
Investigation of Uncertainties in the Re-saturation Model of Bentonite as near-field Buffer in a Spent Fuel Repository in Granite

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ABSTRACT: Bentonite plays an important role in the design of underground repositories for toxic and radioactive waste. The hydrophilic properties of bentonite appear to be ideally suited for minimising any contact of water with the waste. However, the behaviour of bentonite during re-saturation is a very complex phenomenon which may be influenced by hydraulic, mechanical, thermal and chemical processes.

A short discussion of the microstructure of bentonite provides the base for a sound conceptual model of re-saturation. This model does not compare satisfyingly with the already existing numerical simulation codes, every one of which is based on unsaturated flow or two-phase flow in the pore space. Thus, new conceptual models were developed at GRS for the re-saturation of bentonite. In accompanying experiments a data base for checking the new re-saturation models as well as the already existing models was created.

The experiments show a high relevance of re-saturation via water vapour. Without further experiments it cannot be excluded that the main water transport mechanism is vapour diffusion in the pore space and not flow of liquid water. This would call the applicability of two-phase flow based codes to complex physical situations into question.

1 INTRODUCTION

One of the main problems of the underground disposal of radioactive and toxic waste is the water present in the host rock. If it can reach the waste packages unhindered, it can attack the waste canisters and thereby mobilise the contaminants. In this case, the water pathway represents a pathway for hazardous substances into the biosphere. For a repository in salt rock water intrusion occurs only in the case of an altered evolution. But in crystalline rock it occurs also during the normal evolution of a repository.

Geotechnical barriers are meant to reduce these detrimental effects by delaying and minimising the flow of water in the near-field of a repository. All over the world, bentonite is considered for this purpose as a material for the backfilling and sealing of emplacement galleries and boreholes. As soon as the bentonite, which is brought in in air-dry condition, comes into contact with water, the process of re-saturation begins. The clay minerals absorb the water, swell and thereby reduce the pore volume, which also drastically reduces permeability. Incoming water is thereby stored to a certain extent in the bentonite, and the influx of further water is hindered very effectively. In a fully re-saturated bentonite, the water moves very slowly due to the low permeability and also in very small volumes due to the low porosity. These characteristics make bentonite appear an ideal material for geotechnical barriers.

However, the behaviour of bentonite at changing water contents is a very complex phenomenon which may be influenced by hydraulic, mechanical, thermal and chemical processes. These

processes are coupled by a range of parameters and equations of state. Some of these couplings are highly non-linear and some are still not fully understood today despite year-long and substantial research. A comprehensive and universally valid description of the processes is therefore difficult. Nevertheless, the relevant characteristics of the bentonite have to be registered so that they can be duly considered in long-term safety analyses.

The present paper is restricted to the re-saturation of an initially dry or partially saturated bentonite, which represents a decisive partial aspect of the performance of bentonite barriers. The simulation of this process requires a thorough description with a numerical model. So far, efforts in this respect have been aimed at covering as many of the complex coupled physical processes as possible by a model, with the result that these models became very intricate by nature. On the other hand, a re-saturation model should be as simple as possible in the context of long-term safety analyses.

Therefore an attempt has been made at GRS to come up with simple but satisfactory mathematical descriptions of the re-saturation behaviour in bentonite and to validate these by way of re-saturation experiments. The results permit a completely different assessment of the relevance of the processes involved in re-saturation than previously assumed even if these alternative interpretations have yet to be confirmed by additional investigations. If these new views indeed hold true this has consequences for the applicability of the already established models.

2 MICROSTRUCTURE OF BENTONITE

Bentonite consists mainly of montmorillonite, a clay mineral. The typical composition of an MX-80 bentonite is: 65-75 % montmorillonite, 10-14 % quartz, 5-9 % feldspar, 3-5 % carbonate, 2-4 % mica and chlorite as well as 1-3 % heavy minerals [12]. The montmorillonite alone is responsible for the swelling of the bentonite. In order to understand the phenomenon of re-saturation it is therefore necessary to have a closer look at the montmorillonite.

The clay minerals as indicated in Fig. 1 consist of stacks of electrically charged crystal layers, the so-called lamellae. They are bonded by cations e. g. Na^+ , Mg^{2+} , Ca^{2+} or K^+ which are located in the space between the lamellae. This space is called the "interlayer" or the "interlamellar space". Depending on the respective interlayer cation between three and twenty lamellae can be stacked on top of each other to form a particle. Upon the entrance of water into the interlamellar space the water molecules deposit around the interlayer cations which means that the interlayer ions become hydrated. This widens the interlamellar space and let the particles swell. The maximum absorbable amount of water depends on the kind of interlayer cations. The movement of water in the interlamellar space is negligible.

Natural bentonite is broken up by the production process, with an ensuing grain formation. The grain size of industrially processed bentonite powder is between 0.1 and 2 mm [10], [12], [13]. Compaction of bentonite powder establishes then a porous medium. Thus, in principle there are two types of volumes to distinguish: the pore space between the clay grains and particles and the interlamellar space within the clay particles.

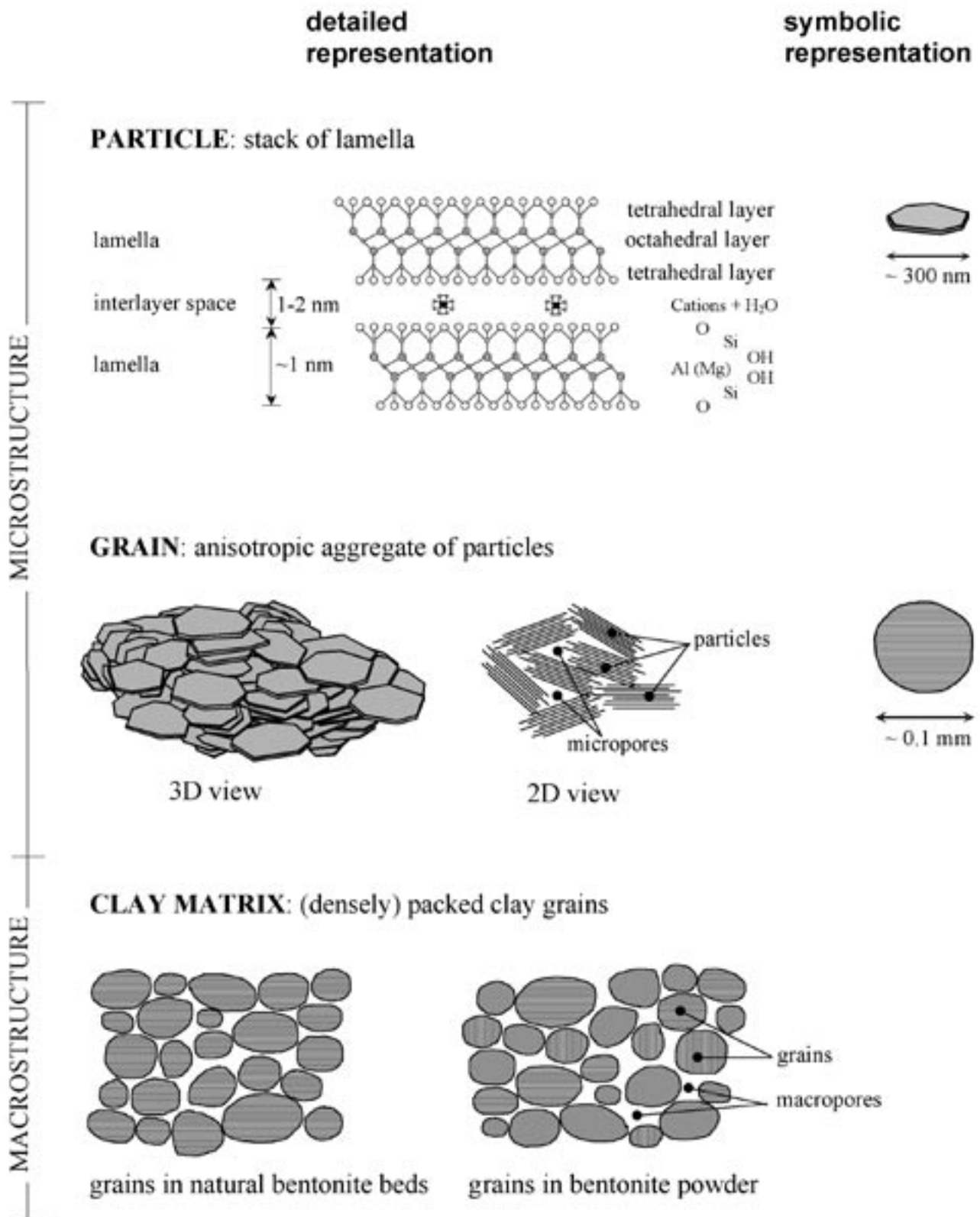


Fig. 1. The structure of montmorillonite on different scales from [7]; detailed particle view after [5]; 3D grain view after [14]; macroscopic grain view after [12].

3 CONCEPTUAL MODEL OF RE-SATURATION

The process of water uptake is illustrated in Fig. 2 considering the re-saturation of a buffer in a repository as an example. For the sake of simplicity this is assumed to be a one-dimensional process. Air-dry bentonite is filled in the annulus between waste canister and host rock and water enters the bentonite from the water-bearing excavation damaged zone (EDZ). At first, water flow is driven by hydraulic pressure and capillary forces into the pore space as assumed in the classic two-phase flow theory. But the chemical potential of the pore water is higher than the chemical potential of the interlayer water and then the process of hydration begins. It means that the cations in the interlayer of the clay particles pull the water molecules from the pore space into the interlamellar space. Thus hydration acts as a sink for the pore water and reduces the pore space at the same time. As a consequence the permeability is reduced concurrently with the porosity and this effect decreases the pore water flow. A reduced pore water flow constricts in turn the hydration. This means, that the re-saturation can basically be described by two coupled processes only, the water transport in the pore space and the hydration.

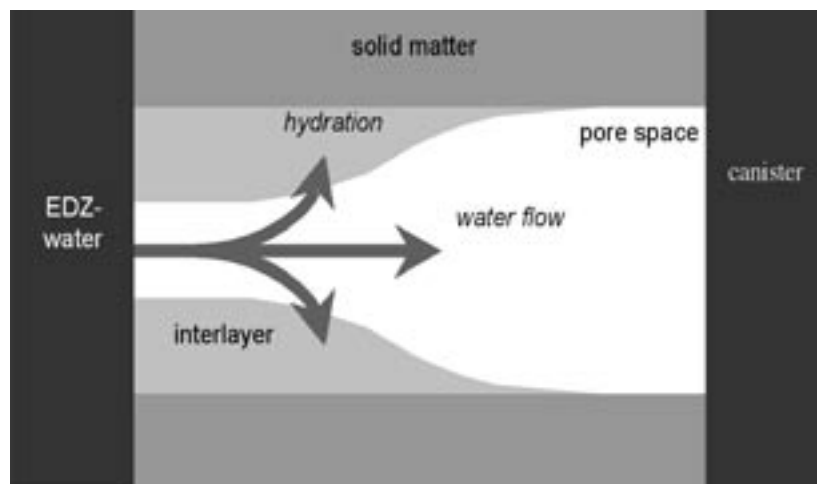


Fig. 2. Re-saturation of a bentonite buffer.

Obviously, the rate of water flow into the interlamellar space – called “hydration rate” further on – is of crucial importance to the dynamics of the resaturation of bentonite. The ratio between the hydration rate and the flow rate in the pore space controls not only the propagation of pore water but also the time-dependent distribution of interlayer water. The underlying mechanism becomes clear if two extreme situations are contemplated: resaturation with a very low hydration-to-flow ratio and resaturation with a very high hydration-to-flow ratio. The resulting effects are illustrated in Fig. 3. In case of a very low hydration-to-flow ratio the pore space is filled with water before a significant transfer of water to the interlayer space occurs. In case of a very high hydration-to-flow ratio the pore water can proceed into the bentonite only after hydration at the pore water front is more or less completed, meaning that the pore space has been reduced to a minimum behind the front.

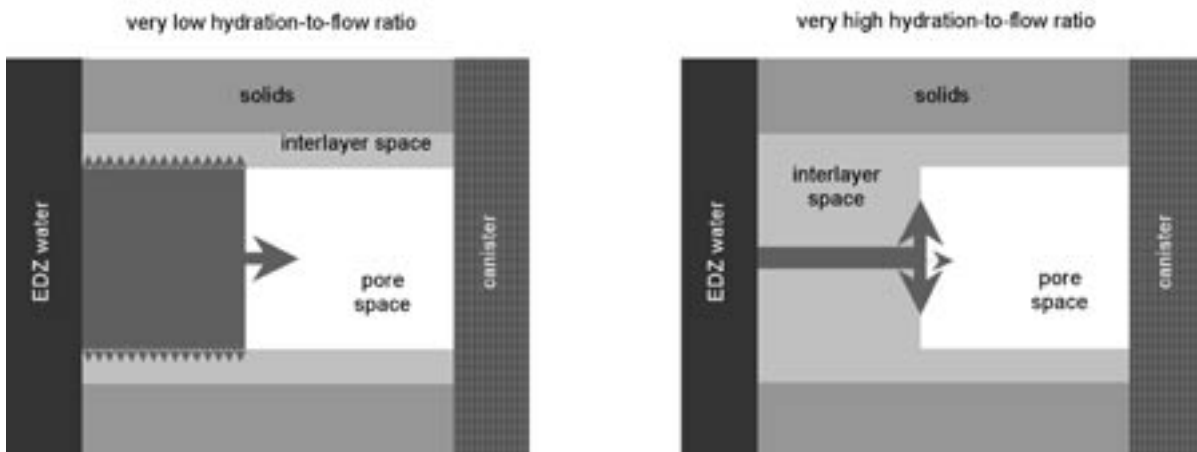


Fig. 3. Effect of the hydration rate on the water distribution in the bentonite.

Having found the relevant processes it is of interest to identify the corresponding primary variables. Water can be present in the pore space in either phase state, as liquid water or as water vapour or both. Transport of liquid water is basically a Darcy flow according to the two-phase flow theory. The driving forces are the capillary pressure and the gradient of the hydraulic pressure. Capillary pressure in the framework of the classic two-phase flow theory is dependent on the volumetric saturation of the pore space with liquid water. In a rigid interpretation it therefore depends on the pore water content and the porosity. The same applies to the effective permeability. The porosity in turn is a function of the local amount of hydrated water.

Transport of water vapour can be described as binary gas diffusion of vapour and air with a Fickian type of flow law. Here, the flow rates depend on the gradient of the vapour partial pressure and the porosity.

The hydration rate depends on the difference between the chemical potential of the pore water and the interlayer water. While the chemical potential of the interlayer water is a function of the local amount of hydrated water, the chemical potential of the pore water depends in the absence of liquid water on the relative humidity. If liquid water is present a chemical potential corresponding to vapour saturated air is generally assumed.

From this short outline of the conceptual model of re-saturation it follows that the mathematical description of both, water flow in the pore space and hydration requires a distinction between the local amount of pore water content and the local amount of interlayer water. It is not sufficient to consider just the total water content as a primary variable.

4 CURRENTLY AVAILABLE NUMERICAL MODELS

Several codes have been developed or advanced in order to enable quantitative predictions of bentonite re-saturation, 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 for THM problems;
- COMPASS, a development of Cardiff University for THM problems;
- FLAC, a commercial structural-mechanics code by Itasca Consulting Group, Inc., USA with adaptation options for THM problems;

- FRACON, a contract development by AECL for THM problems;
- MUFTE_UG, a development of Stuttgart University; multi-phase-flow code for TH problems with extensions for expandable materials;
- ROCKFLOW (ROCKMECH), a development of Hanover University, modified at Tübingen University to address THM problems;
- ROCMAS, a development of the Lawrence Berkeley Laboratory for THM problems;
- THAMES, a development of Kyoto and Iwate Universities, Japan, for THM problems.

These codes are described in more detail in [15], [2], [4], [11] and [16]. All these codes share some fundamental assumptions, especially the concept of a flow in the unsaturated area, an elastic stress-strain behaviour, and the propagation of heat due to thermal conduction. The developers obviously also share the view that thermal conduction is the only relevant process of heat propagation in THM modelling.

It is most interesting to note that the basic approach of unsaturated flow or two-phase flow, respectively, is taken for granted in all the models. In view of the conceptual model discussed above the conventional two-phase flow approach has an essential weak point. The process of hydration is not considered and thus, no difference is made in the balance equations between the pore water and the immobile water in the interlayer. Only the local total water content or an equivalent measure is considered in the above mentioned THM-codes. Additionally, the related balance equations use the assumption of an inert solid matrix.

This has two severe consequences. First, it is not possible to separate the share of the capillary pressure in the matric suction from the share that is caused by the gradient in the chemical potential between pore water and interlayer water. Both suction forces are mixed up and the sum of both is derived from a so-called „retention curve“ as a function of the total water content. This is not correct because capillary pressure and suction depend on different quantities: the capillary pressure is controlled by the volumetric saturation of the pores with the liquid as well as the pore size (this applies to the permeability as well) while the suction is controlled by the chemical composition of the liquid phase and the local amount of interlayer water.

Second, a considerable hydration via water vapour occurs at an elevated relative humidity if no liquid water is present in the pore space of the bentonite. But without the distinction between pore water and interlayer water all the swelling properties have to be described as a function of the local water content which is not significantly increased by the vapour mass. The only way to work around the problem and stick with the total water content as a prime variable is to assume instant equilibrium of the chemical potentials as is done in CODE-BRIGHT. But this way the dynamics of hydration are lost. The models are therefore not able to reproduce the re-saturation via water vapour.

The classic two-phase-flow theory thus does not suffice to describe the phenomenon of re-saturation correctly. The latest modifications to COMPASS [1], ROCKFLOW [6] and very recently also to the MUFTE_UG code [3] are aimed at eliminating at least part of the deficiencies described above. However, this is still done on the basis of the two-phase-flow theory, the applicability of which to bentonite has not been shown yet without doubt.

So far, none of the available models considers pore water and hydrated water separately. Only in the MUFTE_UG code this is intended to achieve but this new option is only at an early stage of development. Thus, none of the available models is able to describe the capillary pressure, effective permeability and the hydration via water vapour correctly.

5 RE-SATURATION VIA WATER VAPOUR

The relevance of the re-saturation via water vapour has by and large been seen as a secondary problem. There are several codes that take vapour diffusion into account but only as a transport process for water in the pore space. And indeed, the contribution of vapour to the hydration is not self-evident. While hydration takes place irrespective of the phase state the density of liquid water is almost 5 orders of magnitude higher than that of vapour saturated air at 20 °C. The only reason that could justify a closer investigation of this process is the fact that binary gas diffusion is a very fast transport mechanism in comparison to the flow of liquid water in the bentonite.

In order to investigate the dynamics of water uptake several resaturation experiments with liquid water as well as with water vapour have been performed at GRS [9]. The basically one-dimensional experimental set-up provided moisture distributions as a function of time. For the first time such water content profiles were gathered with a high resolution in space as well as in time [8]. Parallel to the experiments, a numerical vapour diffusion model has been developed that considers

- vapour diffusion in the pore space;
- hydration controlled by the:
 - relative humidity in the pore space and;
 - the local mass of hydrated water and.
- varying porosity.

Fig. 4 shows that the time dependent moisture distribution in the experiments for uptake of water vapour could be reproduced to a certain extend with the vapour diffusion model. The amount of hydrated water as well as the trend of the distributions coincides well with the measured data. Only the uptake dynamic seems to be somewhat off. Considering that the vapour diffusion model at the present stage incorporates several simplifications the agreement is satisfying.

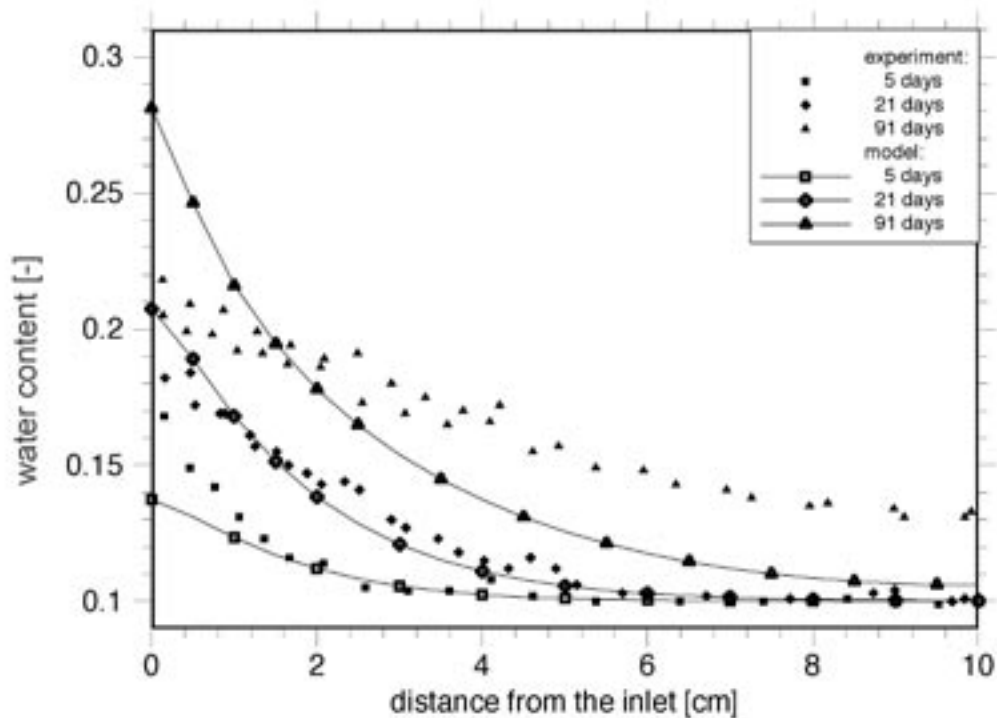


Fig. 4. Water content as a function of penetration depth; re-saturation with water vapour.

Interesting conclusions can be drawn from a comparison of the results of the vapour diffusion model with the data from the uptake experiment with liquid water as illustrated in Fig. 5. It shows that about one half of the hydrated water can be explained by means of binary gas diffusion. This means at least that a considerable part of the water uptake is due to vapour diffusion. Assuming that vapour diffusion fades to Knudsen-diffusion in an advanced stage of saturation it could even be possible to explain the bentonite saturation exclusively by vapour flow due to an increasing diffusion coefficient. In that case the uptake of liquid water may be limited to the wetted bentonite surface. This would mean that no two-phase flow occurs at all during re-saturation.

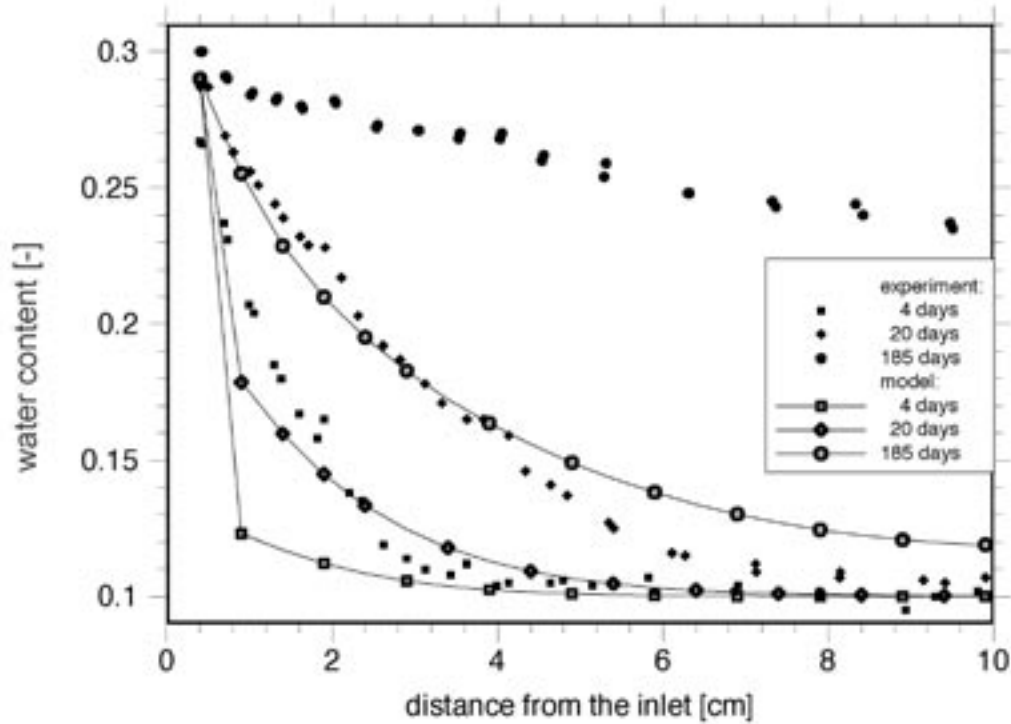


Fig. 5. Water content as a function of penetration depth; re-saturation with liquid water.

6 UNCERTAINTIES IN THE APPLICATION OF RE-SATURATION MODELS

The look into the microstructure of bentonite has shown that hydration is a central process for the re-saturation. A simple conceptual model needs only to consider hydration and water transport in the pore space. Contrary to this the established THM-codes do not take hydration as a process into account. Pore water and hydrated water are not distinguished and hydration via water vapour is not considered with only one exception. On the other hand, the new vapour diffusion model and the corresponding uptake experiments show a high relevance of hydration via water vapour. This means that the two-phase flow concept as currently applied to the re-saturation of bentonite appears to be not too close to reality and not every relevant physical process is captured in the established THM-codes. Physics that are not considered in this approach must therefore be encapsulated in the calibrated parameters if the water uptake is described reasonably well by those models. But there is a price to pay for this kind of approach. Calibration in general means only that an optimum fit of measured and calculated results is sought by varying parameters of a numerical

model which is based on a previously chosen conceptual model. Afterwards the numerical simulator may reproduce the experiment from which the calibrated parameters were derived fairly well. But this is simply an optimisation process for one specific geometrical and physical set-up and does not prove the correctness of the underlying conceptual model. In other words such a model is valid only for the conditions for which the parameters in use are calibrated. The change of just one factor from those conditions can provoke totally misleading numerical results.

This is a severe problem in case of the THM-treatment of re-saturation because there are so many parameters and equations of state involved. In a rigid scientific sense every single parameter and relationship has to be determined under exactly the same conditions to provide a meaningful set of input parameters. And a model can then be trusted only if it is applied to exactly those conditions. This applies as long as it is not clear that all relevant processes are taken into account for the modelling.

But the interest in modelling re-saturation of bentonite is actually that of being able to predict the process under circumstances that are beyond the scope of any realistically conceivable experiment. It is therefore of prime importance that the conceptual model underlying the numerical simulators is proved to be valid to the highest possible extend. This is even more so considering that even if the processes and their relevance are perfectly clear there always remain the uncertainties due to the inhomogeneities of the material and the idealisations of the geometry.

7 CONCLUSIONS

The results of recent investigations cast doubt on the applicability of the currently established THM-codes to the bentonite re-saturation under complex physical conditions. It originates from the fact that the process of hydration via water vapour is on one hand not considered properly in the models - if at all. On the other hand it has been proved to be highly relevant. Without further experimental evidence it has to be conceded that it could even be the dominant process. This would give two-phase flow only relevance for a very short time period after the first contact with liquid water. Thus, it would call the basis for the hydraulic part of any of the re-saturation models into question.

In the light of the new experimental results the actually relevant process of pore water transport during re-saturation appears not to be clear. Further investigations concerning the pore water transport and concurrently a revision of the conceptual model of bentonite re-saturation are therefore highly advisable in order to re-establish confidence in the performance of future re-saturation models.

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