The predicted and actual wetting rate of the buffer in repositories for high-level radioactive waste

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Abstract

Underground test sites for the disposal of high-level radioactive waste (HLW) are commonly equipped with instrumentation for documentation of the evolution of engineered barriers and of changes in the physical state of the host rock. The rate of hydration of the clay buffer surrounding waste canisters determines the physical interaction with the rock and waste canisters and is recorded by use of RH meters and by measuring the swelling pressure exerted on cells installed in the clay. They are connected to cables and tubings extending to the recording units placed in room atmosphere. If they pass from the rock through the buffer groundwater under pressure can migrate through or along bunches of cables and tubings and reach the points of measurement earlier than in clay without instrumentation. The inconvenience is avoided by tight embedment of individual cables and tubings and placement of the pressure gauges such that they do not pass through the clay. Incorrect rates of hydration has led to the inadequate conclusion that water flows in unsaturated parts of the buffer according Darcy's law while it is in fact migrating by coupled diffusion processes.
Keywords: buffer, cables, instrumentation, pressure sensors, thermocouples, tubings.

1 Introduction

The role of the buffer clay in a repository for highly radioactive waste is to provide the waste containers (canisters) with an embedment that is tight and has sufficient bearing capacity and at the same time sufficient ductile for minimizing seismically generated stresses in the containers. The thermal conductivity must

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be sufficiently high to keep the temperature at the buffer/container contact below 100°C. The period of hydration by uptake of water from the surrounding rock shall be as short as possible for minimizing salt accumulation in the buffer, a process that can have an impact on the corrosion rate of the containers. Documentation of the hydration rate has therefore been an important task in the underground laboratories prepared by companies responsible for safe disposal of HLW. We will consider experiments performed by the Swedish Nuclear fuel and Waste Management (SKB) in this paper for identifying difficulties and possibilities in this respect.

2 Evolution of buffer in repository-like environment

2.1 Processes

When the canisters with spent reactor fuel have been placed in the deposition holes with a nominal diameter of 1.9 m and depth of about 8 m the heat production of the decaying highly radioactive waste (HLW) starts to affect the buffer, which has the form of tightly stacked highly compacted blocks of smectite-rich clay ("bentonite") surrounded by clay pellets according to the Swedish concept KBS-3V [1]. The blocks and pellets consist of expandable clay with a content of smectite of at least 70% by weight. The exposure to heat is paralleled by uptake of water that migrates from the rock to the pellet fill and further to the dense blocks (Figure 1).

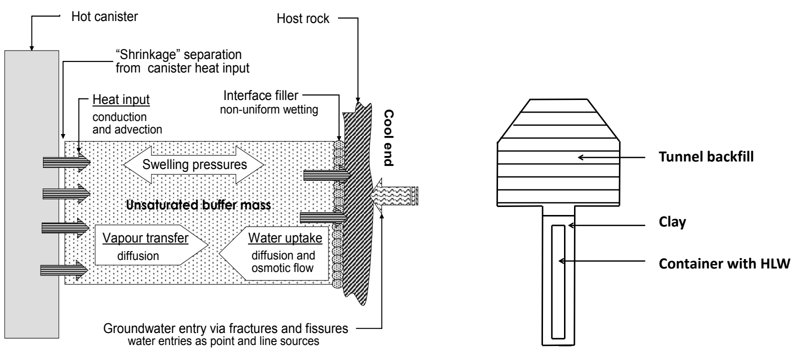

Figure 1: Schematic showing basic elements of the canister-buffer interaction [2].

The main processes in the early phases of buffer hydration under a thermal gradient can be summarized as [2]:

- migration of porewater by gradients in hydration potential,
- flow of water from colder to warmer regions because of differences in heat contents,

- flow of water under pressure,
- reverse migration of porewater by electric potentials set up by porewater flow.

Porewater redistribution in the early phase of buffer evolution is controlled by migration along particle surfaces under tension gradients, and by vapour migration, which are both diffusive processes. In a deposition hole vapourization of porewater takes place in the hottest part of the buffer and brings it towards the rock and to the overlying backfill that is not saturated with water. In the successively narrowing "dry" zone close to the hot canister surface dissolution of clay and accessory minerals takes place and also precipitation of salts like sodium chloride and calcium sulphate.

Swelling of the buffer upon water entry and uptake produces compressive forces throughout the wetted portion, which will be transmitted to the unwetted part resulting in progressive compression of it.

2.2 Laboratory hydration experiments

Laboratory experiments with uniaxial wetting of clay with 70–80% montmorillonite (MX-80) mainly saturated with Na and partly with Ca and Mg and with the dry density 1270 kg/m^3 (1800 kg/m^3 at water saturation) have given information on the rate of hydration from an initial degree of water saturation of about 20% up to about 100% as a function of the composition of the solution, the water pressure and temperature gradient [3]. The major conclusions were that 3.5% $CaCl_2$ solution gives much quicker hydration than electrolyte-poor water, that pressurizing the solution to 500 kPa also gives rapid hydration, that enrichment of chlorides and sulphates at the heated end is quick and strong, and that a high temperature gradient (14^{o}C/cm) with 30^{o}C at the wetted boundary and 100^{o}C at the opposite end significantly retards the wetting.

2.3 Examples of recorded hydration on a larger scale

2.3.1 Czech mock-up test

Mock-up tests representing SKB's concept KBS-3V down-scaled to 1:3 have been performed by Pacovsky *et al.* at the Prague Technical University using the so-called RMN buffer that consists of 75% Ca montmorillonite, 10% finely ground quartz and 5% graphite powder [4]. The blocks had a radial thickness of 18 cm and a dry density of 1800 kg/m^3, the initial water content being 7%. They were surrounded by 50 mm loosely filled RMN granules, which were in contact with a filter that was kept saturated with brackish granite water under 60 kPa pressure. The initial dry density of the pellet fill was 900 kg/m^3. The clay/heater system was initially closed but water was let in after a few weeks when a state of equilibrium was reached. The height and diameter of the confining container were 2.23 m and 304 mm, respectively. The surface temperature was kept at about 95^{o}C at mid-height heater in the 2.5 years long experiment.

At termination the density, temperature, swelling pressure and hydraulic conductivity of bored-out samples were as shown by Table 1. The evolution of the buffer clay was followed by taking samples 2–3 times per year for determining geotechnical data. M1 represents the outer pellet fill, M2 moderately heated dense block, M3 significantly heated dense block, and M3 strongly heated dense block adjacent to the heater [4].

Table 1: Material data of the Czech buffer clay after 2.5 years.

Sample	Distance from heater, cm	T, °C	ρ_{sat}, kg/m^3	p_s, kPa	K, m/s
M1	16-18	45-47	1800	430	1.6E-11
M2	12-14	54-56	1945	650	1.9E-11
M3	6-8	67-69	1910	355	2.3E-10
M4	0-2	85-90	1925	310	2.1E-10

The differences in density of the fully hydrated samples were caused by compression of the drier parts under the early generated swelling pressure of the wet clay [5]. They partly remained in the final state of complete water saturation but in a very long time perspective some homogenization may occur by creep processes.

The hydration rate of the buffer illustrated in Figure 2 was evaluated by determining the water content of samples taken in different positions during the 2.5 year test. The curve is very similar to the predicted hydration assuming diffusive water migration for the coefficient D = 2E-9 m^2/s.

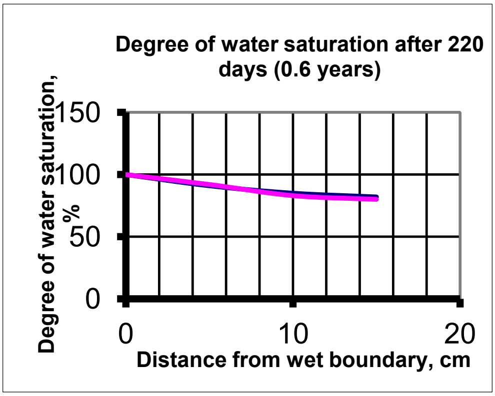

Figure 2: Degree of water saturation at different distances from the contact between the powder fill and the buffer blocks ("wet boundary"). The curve giving the lowest values at 10–15 cm from this contact represents the actual values while the other represents diffusive water migration for D = 2E-9 m^2/s.

Carefully calibrated ROTRONIC moisture sensors gave scattered data in the first 20 months (May 24, 2002, to October 6, 2003), presumably because of water leaking from the granular fill along the cables (Figure 3), [4]. After about 1.5 years the wetting had become uniform.

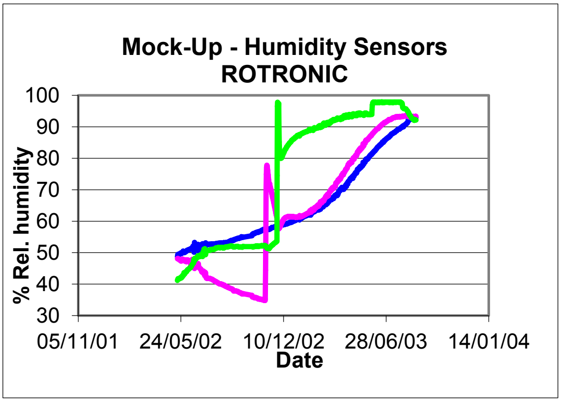

Figure 3: Recorded hydration of the buffer in the Czech Mock-up experiment. Upper: top of clay column; Lower: base of heater; Central: lowest part of clay column [4].

An indirect way of determining the rate of hydration is to measure the swelling pressure exerted by the hydrating clay on pressure cells or sensors installed in the clay buffer. The recorded value is the total pressure, i.e. the sum of the porewater pressure and the true (effective) swelling pressure. It was measured in the Czech mock-up experiment and showed a successively but not uniformly increased total pressure when the wetting started (Figure 4). The variations in pressure were due to breakage and displacements of the buffer blocks and self-sealing of created fractures.

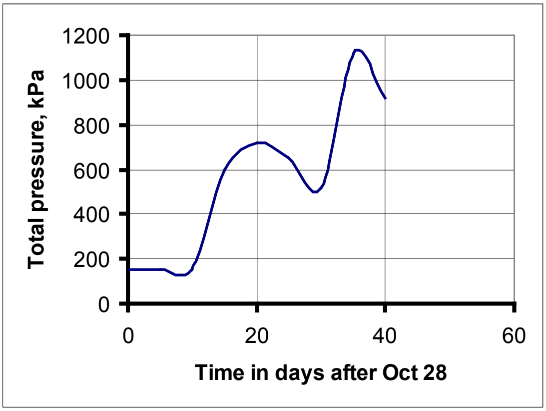

Figure 4: Example of recorded total pressure in the Czech mock-up experiment after letting water into the filter under 60 kPa pressure on October 1 (2003).

2.3.2 Field tests – the Stripa Project

The international NEA Stripa Project comprised the B(uffer) M(ass) T(est) in crystalline rock at about 360 m depth, [6] managed by SKB. Here, six holes with 760 mm diameter and about 3000 mm depth were bored using coring technique in a blasted drift that was backfilled with a mixture of moraine-graded ballast (aggregate) with 10–20% MX-80 bentonite. A 9 mm space was left open between the clay blocks and the rock. The dry density of the buffer blocks was 1710–1780 kg/m^3 and the water content 9–10%. The ultimate average dry density was 1510 kg/m^3 (1950 kg/m^3 at water saturation). The columns of blocks was richly instrumented for recording the thermal and pressure evolutions of the buffer that surrounded electrical heaters of teflon-coated aluminum with 380 mm diameter powered to 600–1300 W. Great care was taken to effectively fill and compact the grooves for the tubings of the Gloetzl pressure cells for avoiding water leakage along them. A significant drop in temperature close to the heater indicated desiccation and fracturing while the outer 50 mm annulus had become largely water saturated. The inflow of water varied among the holes of which No 1, 2 and 5 provided enough water to the buffer for maturing without limiting the rate of hydration.

Pressure cells at the contact between buffer and rock gave the evolution of the swelling pressure and thereby an indication of the rate of hydration in Figure 5.

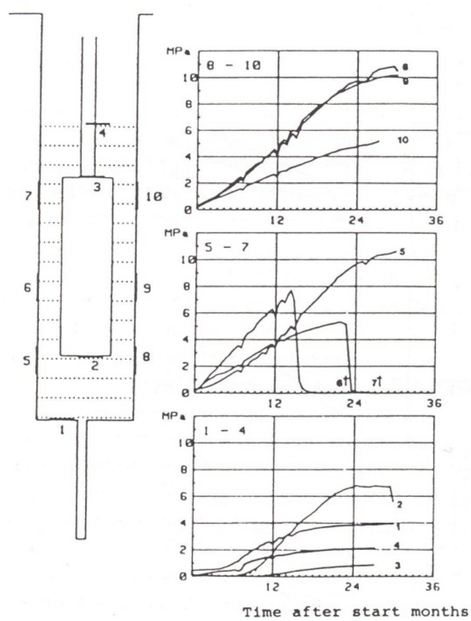

Figure 5: Evolution of swelling pressure of the buffer in the wettest deposition hole (No 2) with 600 W simulated HLW canister at Stripa. Gloetzl cells 6 and 7 failed after more than one year [6].

2.3.3 Field tests – the Äspö prototype

SKB's underground laboratory at Äspö is a successor of the Stripa test site and has a similar outline. Six deposition holes with about 8 m depth and 1.8 m diameter contained heaters powered at 1800 W contained columns of dense clay blocks instrumented so for investigating the early phases of maturation of MX-80 buffer clay. Backfilling of the tunnel was made with a mixture of 30% MX-80 bentonite and 70% crushed rock [1].

The holes were bored by TBM-type technique, which gave an excavation-disturbed zone of 5–10 mm depth. This zone has a considerably higher hydraulic conductivity than the virgin rock crystal matrix, and hence distributes water entering from discrete fractures over the periphery of the holes [6]. Ring-shaped buffer blocks with an average dry density of 1750 kg/m^3 were stacked in the holes and HLW canisters placed in them leaving a gap between rock and blocks of 50 mm and a 10 mm gap between the blocks and the canisters. The outer gap was loosely filled with MX-80 granules, implying that the net ultimate density at saturation with brackish water will be 1950 kg/m3. Inflow tests in the six holes gave 1 to 100 l/day. The hydraulic conductivity of the rock around the holes ranged between E-12 and E-10 m/s with a geometric mean of E-11 m/s [1].

The evolution of temperature and wetting at midheight of the wettest hole is shown in Figure 6 in which the relative humidity (RH) can be taken as the degree of water saturation. The temperature still rose after more than 4 years and reached about 72^{o}C in the clay adjacent to the canister surface and around 60^{o}C at the rock after about 2 years. These temperatures were much lower than planned.

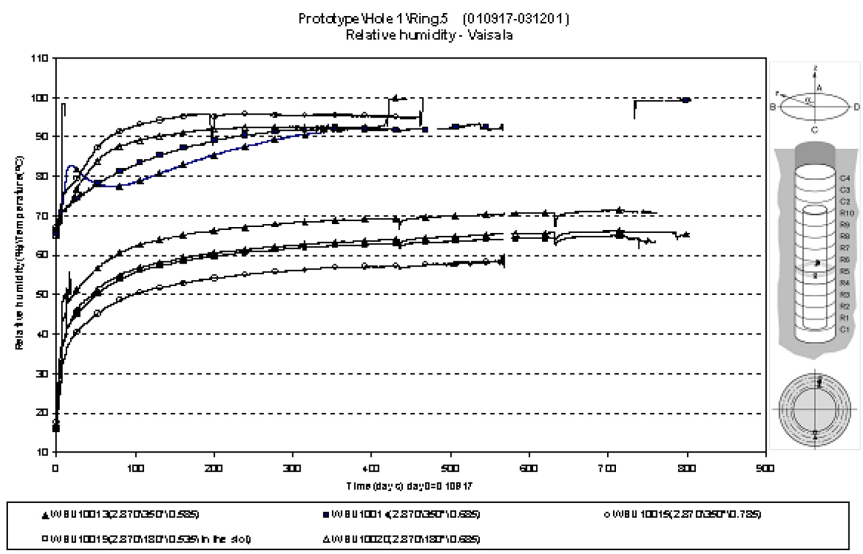

Figure 6: RH and temperature distributions in the buffer in the wettest hole (Hole 1). The upper curve set shows the RH readings, the lower gives the temperature [1, 7].

The RH plottings indicate that nearly complete water saturation (92–95%) had been reached after about one year, which is in contrast with the recorded evolution of the swelling pressure shown in Figure 10. The latter indicates that saturation of the buffer had not taken place even after 2.5 years. The discrepancy is believed here to be caused by water leakage along the cables connecting the RH gauges to the recording units, causing local wetting and early saturation of the clay where the gauges were located.

2.3.4 Field tests – the Äspö retrieval test

A separate experiment was conducted for a detailed study under hydrologically controlled conditions by surrounding the buffer with a filter that was saturated with the same water as in the Prototype experiment and constantly pressurized at 100 kPa. The heater power was 1800 W and the instrumentation similar to that of the Prototype [1, 7]. Figure 7 illustrates the recorded evolution of the total pressure, which includes the 100 kPa water pressure.

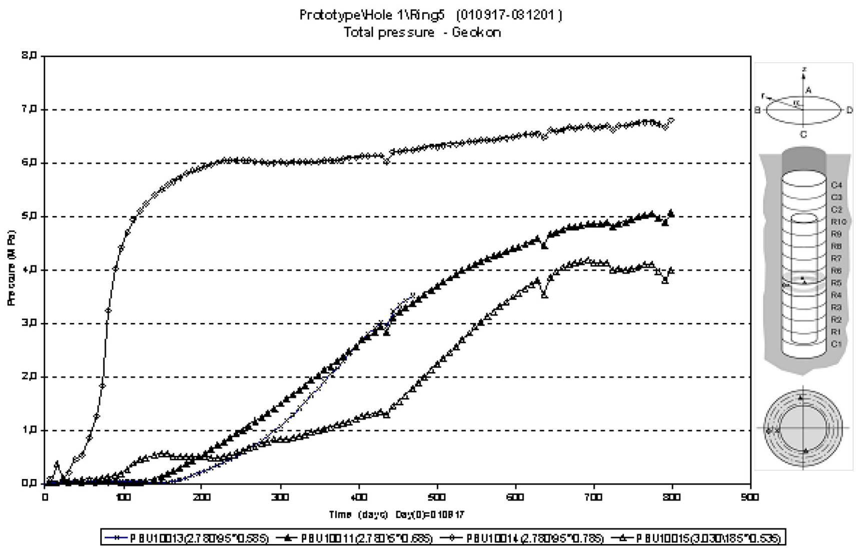

Figure 7: Evolution of total pressure at mid-height of the buffer in the wettest hole. The highest pressure, about 6.7 MPa, was reached after about 2 years while the lowest (4 MPa) appeared close to the canister [1, 7].

The early wetting of the clay near the heater indicated by the pressure recording is explained by migration of water along the pressure tubings from the pressurized filter to the heater (Figure 8). These measurements hence gave an incorrect picture of the wetting trend. The leakage wetted the gap between buffer and heater along its entire length and hence altered the planned conditions for the maturation of the entire buffer.

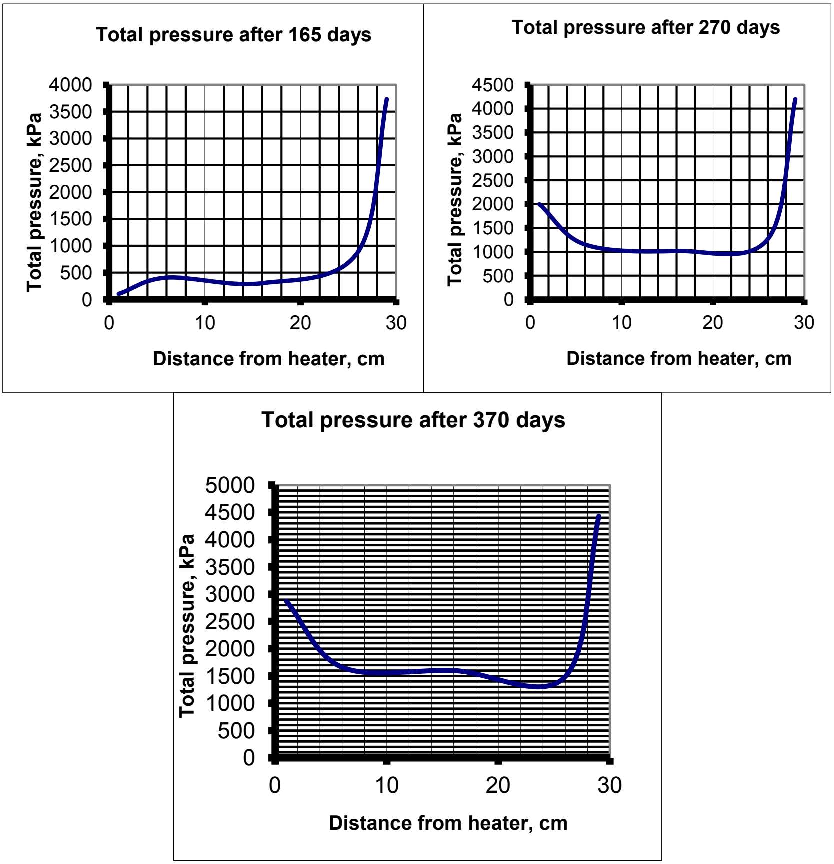

Figure 8: Recorded total pressure in SKB's retrieval test at Äspö [7, 8].

3 Prediction of buffer hydration

3.1 Early attempts

An early attempt was made by Börgesson [9], who realized from the 1D experiments with MX-80 clay referred to in Section 2.2 that the wetting can be described as a diffusion process and that enclosed air in the compacted, initially air-dry clay dissolves and diffuses out without leaving any heterogeneities in the ultimately fully saturated clay. Models based on water flow from the rock combined with diffuse transport in the clay were more complex but gave reasonable results like the model by BGR, Germany, as illustrated by Figure 9 [1]. The difficulty in using such models is to define the hydraulic boundaries of the buffer.

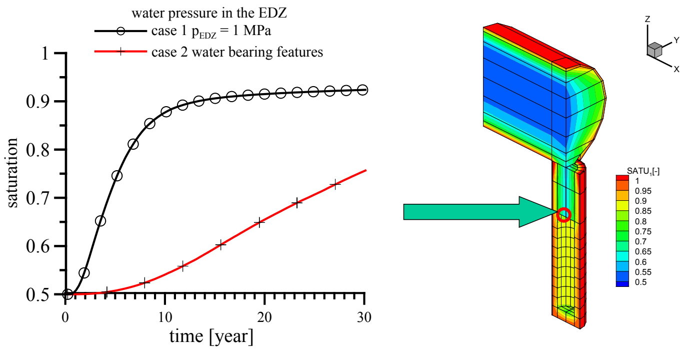

Figure 9: Water saturation versus time for continuously pressurized EDZ in KBS-3 deposition hole by 1 MPa, and without pressurizing and water uptake only through 2 water-bearing fractures (Calculation by Lutz Liedtke, BGR), [1].

3.2 Recent attempts

Later, integration of various physical laws and using the concept of “unsaturated” hydraulic conductivity gave more or less complex semi-theoretical models that were used in the international CROP project for describing the hydration of buffer clay under various conditions respecting boundaries for wetting and expansion/compaction [1]. Such models were tested against the recorded swelling pressure in the Äspö field experiments and the three giving the best agreement with the actual recordings provided the data in Table 2, which also gives the actually recorded data.

Table 2: Recorded and predicted pressure in MPa at the canister and rock at mid-height in the wettest hole after 1 and 2 years from the start of heating [1, 8].

Location	Recorded	COMPASS (UWC)	CODE_BRIGHT (CIMNE, Enresa)	THAMES (JNC)
Canister	1 y = 1.0 2 y = 4.0	1 y = 0.8 2 y = 3.2	1 y = 3.0 2 y = 5.1	1 y = 4.7 2 y = 6.2
Rock	1 y = 6.0 2 y = 6.7	1 y = 2.8 2 y = 3.9	1 y = 5.0 2 y = 7.2	1 y = 6.4 2 y = 7.2

Comparison of predictions and recordings gave the following major conclusions [1, 8]:

- two of the codes gave pressures that were much higher than what was recorded for the clay/canister contact but fairly correct for the clay/rock contact,
- one third code gave fair agreement between measured and predicted pressures for the clay/canister contact but poor agreement for the clay/rock contact,

- none of the codes gave reasonable agreement between measured and predicted swelling pressure for the holes with low inflow rates.

The overall conclusion was that the rates of hydration and maturation for the first 2 years could not be adequately predicted [1, 8]. The discrepancy between measured and theoretically predicted hydration of the buffer was in fact even stronger than indicated by the table since the recordings overrated the hydration because of water leakage through and along cables and tubings as described in this paper. It is therefore not known today whether water saturation of the buffer in KBS-3V type repositories will take a few tens or hundreds of years in deposition holes especially when the deposition holes are not intersected by any significantly water-bearing fracture.

3.3 Present state

At present there is no unanimous way of predicting the hydration and maturation of dense smectite-rich buffer clay even under simple boundary conditions. The various indications of diffusion-controlled water transport suggest, however, that the hydration rate can be estimated for special cases. Thus, for constant buffer volume and unlimited access to weakly brackish groundwater pressurized to a few hundred kPa, and temperature gradients lower than 2.0^{o}C/cm, one can describe the hydration as being diffusive. The various different mechanisms involved in porewater migration, like the electrically generated counteracting impact on porewater flow [8, 10], give different time-related contributions to the transport and hence imply a model with several diffusion parameters. A single averaged diffusion coefficient may do, however, considering the uncertainties in defining the hydrological conditions and constancy of the buffer volume.

Figure 10 illustrates the agreement between measured and predicted hydration of MX-80 clay in one of the Stripa experiments using the diffusion coefficient D = 3E-10 m2/s. The lower value D = 2E-9 m^2/s that gave fair agreement between the predicted and actually measured hydration rate in the Czech experiment (Figure 2) is ascribed to the lower smectite content of the RMN buffer clay.

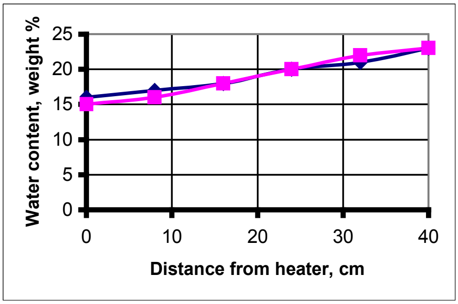

Figure 10: Measured (triangles) and predicted (squares) water content in Hole No 5 in the Stripa buffer test after 27 months for the diffusion coefficient D = 3E-10 m^2/s.

4 Conclusions

- Instrumentation of buffer and backfill in laboratory and field experiments can affect major processes and control the hydration rate. Groundwater leakage can take place along cables and tube connections from the rock to installed sensors and give the impression of much earlier wetting than where there are no instruments.
- The hydration rate of buffer clay is significantly higher for Ca-rich salt water than for electrolyte-poor groundwater.
- The hydration rate increases for water pressures exceeding a few hundred kilopascals.
- The hydration rate of smectite-rich clay is not significantly affected by low and moderate thermal gradients.
- The hydration process of smectite-rich clay can be described as one of molecular diffusion with D = 3E-10 m^2/s to 2E-9 m^2/s depending on the bulk density, mineralogy and initial degree of water saturation.

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