Large-scale Experiment for Water and Gas Transport in Cementitious Backfill Materials (Phase 1) 
COLEX I 
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This report was prepared on behalf of Nagra. The viewpoints presented and conclusions reached are those of the author(s) and do not necessarily represent those of Nagra.

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SUMMARY

In the planned Swiss repository for low- and intermediate-level radioactive waste, the voids between the waste containers will be backfilled with a highly permeable mortar (Nagra designation: mortar M1). As well as providing mechanical stability through filling of voids and sorbing radionuclides, the mortar must divert gases formed in the repository as a result of corrosion into the neighbouring host rock. This will prevent damage which could be caused by excess pressure on the repository structures. Water transport, which is coupled to gas transport, is also of interest. The former is responsible for the migration of radionuclides.

Up till now, numerical simulations for a repository situation were carried out using transport parameters determined for small samples in the laboratory. However, the numerical simulations still had to be validated by a large-scale experiment. The investigations presented here should close this gap.

Investigations into gas and water transport were carried out using a column (up to 5.4 m high) filled with backfill mortar. The column has a modular construction and can be sealed at the top end with a material of defined permeability (plug or top plug). The possibility to vary the material of the plug allows the influence of the more impermeable cavern lining or possible gas escape vents in the cavern roof to be investigated. A gas supply is connected to the bottom end and is used to simulate different gas generation rates from the waste.

A total of 5 experiments were carried out in which the gas generation rate, the column height and the permeability of the plug were varied. Before the start of the experiments, the mortar in the column and the plug were saturated with water to approx. 95%. In all the experiments, an increase in pressure with time could be observed. The higher the gas generation rate and the lower the permeability of the plug, the more quickly this occurred. At the beginning, only water flow out of the top of the column was observed. Only after the saturation of the mortar had dropped to approx. 80% could gas be detected flowing out of the column. This gas flow was not continuous but occurred in pulsed intervals.

The experimental investigations were then numerically simulated and the results compared with those from the experiments themselves. This demonstrated that, with the transport parameters used up to now, the experiments could be simulated, sometimes resulting in a very good correlation with the actual tests. Extensions to the numerical model are suggested, which will allow simulation of phenomena not yet implemented in the model.
ZUSAMMENFASSUNG


Es wurden insgesamt 5 Versuche durchgeführt, wobei die Gasgenerationsrate, die Säulenlänge und die Permeabilität des Abschlusspropfens variiert wurden. Der Mörtel in der Säule sowie der Abschlusspropfen wurde vor Versuchsbeginn auf ca. 95 % aufgesättigt. Es zeigte sich in allen Versuchen ein Druckanstieg mit der Zeit, der mit zunehmender Gasgenerationsrate und abnehmender Permeabilität des Abschlusspropfens rascher erfolgte. Zu Versuchsbeginn wurde nur ein Wasserfluss aus der Säule beobachtet. Erst nachdem die Sättigung des Mörtels bis auf ca. 80 % abgesunken war, wurde auch ein Gasfluss aus der Säule festgestellt. Der Gasfluss erfolgte nicht kontinuierlich sondern in Intervallen pulsartig.

Im Anschluss wurden die durchgeführten experimentellen Untersuchungen numerisch simuliert und die numerischen mit den experimentellen Resultaten verglichen. Es zeigte sich, dass mit den bisher verwendeten Transportparametern die Versuche z.T. in guter Übereinstimmung simuliert werden können. Nachfolgend werden Erweiterungen des numerischen Modells vorgeschlagen, um Phänomene simulieren zu können, die bisher im numerischen Modell nicht implementiert sind.
Dans le projet suisse pour le stockage définitif de déchets radioactifs de faible et moyenne activité, les espaces entre les fûts de déchets seront remplis d’un mortier très perméable (« mortier 1 » selon la désignation de la Cédra). Ce mortier doit permettre, en plus de la stabilisation mécanique due au remplissage des vides et de l’adsorption des radioéléments, l’évacuation des gaz dus à la corrosion vers la roche encaissante. Cela peut éviter des dégâts à la construction de la caverne dus à une éventuelle surpression. Le transport d’eau couplé à celui du gaz est également digne d’intérêt, car il est le vecteur de transport des radioéléments.

Les simulations numériques effectuées à ce jour pour le transport de gaz et d’eau dans un dépôt final sont basées sur des paramètres de transport évalués en laboratoire sur de petits échantillons de mortier. Il manque encore une validation des simulations numériques basée sur une étude à plus grande échelle. Les investigations présentées ici sont destinées à combler cette lacune.

La partie expérimentale examine le transport de gaz et d’eau dans une colonne verticale pouvant atteindre 5.4 m de hauteur, remplie de mortier de colmatage. La colonne est modulaire et peut se terminer dans la partie supérieure par un matériau de perméabilité définie (bouchon terminal). Grâce à cette terminaison modulable on peut étudier l’influence de la roche encaissante moins perméable ou celle d’éventuelles ouvertures pour la vidange du gaz au toit de la caverne. L’arrivée de gaz, avec un débit réglable, est connectée à l’extrémité inférieure de la colonne. On peut ainsi simuler la production de gaz des déchets.

On a effectué 5 essais en tout, en variant le débit de production de gaz, la hauteur de la colonne et la perméabilité de la partie terminale. Le mortier dans la colonne et le bouchon terminal ont été saturés d’eau à 95 % avant le début de chaque essai. Tous les essais ont révélé une augmentation de pression au cours du temps, d’autant plus rapide qu’était forte la production de gaz et faible la perméabilité de la partie terminale. Au début des essais, on n’a observé qu’un débit d’eau à la sortie de la colonne. Ce n’est que lorsque la saturation en eau a baissé jusqu’à 80 % environ que l’on a observé un flux de gaz à la sortie de la colonne. Ce flux de gaz n’était pas continu, mais intermittent, en pulsations.

Ensuite, les essais ont été simulés numériquement et les résultats comparés aux observations. Les simulations basées sur les paramètres de transport utilisés jusqu’ici restituent en bonne partie les observations. Pour terminer, on propose des extensions du modèle numérique permettant de simuler des phénomènes qui n’ont pas encore été pris en compte dans celui-ci.
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1 INTRODUCTION

Based on the protection objectives set out in the HSK R-21 Guideline, the release of radionuclides from a sealed repository for radioactive waste, subsequent upon processes and events reasonably expected to happen, shall at no time give rise to individual doses which exceed 0.1 mSv/a. For this reason, underground caverns in rock formations which have a minimum of tectonic activity and low hydraulic permeability are chosen for repositories. Besides the cavern construction and the waste emplacement concept, the neighbouring host rock itself has an important role to play in isolating radioactive substances from the biosphere. In this respect, low-permeability rock formations are the most favourable. Another factor to be considered in the safety analysis for low- and intermediate-level waste disposal is the production of gases due to progressive chemical alteration of the materials emplaced in the repository. Hydrogen is formed as a result of anaerobic corrosion of steel canisters (WIBORGH, HÖGLUND & PERS 1986; MOETSCH, ZUIDEMA & VAN DORP 1994). Furthermore, microbial degradation of organic substances produces methane gas. Aluminium wastes and zinc also produce elemental hydrogen in the cement-bound backfill mortar. Safety analyses assume that, on the long term, 0.5 m³ gas will be produced annually (at STP) for every cubic metre of waste in the repository.

The gases which form in the cavern cause the pressure in the cavern to increase and must therefore be transported from there into the surrounding host rock and, finally, through the rock formation. The resulting pressure differences lead to stresses which must not exceed the tensile strengths of the materials used.

Special, highly permeable, cement-bound mortars for cavern backfilling were thus developed by the Institute for Building Materials at the ETH Zürich (JACOBS, MAYER & WITTMANN 1994; MAYER & WITTMANN 1995). Permeabilities and porosities were examined in the laboratory for small samples around 10 cm in size. Simultaneously, in order to simulate the gas and water transport in a repository, numerical simulations were performed using TOUGH2 and the transport parameters obtained from the laboratory tests (MAYER, JACOBS & WITTMANN 1994, SENGER et al. 1994). However, these numerical simulations still remained to be validated by a large-scale experiment. The investigations presented here are intended to rectify this situation.

The transport of gas and water was investigated in experiments using a column (up to 5.4 m high) filled with backfill mortar. The initial and boundary conditions were varied. The experiments are then simulated numerically and the results compared with those of the experiments themselves.

The report can basically be divided into two sections. In the first section, the experiments are presented, starting with the experiment configuration, followed by a characterisation of the materials investigated and the test procedures. Thereafter, a total of five different experiments are described.
The second section presents the numerical simulations, starting with the model parameters used and including a simulation of the first test. A comparison of the experimental and simulation results allows the model parameters to be refined and any limitations noted. Finally, an extension to the numerical model is proposed.
2 LABORATORY INVESTIGATIONS

2.1 Test configuration

2.1.1 General

In the planned repository for low- and intermediate-level waste, mortar M1 will be used to backfill voids between emplaced containers. If one considers the case of a gas-producing container in the repository, then the gas in mortar M1 would have to cover distances up to 20 metres in order to reach the host rock via the cavern lining or through a gas escape vent. The width of such flow channels for the gas is determined by the spacing between the containers and is typically 20 - 30 cm. The test configuration described in the following is designed to reproduce such transport processes on the same scale.

Ideally, the experiments should be carried out in an instrumented reproduction of the caverns. This is impossible for financial reasons and also because boundary conditions and parameters can be varied only in a restricted way. A modular, one-dimensional flow experiment with distances which are typical for a repository was therefore conceived. The test set-up consists of four column elements, each 1347 mm long and with an inner diameter of 274 mm. These elements are filled with mortar M1 and each is equipped with two water saturation sensors and two pressure sensors. The elements can be screwed together via a flange at each end, producing a maximum column length of 5.4 m. An adjustable gas supply is connected at the bottom end of the assembled column and is used to simulate different gas generation rates. A sample cell is mounted at the top of the column. Materials with known permeability and porosity can be emplaced in this cell and the influence of the gas vent or the host rock on gas and water transport can then be simulated. Outflowing gas and water volumes are measured at the cell exit.

2.1.2 Column elements

A column elements consists of a 1000 mm long weldless steel tube with a welded neck flange on each side. The overall length is 1347 mm (Fig. 1). The inner diameter of the tube is 274 mm and the wall thickness is 25 mm. Each steel tube is equipped with four drillholes for connecting pressure and water saturation sensors. The steel used for the tube is of grade St. 52.0 according to DIN 1629 and has a tensile strength of 500 to 650 N/mm². The flanges are designed according to DIN 2638 for a nominal pressure of 160 bars.
Fig. 1: Structure of a column element

The following factors were considered when selecting the dimensions for the wall thickness of the tube: in conventional sample cells used for investigating transport processes, the cylindrical surface of the sample is sealed using a rubber sleeve or a synthetic resin. This ensures that transport does in fact occur through the sample and not in a gap between the sample edge and the seal. The sealing requirements can be formulated as follows: for a given driving force, the flow through the sample $Q_p$ must be much larger than the flow through the gap $Q_s$:

$$Q_s \ll Q_p$$  \hspace{1cm} (1)

For a cylindrical sample with radius $r$, permeability $k$ and gap width $b$, it follows that:

$$\frac{2\pi rb^3}{12\eta} \frac{dp}{dx} \ll \frac{r^2\pi k}{\eta} \frac{dp}{dx}$$  \hspace{1cm} (2)

and:

$$b^3 \ll 6rk$$  \hspace{1cm} (3)
If it is accepted that 0.1% of the flow will be through the gap, then the maximum allowable gap width for a tube with an inner diameter of 0.275 m filled with mortar M1 ($k \approx 10^{-10} \text{ m}^2$) will be 10 $\mu$m.

A gap occurs between the mortar and the wall of the tube either if the mortar shrinks inside the tube, this can however be ruled out with the more or less water-saturated conditions, or if the tube widens due to a pressure increase in the interior of the tube. It can be seen from the formula for pressure vessels that the gap width $b$ is a function of the pressure difference between the tube interior and the atmosphere, the dimensioning of the tube and the E-Modulus of the steel:

$$\Delta P \frac{r}{d} = E \frac{b}{r}$$

The selected tube wall thickness $d$ of 25 mm and an assumed operating pressure of maximum 10 bars results in an increase of 3.5 $\mu$m in the tube radius. It can thus be ensured without additional sealing measures that side effects will not occur to a measurable extent.

![Diagram](image)

Fig. 2: Assembled sample cell

### 2.1.3 Sample cell and column end

The sample cell mounted at the upper end of the column basically consists of a bottom plate, a steel tube into which the sample is glued and a lid (Fig. 2). The bottom plate consists of a blind flange into which a hole ($\odot = 150 \text{ mm}$) is drilled. The steel tube has an inner diameter of 150 mm, a wall thickness of 10 mm and a height of 65 mm. A groove is turned into both faces of the tube to allow sealing with an O-ring. The lid has a hose fitting and eight screws for fastening the steel tube onto the bottom plate. There is also a sample cell at the bottom end of the column, but this is used only for connecting the gas supply during the experiments or connecting a water reservoir for preconditioning. The steel tube in this cell is empty.
2.1.4 Measuring equipment and data acquisition

2.1.4.1 Pressure

Two types of pressure sensor, which display the measured pressure relative to ambient pressure, were used for the column elements. One has a maximum permissible pressure of 20 bars and the other 100 bars. An absolute pressure gauge was used for measuring atmospheric pressure and a relative pressure gauge (0 - 70 mbar) was used for the gas separator.

2.1.4.2 Gas generation rate

For simulation of a gas source, a commercially available nitrogen bottle with a mass flow regulator was used to establish a constant gas flux. The type used (Brooks) consists of a flowmeter which measures the mass flow using a thermal process, a control valve and an electronic control unit. The gas generation rate can be set between 0 and 3 ml (normal) by applying a constant voltage (0 - 5 V).

2.1.4.3 Water saturation

The structure of the sensor ($\varnothing = 250$ mm, $h = 30$ mm) is similar to that of a plate capacitor, except that the electrodes are made of wire mesh with a mesh size of 1 mm. This is to prevent any interference with the transport processes to be investigated.

To produce the electrodes, circular discs ($\varnothing = 250$ mm) were cut from the wire mesh and an insulated copper wire soldered on at the edge. A 12 mm thick layer of mortar M1 was then introduced into a cylindrical casing and covered with an electrode. A further 6 mm thick mortar layer was then covered with a second electrode and the remainder of the mortar was then placed on top of this. The sensors were removed after one day and stored under water.

In preliminary experiments, the electrical properties of the sensors were checked and a suitable method identified for measuring water saturation. For this purpose, it was necessary to be able to set different levels of water saturation in the probes. In view of the fact that the later experiments were to be carried under fully saturated conditions from the beginning, the probes were initially stored under water. Conditioning the probes at saturation levels below 100% proved problematic. As soon as the probe was removed completely from the water, an uncontrollable volume of water ran out of the probe because of the low capillary forces of the coarse-grained mortar. To avoid this, the probes were placed in the water in such a way that a segment protruded from the water (Fig. 3) and the rest of the probe remained under water. The saturation of the probe is given by the ratio of the wetted volume of the probe to the total volume. In a preliminary experiment, the capillary rise of water in mortar M1 was measured to be approx. 1 cm.
\[ S = \frac{1}{\pi} \left( \pi - \arccos \left( \frac{r-h}{r} \right) \right) + \frac{r-h}{r} \sin \left( \arccos \left( \frac{r-h}{r} \right) \right) \]  

An LCZ-meter (Hewlett Packard) was used to determine capacities, inductivities, complex AC resistance and the phase angle for different frequencies. The capacity could not be measured using this method at the high water saturations since the meshes of the probes were practically short-circuited by the conductive pore solution. A suitable method proved to be to determine the ohmic resistance \( R \) from the complex resistance \( Z \) and the phase angle \( \theta \) for a frequency of 1 kHz.

\[ R = Z \cos \theta \]  

Fig. 3: Arrangement of a water saturation sensor in a water bath for measuring the calibration curve

Fig. 4: Equivalent circuit diagram for water saturation sensor consisting of three resistances
From the equivalent circuit diagram (Fig. 4) for the water saturation sensor, it follows, that the total measured resistance is:

\[ R = R_{\text{cab}} + \frac{R_{\text{dry}} R_{\text{sat}}}{R_{\text{dry}} + R_{\text{sat}}} \]  

(7)

Assuming that the resistance \( R_{\text{sat}} \) is inversely proportional to the saturation of the probe \( R_{\text{sat}} = R_0/S \) and that the resistance \( R_{\text{dry}} \) is inversely proportional to \( (1 - S) \) \( R_{\text{dry}} = R_1/(1 - S) \), it follows together with equation 7 that:

\[ S = \frac{R_1}{R - R_{\text{cab}}} \]

(8)

The two parameters \( R_0 \) and \( R_1 \) were determined by regression. By way of an example, Fig. 5 shows the calibration curve (continuous line) for water saturation sensor 5 together with the resistances determined by regression. As was the case for sensor 5, it was established for all other sensors that \( R_1 \ll R_0 \). Equation 8 can be simplified by considering limiting values.

\[ S = \frac{R_0}{R - R_{\text{cab}}} \]  

(9)

The dashed line in the diagram shows that equation 9 is appropriate, particularly for high water saturations.

The resistance \( R_0 \) is dependent on both the structure of the sensors (surface area and spacing of electrodes) and the composition of the pore solution in mortar M1. When setting up the initial conditions for the experiments, the column was always filled with tap water, which meant that the composition of the pore solution, and thus also \( R_0 \), was always changing. The resistance \( R_0 \) was determined from the minimum of the measured resistances during the experiment by subtracting the cable resistance. It should be noted that the same cables were used for the test phase and for the actual experiment. In order to rule out any influence due to different cable lengths, all sensors were connected to the measuring instrumentation with an 8 m long cable. The resistance of a cable \( R_{\text{cab}} \) is 0.721 \( \Omega \). Since \( R_0 \) and the measured resistances of the water saturation sensors are dependent on temperature, the temperature dependence of the resistances was measured (Tab. 1). The resistance of the cable \( R_{\text{cab}} \) increases with increasing temperature, while the resistance of the pore solution \( R_0 \) decreases.
Fig. 5: Calibration curve for sensor 5

Table 1: Temperature dependence of resistances of water saturation sensors at 20°C

<table>
<thead>
<tr>
<th></th>
<th>$\frac{\Delta R_0}{R_0 \Delta t}$ [%/°K]</th>
</tr>
</thead>
<tbody>
<tr>
<td>cable</td>
<td>0.4 ± 0.1</td>
</tr>
<tr>
<td>pore solution</td>
<td>-1.0 ± 0.3</td>
</tr>
</tbody>
</table>

2.1.4.4 Reference sensors for water saturation

The fact that the calibration curves for the individual sensors give a very good representation of the measurement points at high saturations should not be allowed to obscure the fact that, in the saturation range above 0.5, the sensors react to large changes in water saturation with only relatively small changes in resistance. During the main experiments, which partly continued over a period of several weeks, unavoidable temperature fluctuations and changes in the composition of the pore fluid led to changes in the measured resistance. For this reason, after the first experiment (autoclaved aerated concrete [aac] as vent material, gas generation rate 1.5 ml/min)
the measuring equipment was expanded and comparative measurements were carried out in parallel using 3 further sensors placed in a water-filled bath. The scatter of the measured values in the reference sensors allows an error estimate to be made for the sensors in the column elements.

2.1.4.5 Gas and water flow

A gas separator was used to determine the volumes of gas and water exiting from the upper sample cell. A gas separator basically consists of a sealed container with both an inlet and an outlet on its underside. On the upper side of the container there is a ventilation valve which is opened only for filling the container with water. Gas entering the water-filled separator through the inlet at the bottom rises to the top and collects below the container lid and a volume of water corresponding to the gas volume is then displaced through the outlet. The gas volume can be then registered, for example with a calibrated scale using the height of the water column. The water volume entering the gas separator passes through without influencing the height of the water column. The outflowing water volume, which corresponds to the sum of the inflowing gas and water volumes, is measured using a calibrated container or a balance.

In the present arrangement (Fig. 6), the gas separator is designed for two measuring ranges. Range 1 (valve a closed) can measure a maximum volume of 1000 ml, while range 2 (valve a open) is designed for a maximum volume of 10'000 ml. Instead of a calibrated scale, the gas volume in the separator is measured using a pressure sensor.

The measured pressure difference $\Delta P$ to ambient pressure corresponds to the hydrostatic pressure of the water column with height $h$. The gas pressure in the separator is thus reduced by $\Delta P$ compared with the ambient pressure $P_u$. The gas volume $V_{gn}$ at STP ($T_0 = 273.15K, P_0 = 101325 Pa$) can then be calculated using the state equation for an ideal gas:

$$ V = V_g \frac{(P_u - \Delta P)T_0}{T_u P_0} $$

(10)

where the gas volume $V_g$ is determined from the volume of the container $V_0$, the pressure difference $\Delta P$ and a proportionality constant $\alpha$.

$$ V_g = V_0 - \alpha \Delta P $$

(11)
two-way stop cock

plexiglas cylinder

water

hole

balance

gas and water from output of upper sample cell

Fig. 6: Gas separator and balance for determining gas and water volumes exiting from the column

Table 2: Calibration constants used to determine the gas volume from pressure $\Delta P$

<table>
<thead>
<tr>
<th>measuring range</th>
<th>$V_0$ [ml]</th>
<th>$\alpha$ [ml/Pa]</th>
<th>absolute error [ml]</th>
</tr>
</thead>
<tbody>
<tr>
<td>1</td>
<td>1'300</td>
<td>0.1576</td>
<td>10</td>
</tr>
<tr>
<td>2</td>
<td>10'950</td>
<td>1.5867</td>
<td>50</td>
</tr>
</tbody>
</table>
The gas volume under normal conditions can thus be obtained:

\[ V_{g} = V_{g} \left( \frac{P_{u} - \Delta P}{T_{u}} \right) \left( \frac{V_{0} - \alpha \Delta P}{P_{0}} \right) \]  (12)

The proportionality constant \( \alpha \) was determined for each measurement range. Table 2 summarises the results of calibrating both ranges of the gas separator.

### 2.1.4.6 Temperature

The air temperature and the temperature at the column elements and the gas separator were measured using thermoelements (Cu - Ni). Temperature-induced changes in the resistance of the water saturation sensors could partly be corrected using the measured temperature profile.

### 2.1.4.7 Data acquisition

The following sensors and parameters were covered using a multipoint measuring unit UGR60 (Hottinger):

- pressure sensors at the column
- pressure sensor of the gas separator
- pressure sensor for measuring ambient absolute pressure
- analogue output signal of the mass flowmeter
- temperature sensors

The water saturation sensors are connected to an LCZ meter HP 4276A (Hewlett Packard) using a channel converter HP 3488A of the same company. A PM400 Mettler balance is connected to the PC via a serial interface (RS232). The remainder of the measuring equipment communicates with the PC via an HP-IB (IEEE) interface. A customised Turbo Pascal program is used for periodic measurement and storage of data. Fig. 7 gives an overview of the data acquisition system for a test configuration with two column elements.
legend:

P  pressure sensor column
P* pressure sensor gas separator
Pa  air pressure
S  water saturation sensor
SRef  ""  "" (reference)
T  temperature
Q  Gas flow controller
U=  power supply

Fig. 7:  Data acquisition
2.2 Characterisation of investigated materials

2.2.1 Mortar M1

Earlier investigations by JACOBS, MAYER & WITTMANN (1994) showed that the properties of mortar M1 are dependent on its processing. In this respect, the key parameter is the bulk (apparent) density of the mortar. The porosity and permeability of the mortar can be calculated from this parameter.

The mean fresh mortar density was calculated (Tab. 3) from the mortar mass required for filling the column and the volume of the column. The mortar fillings for the individual column elements have an average density of 1827 kg/m³, with individual values deviating by less than 1%.

For the interpretation and evaluation of the later gas and water transport investigations, it is necessary to know the total porosity of the mortar. A direct measurement of this was not made, however, because complete drying of the entire column at a temperature of 105°C was not feasible and it was essential to avoid potential damage to the structure via the drying process. For each column element, the average porosity was determined from the known linear relationship between bulk density $\rho_b$ and porosity $\varepsilon$ of the M1 mortar mixture (MAYER & WITTMANN 1995).

$$\varepsilon = -0.0316\rho_b + 84.7$$

(13)

The average porosity is thus approx. 27%.

A further possibility for measuring porosity exists if the true density $\rho_s$ and the bulk density $\rho_b$ of the mortar are known:

$$\varepsilon = 1 - \frac{\rho_b}{\rho_s}$$

(14)

whereby the true density of the mortar can be calculated from the true densities of its components. It must however be borne in mind that, following hydration of the cement, 60 cm³ of pores per kg of cement are generated due to chemical shrinking. For $\rho_s$ the following therefore applies:
\[
\rho_s = \frac{\sum \text{mass proportions of components}}{\sum \text{mass proportions of components}} \cdot \frac{\sum \text{true densities of components}}{-\text{volume of voids}}
\]

\[
\rho_s = \frac{1 + 0.4 + 5.33}{3140 + 1000 + 2650 - 0.00006} = 2520 \text{ kg/m}^3 \quad (15)
\]

With the bulk density of 1827 kg/m\(^3\), an average porosity of 27% is obtained. The results obtained using the two independent methods agree within the range of measuring accuracy. Reference was also made to measurements carried out by MAYER & WITTMANN (1995) for determining the permeability of mortar M1. For a bulk density of 1830 kg/m\(^3\) the permeability was found to be \((4\pm3) \times 10^{-10} \text{ m}^2\).

To summarise, it can be said that the transport properties of the mortar fillings in the individual column elements do not differ within the range of measuring accuracy.

Table 3: Composition of mortar M1

<table>
<thead>
<tr>
<th>component</th>
<th>mass proportion</th>
<th>true density (\rho_s) [kg/m(^3)]</th>
<th>addition for 1 batch [kg]</th>
</tr>
</thead>
<tbody>
<tr>
<td>Sulfacem</td>
<td>1.0</td>
<td>3140</td>
<td>12.1</td>
</tr>
<tr>
<td>water</td>
<td>0.4</td>
<td>1000</td>
<td>4.95</td>
</tr>
<tr>
<td>quartz sand 2-3 mm</td>
<td>5.33</td>
<td>2650</td>
<td>65.65</td>
</tr>
</tbody>
</table>
Table 4: Properties of mortar M1 in the column elements

<table>
<thead>
<tr>
<th>element</th>
<th>1</th>
<th>2</th>
<th>3</th>
<th>4</th>
<th>average</th>
</tr>
</thead>
<tbody>
<tr>
<td>(\rho_0) kg/m(^3)</td>
<td>1834±13</td>
<td>1819±13</td>
<td>1836±13</td>
<td>1818±13</td>
<td>1827±13</td>
</tr>
<tr>
<td>(\varepsilon) %</td>
<td>n.b.</td>
<td>n.b.</td>
<td>27±1</td>
<td>n.b.</td>
<td>n.b.</td>
</tr>
<tr>
<td>(\varepsilon^{1)}) %</td>
<td>n.b.</td>
<td>n.b.</td>
<td>27±1</td>
<td>n.b.</td>
<td>n.b.</td>
</tr>
<tr>
<td>(\varepsilon^{2)}) %</td>
<td>n.b.</td>
<td>n.b.</td>
<td>27±1</td>
<td>n.b.</td>
<td>n.b.</td>
</tr>
</tbody>
</table>

1) acc. to MAYER & WITTMANN 1995  
2) derived from true and bulk densities

2.2.2 Autoclaved aerated concrete (aac)

Aac (previously termed gas concrete) is produced industrially using an autoclave. Aluminium powder is added to a mixture of lime, cement, fine sand and water; in an alkaline milieu this releases hydrogen gas and pores are formed. The type of concrete used here has a porosity of 70% and a permeability of \(1.4 \cdot 10^{-15} \text{ m}^2\).

2.2.3 Sandstone

The lime-bonded sandstone used here comes from Ostermundingen (BE) and bears the designation "grey Molasse from Ostermundingen". By controlled manufacturing permeability and porosity can be varied within wide limits, (JACOBS, MAYER & WITTMANN 1992). The sandstone has a total porosity of 18% and a permeability of \(5 \cdot 10^{-16} \text{ m}^2\).

2.3 Test procedure

2.3.1 Filling the column elements with mortar M1 and instrumentation

A column element is erected on a poling board covered with plastic foil. The faces of the flanges are protected with adhesive band.

Mortar M1 was produced in a paddle mixer with a 70 l volume. Two batches were produced for filling a column element with a void volume of 79.4 l. The mortar is infilled in layers approx. 10 cm thick. After each layer is introduced, it is compacted by hand using a tamper. When the level of the side drillholes was reached, the connection for the pressure sensor was screwed in. The cable leading to the centre of the tube was then covered with a layer of mortar a few mm thick. A water saturation sensor was
placed on this and the electric cables led out through the other opening (Fig. 8). The gap remaining between the sensor and the tube wall was also filled with mortar M1. The inner wall of the tube in the vicinity of the water saturation sensors was covered with a layer of epoxy resin in order to prevent any electrical contact of the sensor mesh with the tube wall.

Fig. 8: Filled element with pressure- and water sensors

2.3.2 Assembly of the columns

Columns consisting of 2 and 4 elements were used in the experiments. The first step was to assemble the lower sample cell (without sample) and mount a column element on it. The 16 screws and nuts (size M39) used for this were tightened using a hydraulic torque key. Gas-impermeable flat packing (inner diameter 275, outer diameter 350 mm) was used between the sample cell and column element and between the column elements.

2.3.3 Setting up initial conditions

Before each experiment, the column was first ventilated and the relative pressure transducer of the column elements set to zero. The column was then filled with tap water. The outlet from the upper sample cell (without sample) was then connected to a vacuum pump and the column evacuated for approximately 3 hours. Finally, the lower
column connection was connected to a water reservoir. Because of the underpressure in the column, water was slowly sucked into the column until it was full. Filling of one column element took around 30 minutes. The water inlet was then closed and the empty steel tube of the upper sample cell replaced by one with a glued-in sample (aac or sandstone).

### 2.3.4 Overview of test conditions

Table 5: Properties of the materials used in the column elements

<table>
<thead>
<tr>
<th>material</th>
<th>porosity [%]</th>
<th>permeability [m²]</th>
</tr>
</thead>
<tbody>
<tr>
<td>mortar M1</td>
<td>27</td>
<td>$4 \times 10^{-10}$</td>
</tr>
<tr>
<td>aac</td>
<td>70</td>
<td>$1.4 \times 10^{-15}$</td>
</tr>
<tr>
<td>sandstone</td>
<td>18</td>
<td>$5 \times 10^{-16}$</td>
</tr>
</tbody>
</table>

Table 6: Parameters of the different experiments

<table>
<thead>
<tr>
<th>test</th>
<th>plug material</th>
<th>column height</th>
<th>flow rate</th>
</tr>
</thead>
<tbody>
<tr>
<td></td>
<td>aac</td>
<td>sandstone</td>
<td>2.7 m</td>
</tr>
<tr>
<td>A</td>
<td>x</td>
<td>x</td>
<td>x</td>
</tr>
<tr>
<td>B</td>
<td>x</td>
<td>x</td>
<td>x</td>
</tr>
<tr>
<td>C</td>
<td>x</td>
<td>x</td>
<td>x</td>
</tr>
<tr>
<td>D</td>
<td>x</td>
<td>x</td>
<td>x</td>
</tr>
<tr>
<td>E</td>
<td>x</td>
<td>x</td>
<td>x</td>
</tr>
</tbody>
</table>

### 2.4 Results

#### 2.4.1 Experiment A: 2 column elements, aac, gas generation rate 1.5 ml/min

In the first experiment, aac was used as plug and the gas generation rate was set at 1.5 ml/min. Fig. 9a shows the pressure registered by the four sensors on the column. In contrast to the other experiments, the column was filled in this experiment by
connecting it directly to a water tap. The initial pressure in the column is therefore increased. All sensors show the same type of pressure increase. The pressure differences between the individual sensors is caused by the hydrostatic pressure of the water column. With a vertical spacing of the sensors of 68 cm, the pressure difference is 0.068 bars. Pressure fluctuations were observed in all sensors after approximately 180 hours.

Fig. 9b shows the gas and water fluxes flowing out of the column. At the beginning of the experiment only water flows out of the column. Gas flow is observed after 175 hours, together with a simultaneous decrease in water flow. Gas flow is not continuous but in pulses with an interval of about 10 hours.

Fig. 9c shows the total introduced gas volume (under STP), as well as the water and gas volumes flowing out of the upper sample cell as a function of time.

Fig. 9d shows the saturation of the installed water saturation sensors as a function of time. At the beginning of the experiment, the sensors (with the exception of sensor 1) show a saturation of 90 to 95%. The saturation in all the sensors then increases as the air bubbles enclosed in the sensors become compressed with the increase in pressure. Apart from sensor 1, the water saturation then decreases first in sensor 2, then in sensor 3 and finally in sensor 4. It can be concluded from the time shift in the decrease in water saturation that a gas front is moving slowly through the mortar, pushing the displaced water before it. At the point when the gas exits from the upper sample cell, the sensors show a saturation of approximately 80%.
Fig. 9: Experiment A: 2 column elements, aac, gas generation rate 1.5 ml/min
Fig. 9 cont.: Experiment A: 2 column elements, aac, gas generation rate 1.5 ml/min
Fig. 10: Experiment B: 2 column elements, aac, gas generation rate 3 ml/min (different time scales !)
Fig. 10 cont.: Experiment B: 2 column elements, aac, gas generation rate 3 ml/min (different time scales !)
2.4.2 Experiment B: 2 column elements, aac, gas generation rate 3 ml/min

In the second experiment, the gas generation rate was doubled. Fig. 10a shows the pressure registered by the four sensors on the column. All sensors show the same form of pressure increase but, as expected, the pressure rises more rapidly than in the first experiment. After around 270 hours, a pressure of 2 bars is reached in the lowest sensor; this then decreases slightly and reaches 2 bars again after 430 hours. After this, the pressure drops slightly up to the end of the experiment after 770 hours. As for the first experiment, at the beginning only water flows out of the column (Fig. 10b shows only the first 200 hours of the experiment). After 64 hours, the first gas outflow is observed, with the water flow decreasing at the same time. Because the computer used for data acquisition crashed, measured data are lacking between the 8th and 24th hours after starting measurements. The gas supply was not affected by this problem. Fig. 10c shows the overall balance for the total duration of the experiment. Corresponding with the drop in pressure after around 250 hours (Fig. 10a), the curve for the outflowing gas rises more sharply than that for the inflowing gas.

Fig. 10d shows the saturation of the installed water saturation sensors as a function of time in the first 100 hours of the experiment. At the beginning of the experiment, all the sensors show a saturation of approximately 97%. The water saturation then shows a slight increase as the air bubbles in the sensors are compressed by the rising pressure. Saturation decreases first in sensor 1, then, in order, in sensors 2, 3 and 4. At the time when the gas exits from the upper sample cell after 64 hours, the sensors show a saturation between 78 and 92%.
2.4.3 Experiment C: 2 column elements, sandstone, gas generation rate
3 ml/min

In this experiment, a sandstone sample was installed in the upper cell; this has a lower permeability than the aac. At the beginning of the experiment, the pressure rises sharply in the column (Fig. 11a), reaching a pressure of 2 bars after only 10 hours. After 100 hours, the pressure in the column is 13.5 bars; this drops for a short time, reaching a maximum of 16 bars after 135 hours. After this the pressure drops sharply, reaching a value of 9.5 bars after 340 hours. It then rises again, reaching a value of 12 bars at the end of the experiment, after 880 hours.

In the first 135 hours, neither gas nor water flow out of the sandstone sample is observed (Fig. 11b). At time \( t = 135 \) hours, there is a sudden increase in water flow from the sample, which occurs simultaneously with a drop in pressure (Fig. 1a). The sudden onset of water flow is probably due to damage (microcrack) to the sandstone sample caused by the high pressure. The first gas flow from the sample is observed after 480 hours. Fig. 11c shows the overall volume balance. As for the previous experiments, the water saturation (Fig. 11d) in the sensors drops in time steps, starting with the lowest sensor. At the time of gas outflow, the saturation in the column is between 70 and 85%.
Fig. 11: Experiment C: 2 column elements, sandstone, gas generation rate 3 ml/min
Fig. 11 cont.: Experiment C: 2 column elements, sandstone, gas generation rate 3 ml/min
Fig. 12: Experiment D: 4 column elements, aac, gas generation rate 3 ml/min
Fig. 12 cont.: Experiment D: 4 column elements, aac, gas generation rate 3 ml/min
2.4.4 Experiment D: 4 column elements, aac, gas generation rate 3 ml/min

Prior to this experiment, the existing column was extended by two further elements to a total height of 5.4 m. Fig. 12 shows the overall results. Fig. 12a shows the pressure, which reaches a maximum of 3.1 bars in the uppermost sensor after approx. 1000 hours and then drops up to the end of the experiment (after 2000 hours) to a value of 2.4 bars. Fig. 12b shows the gas and water flows. Gas breakthrough occurs after 275 hours. After around 60 hours, the gas flow drops briefly to zero, at which point the gas bottle was changed. Following this, the gas flow showed slight fluctuations. Fig. 12c shows the overall balance and Fig. 12d shows the saturations of the individual sensors.

The individual values shown are mean values over a timespan of 5 hours. Especially at the beginning of the experiments large fluctuations were observed which are related to changes in the chemical composition of the porewater as the experiment was started immediately after filling the column with tap water. The advancing of the gasfront is marked by strong changes in saturation (marked by arrows).
2.4.5 Experiment E: 4 column elements, sandstone, gas generation rate 3 ml/min

As expected, in the last experiment using a sandstone sample, a stronger increase in pressure was observed. At the end of the experiment, after more than 1000 hours, the pressure had risen to more than 10 bars (Fig. 13a). At the beginning of the experiment, water flow from the column initially rose rapidly to a value of 0.5 ml/min and then slowly decreased (Fig. 13b). The first gas outflow from the column was observed after 950 hours. The water saturation at the end of the experiment was approximately 75 - 90% (Fig. 13d).
Fig. 13: Experiment E: 4 column elements, sandstone, gas generation rate 3 ml/min (different time scales !)
Fig. 13 cont.: Experiment E: 4 column elements, sandstone, gas generation rate 3 ml/min (different time scales !)
2.5 Overview and analysis

A further analysis was performed because of the relative inaccuracy water saturation sensors. Using the ideal gas law, a relationship was established which allows the initial saturation to be determined from the pressure increase rate in the column and the water flow out of the column at the beginning of the experiment. For the column with a gas volume $V_{sg}$ and a pressure $P_s$, connected to a gas source with generation rate $dn/dt$, the following applies:

$$\frac{dn}{dt} = \frac{1}{RT} \left( \frac{dP_s}{dt} V_{sg} - \frac{dV_{sg}}{dt} P_s \right)$$

(16)

If $P_0 V/RT$ is equal to $dn/dt$, with $P_0$ being the normal pressure and $V$ the gas generation rate at STP (here 1 ml/min), then it follows for $V_{sg}$ that:

$$V_{sg} = \frac{P_0 V - \frac{dV_s}{dt} P_s}{\frac{dP_s}{dt}}$$

(17)

Together with the total pore volume of the column $V_s$, it follows for the initial saturation $S_0$ that:

$$S_0 = \frac{V_s - V_{sg}}{V_s}$$

(18)

Experiment A is used here as an example. If the first 2 hours of the experiment are taken for analysis purposes, this results in a gas volume of 2.02 l. Together with total pore volume of the mortar in the two column elements (= 2x0.27x79.4 l), this results in an initial saturation $S_0$ of 95.3%. The water volume $V_{bt}$ which is displaced from the column up till the time when gas flow starts can be used to calculate the average saturation $S_{bt}$ at which gas flow through mortar M1 can occur:

$$S_{bt} = \frac{V_s - V_{sg} - V_{bt}}{V_s}$$

(19)

In the case of experiment A, this Fig. is 81.1%.

The results of the experiments are summarised in Table 7. The initial saturation for the individual experiments varied between 90 and 99%. The resulting saturations at gas
breakthrough are between 77 and 84%. There is no recognisable relationship between initial saturation and saturation at gas breakthrough, nor is there any direct relationship with the gas production rate.

Table 7: Overview of experimental results

<table>
<thead>
<tr>
<th>experiment</th>
<th>A</th>
<th>B</th>
<th>C</th>
<th>D</th>
<th>E</th>
</tr>
</thead>
<tbody>
<tr>
<td>no of elements</td>
<td>2</td>
<td>2</td>
<td>2</td>
<td>4</td>
<td>4</td>
</tr>
<tr>
<td>gasflow q [ml/min]</td>
<td>1.5</td>
<td>3</td>
<td>3</td>
<td>3</td>
<td>3</td>
</tr>
<tr>
<td>plug material</td>
<td>aac</td>
<td>aac</td>
<td>sandstone</td>
<td>aac</td>
<td>sandstone</td>
</tr>
<tr>
<td>saturation S₀ [%]</td>
<td>95.2±0.4</td>
<td>89.7±0.4</td>
<td>98.0±0.1</td>
<td>97.2±0.2</td>
<td>99.1±0.1</td>
</tr>
<tr>
<td>gas break-through</td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
</tr>
<tr>
<td>saturation S₀ [%]</td>
<td>81.0±0.8</td>
<td>80.9±0.8</td>
<td>84.1±0.6</td>
<td>77.4±0.9</td>
<td>80.1±0.7</td>
</tr>
<tr>
<td>time t₀ [h]</td>
<td>170</td>
<td>64</td>
<td>480</td>
<td>275</td>
<td>950</td>
</tr>
<tr>
<td>pressure P₀ [bar]</td>
<td>0.8±0.1</td>
<td>1.1±0.1</td>
<td>10.5±0.1</td>
<td>1.4±0.1</td>
<td>9.5±0.1</td>
</tr>
<tr>
<td>max. pressure</td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
</tr>
<tr>
<td>saturation at Pₘₐₓ [%]</td>
<td>n.b.</td>
<td>74.1±1.0</td>
<td>n.b.</td>
<td>72.4±1.0</td>
<td>n.b.</td>
</tr>
<tr>
<td>max. pressure Pₘₐₓ [bar]</td>
<td>n.b.</td>
<td>2.0±0.1</td>
<td>n.b.</td>
<td>3.6±0.1</td>
<td>n.b.</td>
</tr>
<tr>
<td>time t(Pₘₐₓ) [h]</td>
<td>n.b.</td>
<td>260</td>
<td>n.b.</td>
<td>1200</td>
<td>n.b.</td>
</tr>
<tr>
<td>end of experiment</td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
</tr>
<tr>
<td>saturation Sₘ₀ [%]</td>
<td>80.4±0.9</td>
<td>76.6±1.0</td>
<td>82.9±0.8</td>
<td>70.3±1.0</td>
<td>80.0±0.7</td>
</tr>
<tr>
<td>time tₘ₀ [h]</td>
<td>210</td>
<td>770</td>
<td>740</td>
<td>2070</td>
<td>1020</td>
</tr>
</tbody>
</table>

1) pressure in uppermost sensor  2) after 1600 h no more water outflow out of column

There is potentially a link with the material used in the sample cell and the number of column elements (i.e. the length of the column). If the results for two column elements are compared with those for four column elements, then for the experiment with sandstone gas breakthrough always occurs with the highest water saturation. Using sandstone also produces the highest pressures in the column.
3 NUMERICAL MODELLING

The following section begins by presenting the model parameters for the simulations with TOUGH2 (PRUESS 1991). This is followed by a description of the simulation of experiment A and a comparison with the experimental results. The reasons for any discrepancies are discussed and the model parameters are adjusted as far as possible.

3.1 Model Parameters

3.1.1 Mesh with Finite Volume

The flow geometry in the column experiments was realised using a one-dimensional mesh of finite volume. The mortar M1 in a column element is discretised into 50 elements, with each element representing a 2.7 cm thick layer. The hole in the blind flange of the sample lid - between the mortar M1 and the top plug - is realised as one element. The 6 cm long top plug is modelled with 10 elements. The column outlet is modelled with one element whose volume is selected to be very large (10^{50} m^3) in order to guarantee the boundary condition of constant pressure.

3.1.2 Transport Parameters, Boundary and Initial Conditions

In addition to the permeabilities and porosities in section 2.2, the relative permeabilities and capillary pressure as a function of degree of saturation are additional material parameters which are necessary for simulating two-phase transport processes.
Table 8: Transport parameters

<table>
<thead>
<tr>
<th>experiment</th>
<th>mortar M1</th>
<th>aac</th>
<th>sandstone</th>
</tr>
</thead>
<tbody>
<tr>
<td>e</td>
<td>[\text{-}]</td>
<td>0.27</td>
<td>0.7</td>
</tr>
<tr>
<td>k</td>
<td>[m$^2$]</td>
<td>4.10$^{-10}$</td>
<td>8.10$^{-16}$</td>
</tr>
<tr>
<td>model rel. permeability</td>
<td>linear</td>
<td>Corey</td>
<td>Corey</td>
</tr>
<tr>
<td>$S_{rl}$</td>
<td>[\text{-}]</td>
<td>0.3</td>
<td>0.3</td>
</tr>
<tr>
<td>$S_{rg}$</td>
<td>[\text{-}]</td>
<td>0.3</td>
<td>0.2</td>
</tr>
<tr>
<td>model capillary pressure</td>
<td>Narasimhan</td>
<td>Narasimhan</td>
<td>Narasimhan</td>
</tr>
<tr>
<td>$P_0$</td>
<td>[\text{bar}]</td>
<td>0.005</td>
<td>0.054</td>
</tr>
<tr>
<td>$S_{00}$</td>
<td>[\text{-}]</td>
<td>0.3</td>
<td>0.45</td>
</tr>
<tr>
<td>$1/\eta$</td>
<td>[\text{-}]</td>
<td>2.0</td>
<td>2.2</td>
</tr>
</tbody>
</table>

3.1.2.1 Relative Permeabilities

The relative permeability of the gaseous and liquid phases of the mortar M1, $k_{rg}$ and $k_{rl}$ respectively, can be described using the following linear function (MAYER, JACOBS & WITTLMANN 1994):

\[
k_{rl} = \begin{cases} 
\frac{S_g - S_{rg}}{1 - S_{rg}} : S_g > S_{rg} \\
0 : S_g \leq S_{rg}
\end{cases}
\]

\[
k_{rl} = \begin{cases} 
\frac{S_l - S_{rl}}{1 - S_{rl}} : S_l > S_{rl} \\
0 : S_l \leq S_{rl}
\end{cases}
\]

Neither aac nor sandstone have been investigated in detail. They are described using the conventional COREY model (COREY 1954):

\[
S^* = \frac{S_l - S_{rl}}{1 - S_{rl}}
\]

\[
k_{rl} = (S^*)^4
\]

\[
k_{rg} = (1-S^*)^2 (1-(S^*)^2)
\]
3.1.2.2 Capillary Pressure

The capillary pressure curve for mortar M1 (JACOBS, MAYER & WITTMANN 1994) was parameterised with a model from Narasimhan (PRUESS 1987):

\[
P_{\text{cap}} = \begin{cases} 
- P_0 \left( \frac{1 - S_i}{S_i - S_{i0}} \right)^{(1/\eta)} : S_i < 1 \\
0 : S_i = 1
\end{cases}
\]

The capillary pressure curves for aac and sandstone were derived from the pore size distribution - determined by mercury porosimetry - and were also parameterised with the Narasimhan model. The parameters used for two-phase transport are shown in Table 6.

Table 9: Initial and boundary conditions for the tests

<table>
<thead>
<tr>
<th>Experiment .</th>
<th>A</th>
<th>B</th>
<th>C</th>
<th>D</th>
<th>E</th>
</tr>
</thead>
<tbody>
<tr>
<td>Gas prod. rate</td>
<td>q [ml/min]</td>
<td>1.5</td>
<td>3.0</td>
<td>3.0</td>
<td>3.0</td>
</tr>
<tr>
<td>Mortar M1</td>
<td>$S_0$ [-]</td>
<td>0.95</td>
<td>0.90</td>
<td>0.98</td>
<td>0.97</td>
</tr>
<tr>
<td></td>
<td>$P_0$ [bar]</td>
<td>1.20</td>
<td>0.96</td>
<td>0.96</td>
<td>0.96</td>
</tr>
<tr>
<td>Plug</td>
<td>$S_0$ [-]</td>
<td></td>
<td>0.95</td>
<td></td>
<td></td>
</tr>
<tr>
<td></td>
<td>$P_0$ [bar]</td>
<td></td>
<td>0.96</td>
<td></td>
<td></td>
</tr>
<tr>
<td>Edge</td>
<td>$S$ [-]</td>
<td></td>
<td>0.0</td>
<td></td>
<td></td>
</tr>
<tr>
<td></td>
<td>$P$ [bar]</td>
<td></td>
<td>0.96</td>
<td></td>
<td></td>
</tr>
</tbody>
</table>

3.1.2.3 Initial and Boundary Conditions

The initial and boundary conditions are the same as those for the previous experiments and can be found in Table 7. By way of simplification, isothermal boundary conditions are assumed ($T = 293\, ^\circ\text{K}$). The maximum $\pm 3^\circ\text{K}$ fluctuations in ambient air temperature observed in the experiment throughout the day can be ignored.
3.2 Numerical Simulation of Experiment A

The left-hand graphs in Fig. 14 show the results of simulating experiment A and the right-hand graphs show the experimental results. Discrepancies between the two are discussed in the next section.

A pressure increase with time in the column was observed (Fig. 14a). The pressure difference between the individual sensors remains constant. Following gas breakthrough, the pressure in the column drops. Water flow out of the column increases with time until, after 225 hours, it drops dramatically and, after 250 hours, practically ceases (Fig. 14b). At the same time, gas starts to flow from the column. The simulated water saturation of the sensors can be seen in Fig. 14c. A slight increase in saturation is observed at the beginning of the test. After approx. 30 hours the saturation in sensor 1 drops to 0.7, followed by sensors 2 and 3 and, finally, after 200 hours, sensor 4.

3.3 Comparison between Simulation and Experiment

The simulated pressure increase also takes the same form, although it increases more slowly than in the experiment where higher pressures are achieved. In particular, the sharp fall in pressure after the gas breakthrough seen in the simulation is not observed in the experiment, where the pressure continued to increase still further.
Fig. 14: Test A: simulation (base case)
Fig. 14 cont.: Test A: experiment
In the simulation, the flow of water out of the column before gas breakthrough is of the same order of magnitude as that seen in the experiment. However, in the experiment, a maximum is reached after approx. 25 hours while, in the simulation, the water flow continues to increase for 225 hours. Whereas gas breakthrough occurs after around 240 hours in the simulation, in the experiment it occurs after only 170 hours. The pulsed gas breakthroughs seen in the experiment are not present in the simulation.

In the simulation, the water saturation in the individual sensors drops to a value of 0.7 while, in the experiment, a value of 0.8 is reached.

3.4 Adjustment of Transport Parameters

3.4.1 Introduction

The comparison between experiment and simulation in the previous section shows that the most important features can be reproduced correctly in qualitative terms; in quantitative terms, however, there are still considerable differences. The reasons for these discrepancies can be found almost exclusively in the numerical modelling. Assuming that the initial and boundary conditions for the modelling are correct, and disregarding straightforward numerical errors, reasons for the discrepancies could be:

- **The transport parameters (permeability, relative permeability, capillary pressure) are incorrect.** If this is the case, then adjusting these parameters will reduce the discrepancy between simulation and experiment. As well as "trial and error", inverse analysis can be carried out, for example with ITOUGH (FINSTERLE 1993). Due to the large number of possible transport parameters, both cases require that a careful pre-selection of relevant parameters be made.

- **The transport model is incorrect.** In this case, modifying the transport parameters will not improve the correlation between simulation and experiment. There is likely to be a danger that "unreasonable" adjustment of the transport parameters will result in physical effects being overlooked.

The connection between the simulation results and the transport parameters should be examined first. This analysis will provide possibilities for adjusting transport parameters and will draw attention to experimental effects which could not be simulated by the current TOUGH2 model. Further procedure includes adjusting some of the transport parameters and comparing the simulation results again with the experiment.
3.4.2 In What Way are the Simulation Results Dependent on the Transport Parameters?

3.4.2.1 Water Saturation in Mortar M1 as a Function of Time

In the simulations under discussion here, the residual gas saturation $S_{rg}$ of mortar M1 is of significance. The value of 0.3 assumed here means that no mobile gas phase exists with a water saturation of more than 0.7 ($\equiv 1 - 0.3$). This stipulation is met at the beginning of each experiment. The gas will first accumulate in a mortar layer directly at the gas inlet. As soon as the water saturation here reaches 0.7 then the gas phase in this layer mobilises and can enter the next mortar layer, where it will again accumulate until the formation of a mobile gas phase, etc. The gas injected into the bottom of the column cannot rise up in the form of small bubbles through the column, which is filled with more or less water-saturated mortar M1. Instead, it forms a front with a water saturation of $S_t = 1 - S_{rg}$ (0.7 here), which migrates upwards through the mortar. The speed with which the front progresses depends on how quickly the water can be expelled from the column.

3.4.2.2 Water Saturation in the Mortar M1 at Gas Breakthrough

In the simulations, the water saturation $S_{bt}$ of the mortar M1 at gas breakthrough depends only on the residual gas saturation $S_{rg}$:

$$S_{bt} = 1 - S_{rg}$$

This is independent of column height and of the end-plug used.

3.4.2.3 Maximum Water Flow from the Column

As soon as the gas breakthrough point is reached, no more liquid is expelled from the mortar. With the model assumptions made, a further desaturation of the mortar M1 could only occur as a result of removal of water vapour in the saturated gas phase. Since air contains a maximum of 17 g of water per m$^3$ at 20°C, with the gas flows used here of maximum 3 ml/min it is not possible to simulate a measurable desaturation.

3.4.2.4 Flow and Pressure

The driving force for the flow of gas and water through the column is the gas pressure generated within the column. The gas and water flow experience a pressure drop in
both the mortar M1 and in the end-plug. Since the permeability of the mortar M1 is several orders of magnitude higher than that of the end-plug, the pressure drop in the mortar due to the flow of water and gas can be ignored. Besides the gas generation rate, only the permeability of the end-plug can determine the development of pressure within the column.

Before gas breakthrough initially only water is expelled from the column through the end-plug. Flow and pressure are determined by the hydraulic permeability of the saturated end-plug (absolute permeability).

The simulated increase in pressure within the column must also result in a corresponding increase in the flow of water with time out of the column. A drop in water flow with increasing pressure, as observed in the experiment, can only be explained by a progressive decrease in the permeability of the end-plug. Such effects have already been observed in permeability measurements on cement-bound materials (JACOBS, MAYER & WITTMANN 1992). Suspended particles which block up the pores could be one possible explanation for this. In principle, a simulation of these effects is possible, but has not yet been included in the currently available version of TOUGH2.

In the simulation, the water flow drops sharply immediately before gas breakthrough. At this point no more water is expelled from the mortar M1 and the gas front now moves through the end-plug. Here, too, the water must be expelled before the gas phase can become mobile.

After the gas breakthrough only gas flows out of the column in the simulation. The pressure development within the column is now determined from the effective gas permeability of the end-plug which, in turn, depends upon the degree of water saturation of the plug.

3.4.3 Simulations with Adjusted Transport Parameters

This section presents simulations in which specific individual transport parameters have been adjusted to achieve better agreement with the experiment. It was not necessary to perform intensive inverse modelling since the fundamental dependence of the results on the transport parameters had already been noted.

3.4.3.1 Experiment A

Firstly, the residual gas saturation $S_{rg}$ of mortar M1 and the permeability of the end-plug were used in an attempt to achieve a better correlation before gas breakthrough.

The first step was to reduce the residual gas saturation of the mortar M1 to a value of $S_{rg} = 0.2$. The corresponding simulation can be seen in Fig. 15. The saturation in the individual sensors (Fig. 15c) now drops to a value of $S = 0.8$, which agrees well with
the experiment. Since the gas front now moves more quickly through the mortar M1, the gas breakthrough in the simulation occurs sooner, after approx. 150 hours.

A further simulation, in which the permeability of the end-plug is also reduced ($k = 5 \cdot 10^{-16}$), can be seen in Fig. 16. This results in higher pressure being reached in the column (Fig. 16a), which corresponds better with the experiment. Water flow before gas breakthrough is also decreased (Fig. 16b), which causes the gas breakthrough to occur after around 170 hours, similarly to the experiment.

In order to achieve a better correlation after gas breakthrough, the effective gas permeability of the end-plug has to be reduced. This can be accomplished by increasing the capillary pressure, as a result of which the end-plug will desaturate less. Fig. 17 shows the results of a simulation with increased capillary pressure of the aac ($P_0 = 2$ bar, $1/\eta = 1$). Although this gives a better agreement with the experiment, it must be pointed out here that this modified value for capillary pressure is far removed from the base case ($P_0 = 0.054$ bar). A further possibility for achieving a better agreement is described in section 3.4.5. Water flow after gas breakthrough can also be explained with this approach.
Fig. 15: Test A: simulation, mortar M1: $S_{tg} = 0.2$
Experiment

Fig. 15 cont.: Test A: experiment
Fig. 16: Test A: simulation, mortar M1: $S_{\text{rg}} = 0.2$, aac: $k = 5.0 \cdot 10^{-16} \text{ m}^2$
Fig. 16 cont.: Test A: experiment
3.4.3.2 Experiment B

For simulating experiment B, the adjusted model parameters from the previous section are used together with the corresponding boundary conditions for experiment B. The gas breakthrough now occurs after around 100 hours, somewhat later than in the experiment (Fig. 18b). In the simulation, the pressure continues to increase after gas breakthrough, reaching a value of 2 bars which agrees well with the experiment.

3.4.3.3 Experiment C

After 135 hours of this test (p. 19), excessively high pressure damaged the sandstone. As a result of this, the flow of water out of the column increased erratically. Splitting the entire simulation into two parts would allow TOUGH2 to accommodate such effects. In the first part, up to 135 hours, the sandstone would have a low permeability and, in the second, a higher. However, since neither the time of failure nor the increase in permeability of sandstone a priori is known (or can be simulated), no simulation was carried out. Reference should be made here to test E, carried out with the damaged sandstone plug from this experiment.

3.4.3.4 Test E

Finally, test E was modelled with 4 column elements and sandstone as the end-plug. The modified transport parameters for mortar M1 are used with a residual gas saturation of 0.2. The permeability of the sandstone is taken from test C: after 200 hours of this test (the sandstone is already damaged, see experiment C), approx. 0.3 ml/min of water flows from the column. For a pressure gradient of 11 bar/6 cm, this corresponds to a permeability of $k = 1.6 \cdot 10^{-17}$ m$^2$.

The simulation and corresponding experiment can be seen in Fig. 19.

3.4.4 Which Effects Cannot be Simulated?

- **Time-dependent permeability**
  There are no plans to model time-dependent permeability, as has been mentioned already.

- **Water saturation of mortar M1 at gas breakthrough**
  The different water saturations at gas breakthrough observed in the experiments cannot be simulated.
• *Oscillating pressure, gas and water flow after gas breakthrough*

Neither the water flow after gas breakthrough, nor the observed oscillations in pressure and the resulting pulsed escape of gas out of the column can be simulated by this model. These effects can possibly be explained by the inevitable presence of isolated gas bubbles due to the incomplete initial saturation $S_0$ of the column in all experiments.
Fig. 17: Test A: simulation, mortar M1: $S_{tg} = 0.2$, aac: $k = 5.0 \cdot 10^{-16}$ m$^2$, $P_0 = 3$ bar, $1/\eta = 1$
Fig. 17 cont.: Test A: experiment
Fig. 18: Test B: simulation, mortar M1: $S_{\text{rg}} = 0.2$, aac: $k = 5.0 \cdot 10^{-16}$ m$^2$, $P_0 = 3$ bar, $1/\eta = 1$
Fig. 18 cont.: Test B: experiment
Fig. 19: Test E: simulation, mortar M1: Srg = 0.2, sandstone: k = 1.6 \times 10^{-17} \text{ m}^2, P0 = 3 \text{ bar}, 1/\eta = 1
Fig. 19 cont.: Test E: experiment
Fig. 20: Isolated gas bubble; (a) contracting due to increasing pressure and (b) expanding due to falling pressure.
3.4.5 Proposal for an Extension to the Model

In the transport model used up till now, it was assumed that the entire gas phase mobilised at a gas saturation above $S_{gr}$. This assumption is indeed correct if there is complete water saturation at the beginning of the experiment. The gas penetrating into the mortar then forms a continuous transport channel.

In our case, a few gas bubbles are already distributed throughout the mortar M1 at the beginning of the experiment. The gas entering the mortar M1 forms a gas pathway which does not necessarily meet up with an existing gas bubble. As well as the gas transport pathways, isolated "passive" gas bubbles can thus exist (Fig. 20).

Such isolated gas bubbles act as air cushions which contract as the external pressure increases. Conversely, if the external pressure falls, they will expand. If the pressure in the mortar falls - as will be the case e.g. after gas breakthrough - the expansion of these enclosed gas bubbles will cause liquid to be forced back into the transport channel, causing the flow of water to resume. However, due to the higher viscosity of the liquid, more energy will be required. The pressure in the system will thus increase again and, as the gas bubbles compress again, the liquid will once more be expelled from the transport channel.

The whole process now starts from the beginning again since the gas phase is mobile once more and the pressure falls. This results in an alternation between gas and water flow and corresponding pressure oscillation.

Implementation of the model taking into account these enclosed air bubbles could not be carried out as part of this work.
4 REFERENCES


