
Increasing the understanding of non-isothermal bentonite re-saturation by the development of an alternative conceptual approach

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Abstract:

Most countries favour compacted air-dry bentonite for engineered barriers in final repositories for nuclear waste. While the hydrophilic properties of bentonite appear to be ideally suited for minimising any contact of water with the waste the process of re-saturation is not fully understood yet. The established codes for modelling the re-saturation of compacted bentonite are based on the assumption that the hydraulic processes can be described as a two-phase flow of liquid water and air. Many models additionally take vapour flow into account. Investigations performed at GRS for some time now suggest, though, that compacted bentonite coming into contact with liquid water will not re-saturate via the liquid water phase but mainly by evaporation in the pore space and subsequently by vapour diffusion.

The present paper describes some of the basics of the established thermo-hydro-mechanically (THM) coupled codes in order to point out discrepancies with microstructural phenomena in a re-saturating bentonite. The alternative conceptual model being more closely based on the microstructural effects avoids these inconsistencies. It is subsequently described in detail as well as the procedure to validate the model. In that the paper also summarises the history of the alternative model.

1 INTRODUCTION

Water present in the host rock is a major problem for radioactive or toxic waste repositories. If it reaches the waste packages, it can corrode the waste canisters and thereby mobilise the contaminants. In this case, the water pathway represents a pathway for hazardous substances into the biosphere. One of the most important functions of geotechnical barriers such as buffer and backfill (see Fig. 1) is therefore to minimise the influence of water on the performance of the repository by reducing and retarding the water flow.

Independently of the type of host rock compacted bentonite is one of the favoured materials for geotechnical barriers. In a repository it is foreseen to be installed in air-dry state. As soon as water reaches the bentonite barrier the process of re-saturation begins. The clay minerals take up water, swell and reduce the pore space and thereby the permeability of the bentonite. Thus, water entering the bentonite is buffered to a certain extent, and concurrently further inflow is impeded effectively. In a fully saturated bentonite the mobile water moves (a) very slowly due to the low permeability and (b) in little quantities due to the small pore space. The present paper is restricted to the re-saturation of a partially saturated (air-dry) bentonite. This period is comparatively short in comparison to the life expectancy of the buffer but understanding the involved processes is nevertheless important for setting up initial conditions for the long-term evolution of the buffer.

The first re-saturation experiments with a view to radioactive waste storage were performed in the early 1980's and were focussed on the time-dependent water uptake of bentonite. It was found that water uptake under laboratory conditions, meaning room temperature and

atmospheric pressure, can be well be described with a Fickian approach (“diffusion law”) with constant “diffusion coefficient”. The bandwidth of values found for the empirical “diffusion coefficient” in laboratory tests with compacted MX-80 bentonite is remarkably narrow and amounts to a little more than one order of magnitude (Kröhn, 2005).

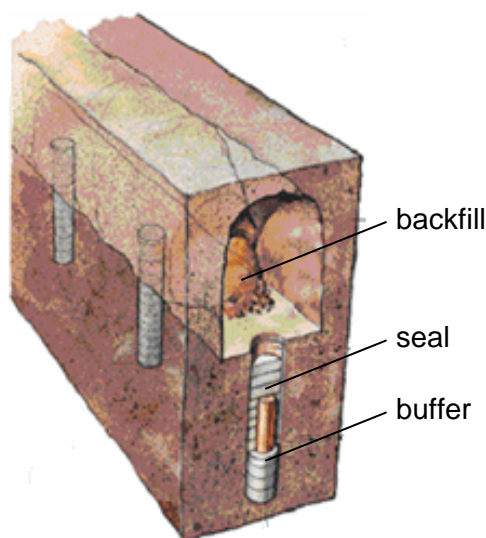


Fig. 1. Bentonite components of the engineered barrier system; after (STUK, 2008).

But the empirical “diffusion” law could not be confirmed for experimental conditions differing from laboratory conditions, especially from room temperature (Börgesson, 2001). New approaches lead in the early 1990’s to the development of thermo-hydro-mechanically (THM) coupled models. Initially developed to investigate the mutual dependencies of heat flow, groundwater flow and mechanical effects in fractured host rock, the necessity to include a bentonite buffer for a complete description of the near-field soon became clear. These models were therefore also used to describe the re-saturation of the bentonite buffer as for instance in the DECOVALEX II exercise (Stephansson, 2001).

Based on the assumption that liquid water migrates in the pore space of the bentonite, these approaches made use of the similarities between the hydraulic phenomena during re-saturation and the classical two-phase flow theory. Several numerical codes have been developed on the basis of the THM-approach, among them:

- ABAQUS, a commercial structural-mechanics code by ABAQUS Inc., USA, formerly HKS Inc. (Hibbitt, Karlsson and Sorensen), with adaptations by Clay Technology AB, Lund, Sweden,
- CODE-BRIGHT, a development of the Technical University of Catalunya, Spain,
- COMPASS, a development of Cardiff University, Great Britain,
- GeoSys/ROCKFLOW, a development of Hanover University, continued at Tübingen University, Germany, and
- LOSTUF, a development of CRIEPI, Japan.

These codes are described in more detail in (Pusch, 2001), (Kolditz et al, 2007) and (Sawada et al., 2006).

With the turn of the millennium GRS became interested in bentonite re-saturation motivated by questions related to the long-term safety of a repository. At that time the literature dealing with the physical as well as the modelling aspects of bentonite re-saturation was scarce. GRS thus commenced its engagement in this field with an intensive theoretical review of the existing codes. Comparing the conceptual models of the codes with the microstructural phenomena in the re-saturating bentonite revealed that the THM-approach to bentonite re-saturation was not entirely consistent (Kröhn, 2003a,b, 2004a). Motivated by these findings GRS decided early to develop an alternative approach that is more closely related to physical phenomena at microstructural scale.

This paper discusses the two-phase flow approach in the THM-concept - which still represents state-of-the-art in modelling re-saturation - as well as the associated problems. An outline of the alternative concept is given together with the stepwise verification of the new approach based on physically increasingly complex problems.

2 COMPARISON OF TWO-PHASE FLOW AND BENTONITE RE-SATURATION

2.1 Macroscopic observations

On a macroscopic level of observation - the level at which the mathematical models are operational - there are striking similarities between the conventional two-phase flow and the hydraulics of bentonite re-saturation in the THM-concept, provided that the above-mentioned assumption of liquid water flow in the bentonite holds. These similarities refer

- to the properties of the forces that drive the fluid flow (capillary forces vs. suction),
- to the flow resistance that depends on the water content and which is generally expressed in terms of a permeability and, finally,
- to saturation as an independent variable.

Fluid flow in the unsaturated bentonite is believed to be driven by the so-called “suction” that is exerted by the clay on the liquid phase as a surface force. In that, suction acts similar on the water as the capillary forces in the conventional two-phase flow.

Suction is inversely proportional to the water content which increases the similarity to the effect of capillary forces even more. However, suction - like most quantities in hydrology - cannot be measured directly. Instead, relative humidity in the pore space at equilibrium with the water content is determined and transformed via Kelvin’s equation into the required suction values. In practise, suction as well as capillary forces are both often described by the well-known equations-of-state (EOS) for two-phase flow.

In the context of two-phase flow a non-deformable matrix consisting of grains with an invariable size leads to the concept of an intrinsic permeability that is depending only on the geometry of the pore space. This intrinsic permeability holds in case of single-phase flow. Reduction of the intrinsic permeability by the effects of a second fluid phase in the pore space is taken into account by a relative permeability that depends on saturation. In case of bentonite re-saturation, a hydraulically non-deformable matrix as well as a constant intrinsic permeability for saturation is assumed. The matrix is hydraulically non-deformable. However, porosity can change according to mechanical displacement.

Like suction the permeability for water cannot be measured directly in the unsaturated bentonite. As in a conventional two-phase flow the permeability for gas is measured instead. Then these values are transformed into relative permeabilities for water using a referring EOS from the conventional two-phase flow theory. For modelling purposes, however, a simple cubic approach is often used in case of bentonite. But the resulting relative-permeability-saturation relations for bentonite re-saturation and conventional two-phase flow are nevertheless comparable.

Finally, while saturation in the conventional two-phase flow depicts the ratio of the fluid volume to the total pore volume, saturation or the degree of saturation in the framework of bentonite re-saturation refers to the ratio of the actual water content to the maximum water content.

2.2 Microscopic observations

Success in many cases seems to confirm the THM-approach to re-saturation. But not all hydraulic phenomena going on at micro scale during re-saturation are properly represented by the macroscopic description as a two-phase flow. Key to an examination of consistency

between micro- and macroscopical description is knowledge about the microstructure of the clay grains and the processes going on during re-saturation. The structure of clay on different scales is illustrated in Fig. 2. On a macroscopic scale clay appears to be formed by grains which seems to comply with the requirements for the two-phase flow theory for a porous medium. But looking more closely at the clay grains they prove to consist of aggregates of discoidal clay particles. On an even smaller scale the particles reveal to be actually stacks of lamellae (platelets of crystalline structure) that contain certain cations in the separating interlamellar space. The cations attract water molecules and bind them onto their surface. This process is called hydration and can bind up to three layers of water molecules around the cation, depending on the cation and the amount of available water. The more water is hydrated the further the lamellae are pushed apart. The process of increasing the interlamellar space is macroscopically observed as swelling of the clay.

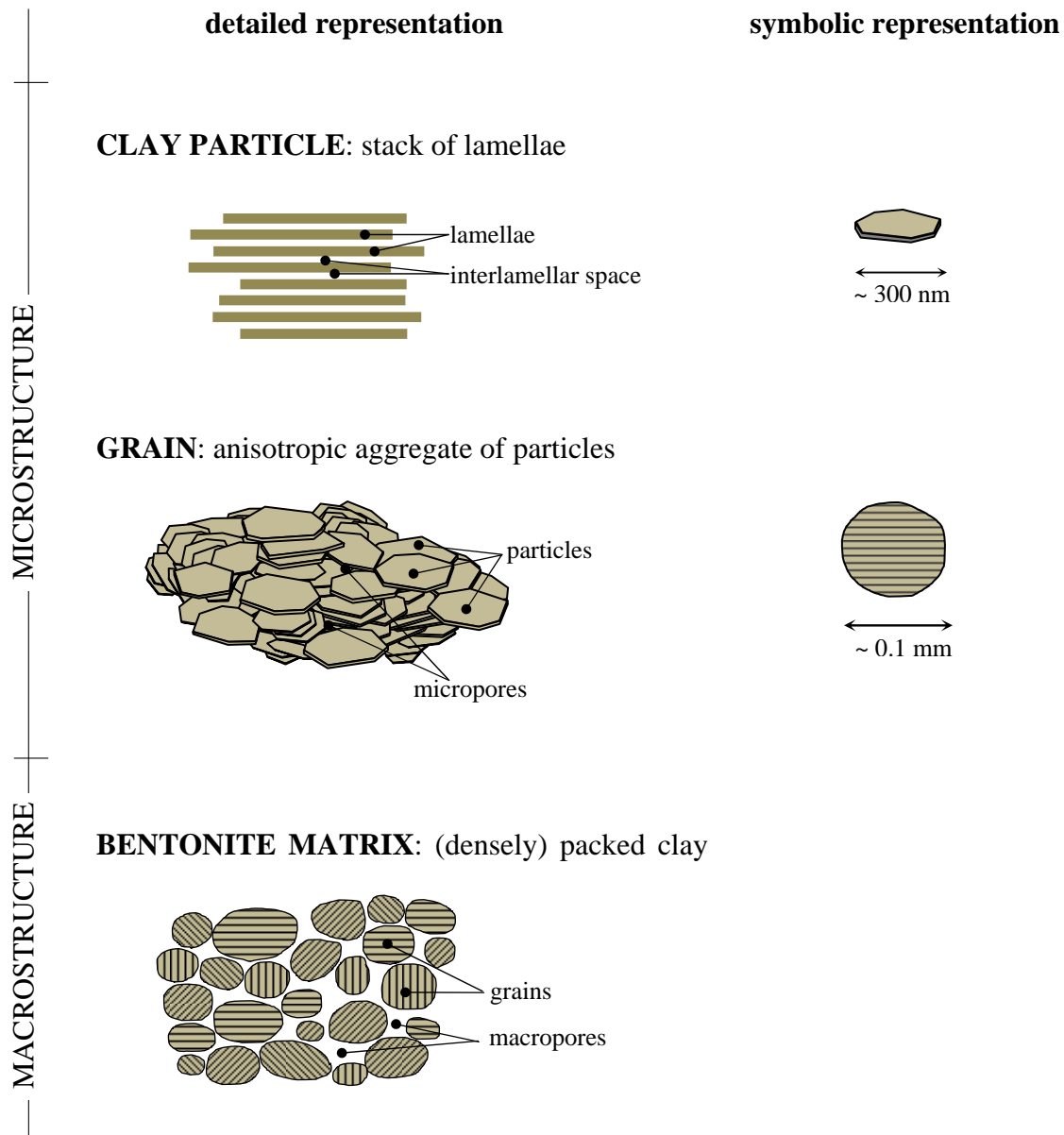


Fig. 2. The structure of montmorillonite on different scales; 3D grain view after Pusch and Yong (2003); modified from (Kröhn, 2003a).

Thus, water propagates not only in the space of the macro-pores between the clay grains. A major amount of water migrates from the macro-pores into the micro-pores of the grains, from where the water is drawn into the interlamellar space of the clay particles. This holds true not only for liquid water but also for water vapour as has been shown by uptake experiments with vapour (e.g. Kröhn 2004b). The particles act as sinks for pore water flow, they immobilise it in the interlamellar space for all practical purposes (e.g. Pusch and Yong (2006) ascribe a “very limited mobility” to hydrated water) and swell at the same time, thereby reducing the available pore space. For a more detailed description of the re-saturation processes with regard to the clay microstructure see also Pusch and Yong (2003). In the end these considerations reveal some inconsistencies between the hydraulic processes going on on a microscopic scale and the macroscopical conceptual THM-model:

- Clay particles and thus clay grains swell with hydration. Under conditions that impede swelling this means also a corresponding decrease of the pore space. But in the THM-models constant grain size is assumed. The mechanical part of a THM-model calculates only the effects of grain displacement.
- Hydration immobilises a considerable amount of water. This effect is not considered in the THM-codes except for a rarely used option in the code CODE-BRIGHT (Gens et al., 1993).
- Clay particles can take up water from water vapour. While vapour flow is considered in most THM-codes it does not contribute to the process of hydration in these models.

So the two-phase flow approach adopted in the hydraulic part of the THM-models is not without contradictions to the physics of re-saturation.

3 THE ALTERNATIVE CONCEPTUAL MODEL

Even if not all of the contradictions pointed out in the previous section were immediately clear after the initial analysis of the existing THM-codes the doubt about the referring concept was strong enough to initiate uptake experiments and concurrent theoretical investigations at GRS. The possibility that vapour diffusion could contribute to bentonite re-saturation as a significant water transport process was already suggested in (Kröhn, 2003a). Experiments performed with MX-80 bentonite under constant volume conditions, room temperature and atmospheric pressure demonstrated a high relevance of vapour transport (Kröhn, 2004b). The results indicated a radically different conceptual model of the re-saturation which is mainly explained by vapour diffusion in the pore space and hydration of water in the clay minerals (Kröhn, 2004c). It consists of three phases, pre-stage, main stage and abandonment stage as described in the following.

The pre-stage begins with the contact of water with the bentonite. In a short period of time water is sucked into the pore space rather fast, mainly by capillary forces, and hydration begins as soon as water is present in the pore space. The reduction of pore space caused by the swelling of the minerals leads to a dramatic reduction of permeability. Since uptake of liquid water by clay minerals is a fast process (Pusch and Yong, 2003), only very little water can enter the bentonite before further water inflow is impeded effectively due to the low permeability. This view is supported by experimental evidence (Kröhn, 2004b) which showed unproportionally high water content and a particularly low dry density only at distances of less than 4 mm from the water contact.

Parallel to the uptake of liquid water evaporation takes place in the pore space at the interface between the fluid and the gas-phase. This becomes significant when the influx of liquid water is low at the end of the pre-stage because progress of the water/air-interface in the pore space is slowed down even further by this process. A rough estimation of the flux densities at the end of the first stage shows that the supply of liquid water through the narrow, already saturated zone equals the subsequent vapour flow further into the bentonite (Kröhn, 2004b). Thus the saturation with liquid water is temporarily limited to the wetted bentonite surface. Measurements of Pusch and Kasbohm (2002) indicate that capillary water uptake during pre-stage takes time in the order of minutes rather than hours or even days.

The water-air interface at the end of this zone thus constitutes a convenient boundary with full vapour saturation for vapour diffusion.

According to the alternative concept the main stage is a phase in which no two-phase flow occurs at all. All the water entering during this stage evaporates at the established interface between liquid water and air. Water transport further into the bentonite takes place only by means of vapour diffusion in the gas phase. The low density of the vapour is here compensated by the high process velocity of the gas diffusion. This stage covers most of the time of re-saturation. Water transport processes during the main stage are depicted in Fig. 3. During main stage the vapour flux at the water/air-interface decreases as the gradient of the relative humidity decreases. Concurrently the water vapour increases the amount of hydrated water next to the interface. This reduces the suction and thus the liquid water flow through the thin saturated zone at the inlet. It is therefore assumed that the water/air-interface moves only little for quite some time.

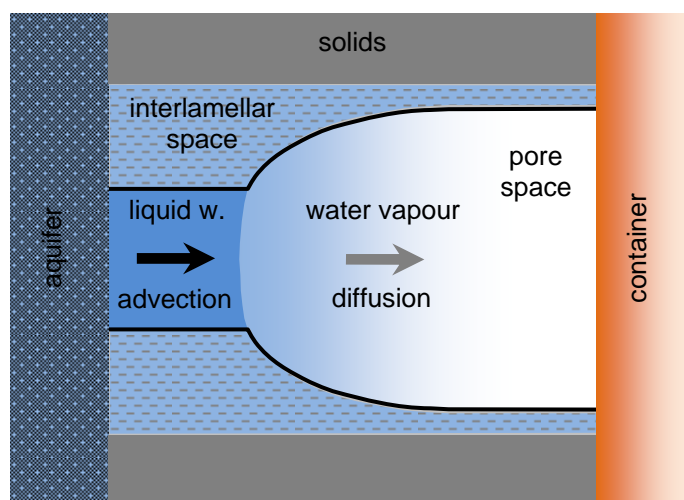


Fig. 3. Water transport during the re-saturation of bentonite; after (Kröhn, 2005).

Note: It is the main stage that makes numerical modelling necessary. Water vapour is transported along the pore channels due to the vapour partial density gradient and concurrently drawn into the clay particles due to the difference of the chemical potential between the water vapour in the pore space and the hydrated water. A balance equation comprising these processes was therefore derived to develop the experimental numerical code VAPMOD (Kröhn, 2004b).

In the *abandonment stage* two-phase flow could begin again when the vapour flux eventually drops below the liquid water flux decreasing likewise. But by then suction would have been reduced considerably due to re-saturation via water vapour. Additionally, the permeability would be very low in an advanced stage of re-saturation. The significance of the third stage is therefore probably low.

Remark: this conceptual model is consistent with a statement in (JNC, 1999) where it is indicated that water movement in the pore space at low water contents is dominated by vapour transport while transport of liquid water is predominant at high water content. Unfortunately, the primary literature is in Japanese so the background of this remark could not be checked.

4 VALIDATION

From the beginning it was planned to test the alternative conceptual model step by step against uptake tests with a physically increasing complexity, beginning at ambient conditions, until the model was shown to be viable even under repository conditions. This procedure was

foreseen to ensure that effects previously not considered could clearly be identified and incorporated in the model.

Just one parameter for the mathematical model describing the hydration dynamics had to be derived newly from experiments. However, after supplying the numerical model with realistic values for this parameter first generic calculations lead to surprising conclusions. It became clear that hydration is a very fast process in comparison to vapour diffusion in the pore space. Thus, when hydration dynamics were dropped in favour of instantaneous hydration the previously derived balance equation of the new conceptual model could be transformed into the well-known empirical “diffusion law” as discussed in detail in (Kröhn, 2005). This transformation allows to quantify the empirical “diffusion coefficient” using exclusively physically meaningful parameters. In an example that is given in (Kröhn, 2005), too, a “diffusion coefficient” is calculated by this means which differs from the calibrated value only by a factor less than 3. Therefore, the validation procedure was continued on the basis of this simplified balance equation.

Having shown a good correspondence between the vapour diffusion model and experimental results for laboratory conditions as shown exemplarily in Fig. 4, water uptake under an increased hydraulic pressure was tackled in the next step. Here, the new model produced convincing results in comparison to a referring laboratory experiment - as shown in Fig. 5 (Kröhn, 2006) - after correcting the thickness of the water saturated zone according to the hydraulic pressure at the water contact.

When modelling the isothermal tests proved to be successful, temperature dependencies were incorporated in this equation. The development of a new code called VIPER (**V**apour transport **I**n **P**artially saturated bentonite as **E**ngineered barrier for **R**epositories) was begun to model non-isothermal re-saturation. The complete set of assumptions underlying the non-isothermal model can be found in (Kröhn, 2008). A laboratory test consisting of a heating-phase in a closed system and a second phase with an additional liquid water uptake provided the basis for this exercise. In the first phase only moisture redistribution due to temperature changes took place which was used to check the validity of the non-isothermal vapour flow approach. Afterwards, the second phase was used to draw conclusions from modelling water uptake. The results are shown in Fig. 6. In the end the alternative conceptual model in the non-isothermal form proved to be valid even under repository conditions (Kröhn, 2008).

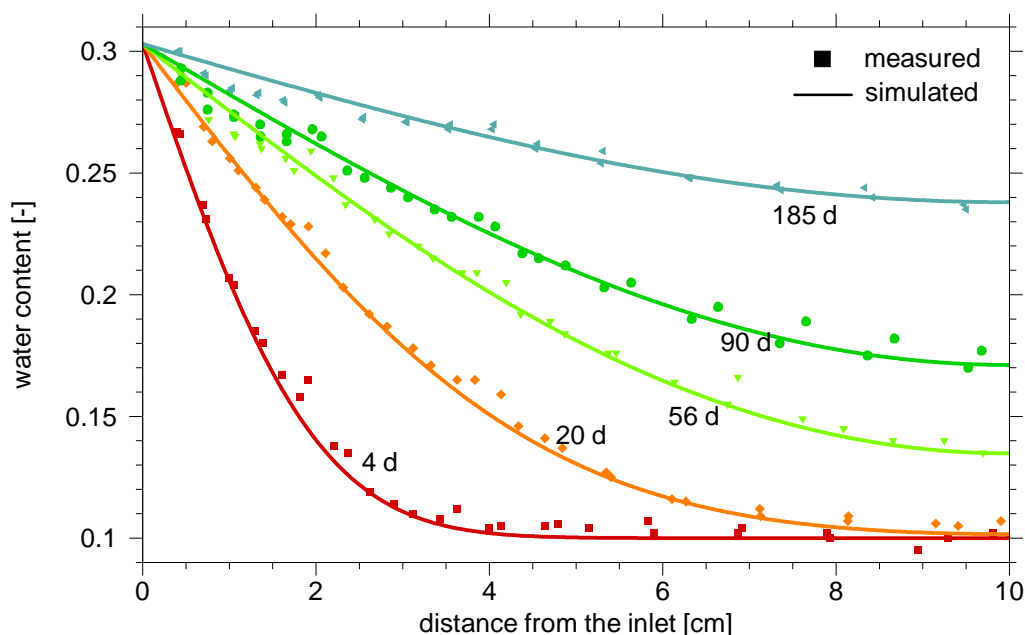


Fig. 4. Measured and calculated water content distributions in an isothermal uptake experiment with atmospheric water pressure; after (Kröhn, 2005).

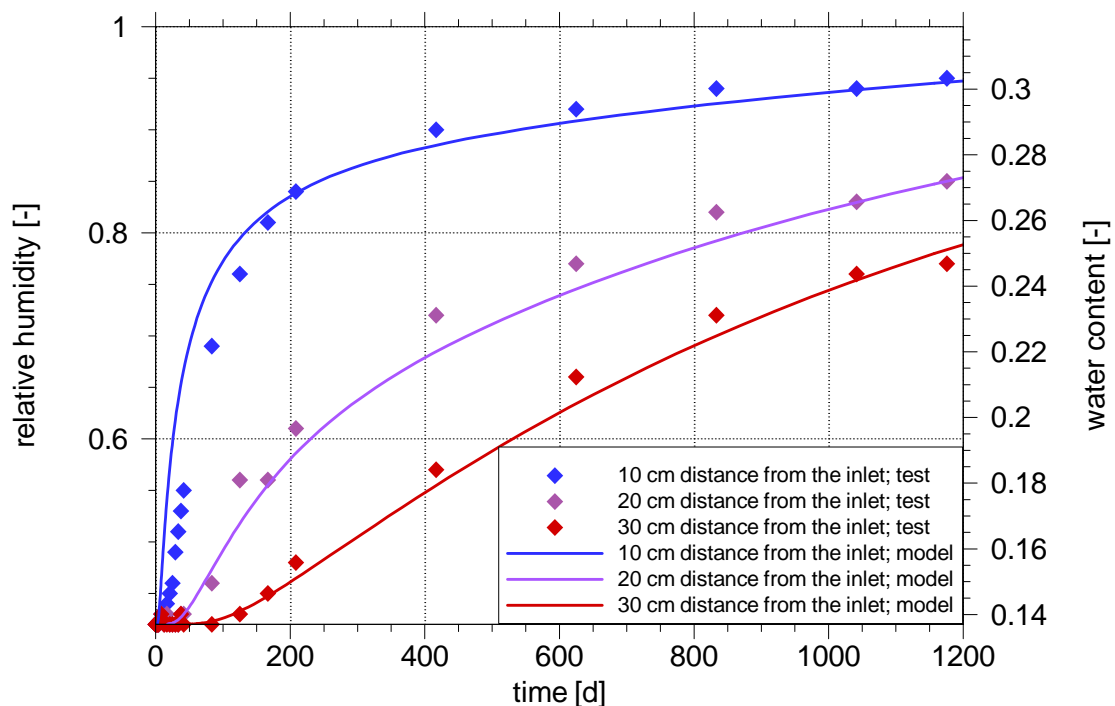


Fig. 5. Measured and calculated breakthrough curves of relative humidity in an isothermal uptake experiment with increased water pressure; from (Kröhn, 2006).

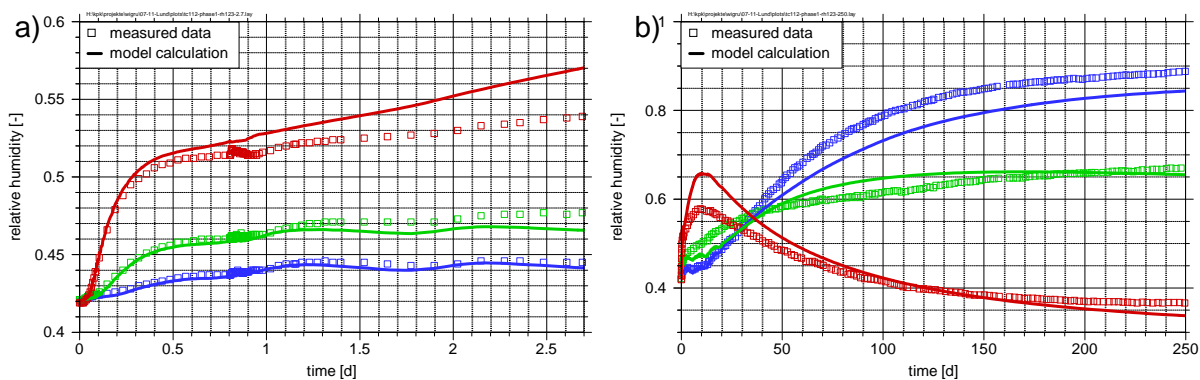


Fig. 6. Measured and calculated relative humidity in a non-isothermal experiment with a) no water inflow and b) with water inflow at increased pressure; from (Kröhn, 2008).

5 DATA REQUIREMENTS

A particularly appealing aspect of the alternative re-saturation approach is the fact that most data and relations required by the model are already well established. In the following a complete list of the necessary relations and data is given:

Relations used in the code:

- partial saturation pressure of water vapour (a function of temperature)
- saturation vapour partial density (a function of partial saturation pressure)
- relative humidity (derived from vapour partial density and saturation vapour partial density)
- porosity (a function of water content and bentonite dry density)
- molecular diffusion coefficient (a function of temperature and pressure)
- macroscopic diffusion coefficient (a function of the molecular diffusion coefficient, porosity, and tortuosity)

Problem specific data:

- density of the solids
- density of the interlamellar water
- initial relative humidity or initial water content
- initial bentonite dry density
- end porosity
- water content (a function relative humidity and temperature (adsorption isotherm) for a specific bentonite)
- time-dependent temperature field

The macroscopic diffusion coefficient, the end porosity, and the temperature-dependency of the adsorption isotherm are not exactly known. But they can vary only within very limited boundaries. All other quantities cannot be altered at all. Under these stringent conditions a good data correlation between measurements and numerical simulations as demonstrated in Fig.s 4 to 6 is evidence for a viable alternative conceptual model.

6 CONCLUSIONS AND OUTLOOK

A new model has been developed that is now able to describe the non-isothermal re-saturation of compacted bentonite under constant volume conditions. This type of problem does apparently not require a coupling of hydraulics to the mechanics. Whether the coupling of hydraulics to heat transport can be neglected as well is still to be investigated. Application of the model to laboratory experiments shows good results. Isothermal as well as non-isothermal uptake tests could be simulated with code VIPER satisfyingly well. The measured and the calculated transient relative humidity distributions coincide in terms of absolute values as well as in development of the relative humidity. In the light of the complexity of the model on the one hand and the fact that very little possibilities exist to calibrate the model to a specific problem on the other hand this outcome is quite encouraging.

However, simply presenting a working model can not be considered as a validation of the conceptual model. This must be the conclusion from the fact that the THM-approach as well as the alternative vapour diffusion approach are able to reproduce the re-saturation phenomena despite the radically different physical basis of these models. It appears, though, that the conceptual basis of the vapour diffusion approach is sounder than the THM-approach since it is not only consistent with the macroscopic observations but also with the phenomena on the microscopic scale. The alternative concept indicates therefore nevertheless strongly that a diffusion process dominates the transport of water in the re-saturating bentonite and not an advective liquid water flow. In fact, after promoting the new model for some time this idea is also either considered or even followed by other parties (cp. Pusch et al. (2004), Hokr and Frydrych (2008), Åkesson et al. (2007)).

While an important milestone has been reached with the non-isothermal model a wide field of further work and research still remains to be performed like

- checking the model against in-situ tests in order to increase confidence in the model
- investigating applicability of the model to re-saturation of bentonite-sand mixtures. If successful, the input data could easily be adapted to other bentonite-sand ratios provided the same specific type of clay is used, e.g. Ca- or Mg-montmorillonite so there will be no need for additional time-consuming and expensive laboratory measurements.
- developing a concept to model the uptake of liquid water in order
 - to provide more realistic boundary conditions for the vapour flow model, and thus
 - to include the capability of the model to simulate the abandonment phase
- including a thermal part in code VIPER to get a full TH-model
- extending code VIPER to 2D (Cartesian or cylindrical coordinates)

With the work presented here it has become even more important to reconsider the conceptual basis of the established THM-models as has already been suggested earlier. By and large these models still ignore the fact that a considerable amount of water, if not

practically all water, is located in the interlamellar space where its mobility is severely restricted. But it would be of utmost importance to know how much water actually moves freely in the pore space if chemical or biological processes are additionally to be considered.

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