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Transient boundary conditions in the frame of THM-processes at nuclear waste repositories

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Abstract. In nuclear waste repositories, initially unsaturated buffer is subjected to constant heat emitted by waste canister in conjunction with peripheral hydration through water from host rock. The transient hydration process can be potraied as transformation of initial heterogeneity towards homogeneity as final stage. In this context, this paper addresses the key issue of hydro mechanical behaviour of compacted buffer in context of clay microstructure and its evolution under repository relevant loading paths and material heterogeneity. This paper also introduces a unique column experiment facility available at Ruhr Universität Bochum, Germany. The facility has been designed as a forerunner of field scale testing program to simulate the transient hydration process of compacted buffer as per German reference disposal concept. The device is unique in terms of having proficiency to capture the transient material response under various possible repository relevant loading paths with higher precision level by monitor the key parameters like temperature, total suction, water content and axial & radial swelling pressure at three different sections along the length of compacted soil sample. In general, a larger spectrum of loading paths/scenarios, which may arise in the nuclear repository, can be covered precisely with existing device.

1 Introduction

In recent years, considerable research has been carried out to evaluate the behavior of compacted clay based buffer material in the frame of coupled thermo-hydro-mechanical (THM) processes at nuclear waste repositories. In this context, initially unsaturated buffer is subjected to radioactive decay heat emitted by waste canister in conjunction with peripheral hydration through water from host rock. The process of hydration triggers swelling of inter layers space in clay particles and transform the material from initial heterogeneous to homogeneous one as final state. This shift of heterogeneity towards homogeneity simultaneously changes the local volume constraint conditions. The challenge is to anticipate the material behavior under such transient boundary conditions in the frame of THM-processes.

The inherent heterogeneity in compacted clays is characterized by its two distinct continua of pore space namely macropores and micropores, whereas the global heterogeneity in compacted buffer bricks in disposal pits is attributed by technological voids. The technological voids are referred to as the macro-pores related to different interfaces involving the buffer material [1-2]. Dual porosity model appears to be well suited to depict this characteristic of compacted clays. The macroscopic volume change due clay-water interaction is associated with evolution of microstructure and its interaction with macrostructure. Thus, to anticipate the complex hydro-mechanical behaviour of compacted buffer in nuclear waste repository, the influence of initial heterogeneity and applied boundary conditions on microstructure evolution must be considered.

Based on the inputs from phenomenological investigations on microstructure evolution, various dual porosity models have been reported such as volume change behavior due to hydration [3-5], coupled THM behaviour in frame of nuclear waste repository [6], water retention behaviour [7]. Enhancement of existing dual porosity models requires incorporating the inputs regarding clay microstructure and its evolution from phenomenological investigations identical to the actual one. The errors associated with element tests due to scale effect, steady state boundary conditions and lack of generating repository relevant loading scenarios can be eliminated in phenomenological experiments.

This paper addresses the key issue of hydro mechanical behaviour of compacted buffer in context of clay microstructure and its evolution under repository relevant loading paths and material heterogeneity. This paper also introduces a unique column experiment facility available at Ruhr Universität Bochum, Germany. The facility has been designed as a forerunner of field scale testing program to simulate the transient hydration process of compacted buffer as per German reference disposal concept.

2 Dual porosity models: theoretical background

The studies pertinent to clay microstructure reveal that as compacted clay samples have different structural levels, which can be scrutinized by its pore size distribution

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curve. The pore size distribution curve based on MIP data revealed about two or more overlapping but distinct continua [3, 8-10]. Later these continua or structural levels were termed as micro and macrostructure. Clay microstructure level comprises the aggregation of clay particles or quasi-crystals. Thus the intra aggregate pores include inter layers and inter particles pores. Where as the pores in between aggregates are inter aggregate or macropores. A typical PSD curve for compacted bentonite is shown in Figure 1.

In dual porosity model the overall medium is assumed to have two interacting sub domain named as micro (m) and macropores (M). These sub domains are two totally distinct environments due to associated physicochemcial forces, hence show peculiar response and sensitivity to applied boundary conditions. For an example, the change in macroporosity is dominant during compaction, whereas the change in microporosity is dominant during hydration (Figure 1). The measurable volume change on macrolevel during clay water interaction is the upscaled microstructure response. Therefore, the material behavior during hydration is governed by initial heterogeneity in terms of micro to macroporosity fraction, evolution of microstructure under applied loading paths and its interaction with macrostructure. The micro to macroporosity fraction is attributed by various factores like initial physical state (powder/pellets), initial water content and dry density, fine content for clay based mixtures, sample preparation method, soil deformation history in term of wetting and drying cycles, clay mineralogy, geological (host rock convergence) and construction features (technical voids).

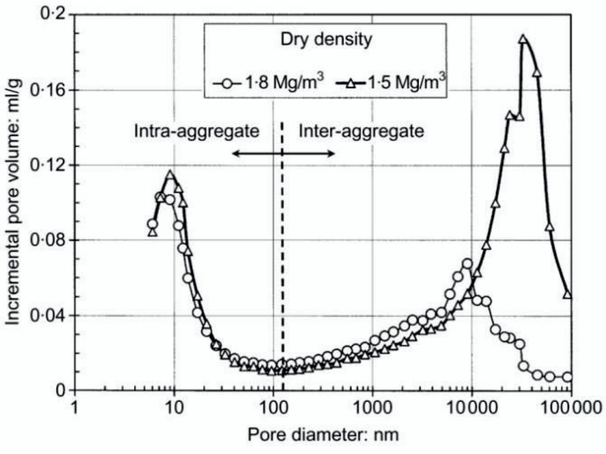


Figure 1. Distribution of incremental pore volume for two compacted bentonite samples at different dry densities. Mercury intrusion porosimetry test [9].

The microstructure after compaction depends on initial water content for a given dry density. The aggregates form, while mixing the clay powder with water and the aggregates themselves associate to form assemblies of aggregates. The aggregates are more prone to deformation of the wet side of optimum, thus filling the larger inter-aggregate voids and creating a single class of pores. Contrary, the association of aggregates remains more stable at the dry side of optimum water content, thus, the larger inter-aggregate pores exist additionally to the pores inside the aggregate. Figure 2 shows the PSDs of as compacted sample and saturated sample having same void ratio [12]. Distinct bimodal PSD features can be seen for as compacted sample at optimum moisture content, where as a weak bimodal PSD features are evident in PSD of saturated sample. In other words, the PSD is leading from a heterogeneous PSD to a more homogeneous one. Considering the initial physical state of material, the PSD of bentonite pellets mixed with powdered bentonite is shown in Figure 3. The other influencing factors for initial micro to macroporosity fraction are not discussed here in detail, as the paper mainly addresses the evolution of microstructure under transient boundary conditions.

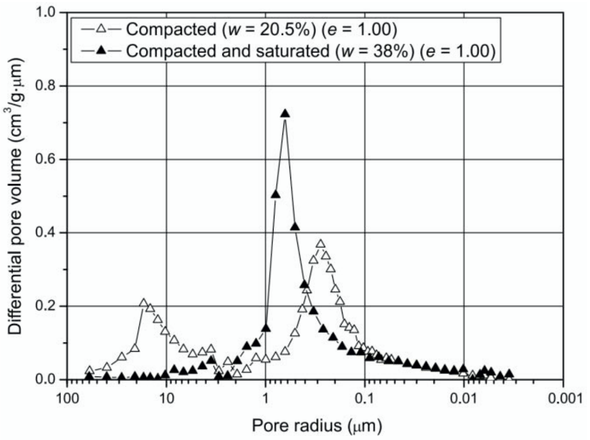


Figure 2. Comparison of PSDs of saturated compacted sample and samples compacted at optimum water content having same void ratio [11].

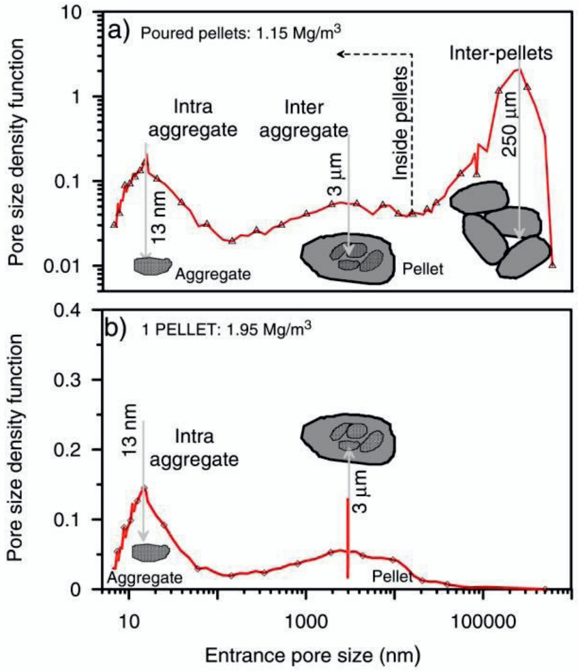


Figure 3. Pores size distribution of the bentonite pellets (a) Sampled poured into the sample holder. (b) High density pellets [12].

3 Microstructure evolution

Within the frame of coupled THM processes at nuclear waste repository, the augmentation of microporosity and its transformation as macroscopic swelling potential are highly senssitive to locally evolved mechanical constraints / transient boundary conditions and controlled by initial heterogeneity (e_m/e_M) and applied global boundary conditions. During hydration, the initial heterogeneity converts into homogeneity, the process can be portraied as a transient shift of one continua (microporosity e_m) towards the other continua (macroporosity e_M). The rate of this shift depends on the applied boundary conditions. Considerable research works have been reported to address the issue of volume constraints on the material response either in from of swelling pressure or in from of water retention behaviour [5, 13-23].

Pusch and Yong (2003) distinguished three boundary conditions that are believed to cause different rates of microstrucure evolution, such as clay exposed to water vapour, non-pressurized liquid water and pressurized water. For the clay hydrated with water vapour, the water molecules migrate into the open channels and adsorbed on exposed mineral surface. The water molecules then migrate into the elementary sheets that have higher hydration potential. As a result, swelling occurs uniformly for all the clay particles in an aggregate.

For the clay exposed to non-pressurized liquid water, water is absorbed by capillary forces in the open channels or macro pores. Then the water migrates into finer void and further into the elementary sheets. This results in expanding of clay particles near the boundary of the aggregate and closes the open channels or macro pores. Thus in this case, the hydration process is then controlled by diffusion process in its later stages.

For the clay exposed to pressurized water, water is pressed into the large channels and move quickly. The penetrating water displaces air and compresses the unsaturated matrix. The hydration rate is faster than those of first and second conditions (i.e., water exposed to vapor and non pressurized water). However, when the large channels become closed by the expansion of the clay aggregates, the hydration process follows the same procedure as the second procedure. This occurs in case of the heavily compacted bentonite. Whereas in case of compacted bentonite with available technical voids, all the large channels surrounding the compacted bricks are initially filled with water following the Darcy's advective flow regime and later bentonite bricks get hydrated through these water filled channels. With the progressive hydration process, these wider channels are filled due to expansion of compacted bentonite bricks due to swelling.

Agus and Schanz (2005) determine the PSD of the compacted bentonite sand mixture at three different states; namely, as-prepared, oven-dried, and swollen states under free swell condition. The PSD curve of the as-compacted, oven-dried, and swollen specimens was reported as bimodal indicating the presence of micro- and macropores in the specimens (Figure 4). Clay clusters enlarged and separated each other without being disintegrated when the specimen was wetted under unconfined conditions. During unconfined wetting, water was absorbed in the micro- and macro-pores which led to an increase in the volume of both pores. The similar sensitivity of the intra-aggregate pores to hydraulic and mechanical paths was exemplified by Romero et al. (2011) in Figure 5. The as-compacted sample is characterized by a clear bimodal distribution with dominant pore modes around 13 µm (macropores) and 60 nm (micropores). Loading the sample at vertical stress of 0.6 MPa shifts the size of dominant macropores towards a slightly lower value, while the micropore volume and its dominant pore size are not affected. On the contrary, the two saturation paths affect both inter-aggregate samples display a similar fabric dominated by the single peak at around 1.2 µm, which is originated by both expansion of the micropores and reduction of macropores.

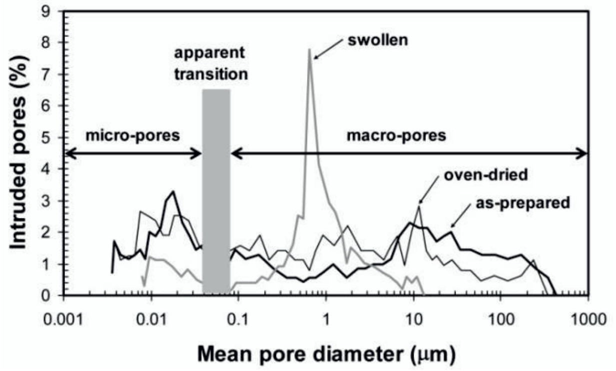


Figure 4. Intruded pore volume versus mean pore diameter plotted from the MIP data [15].

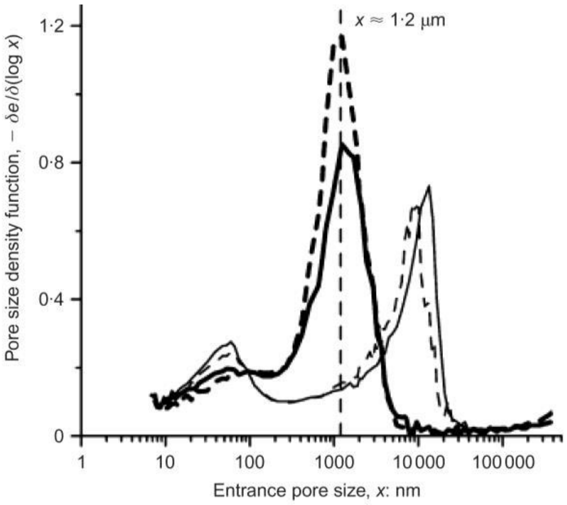
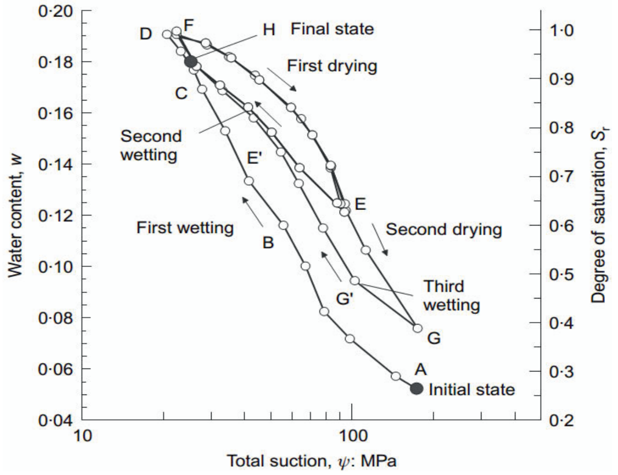


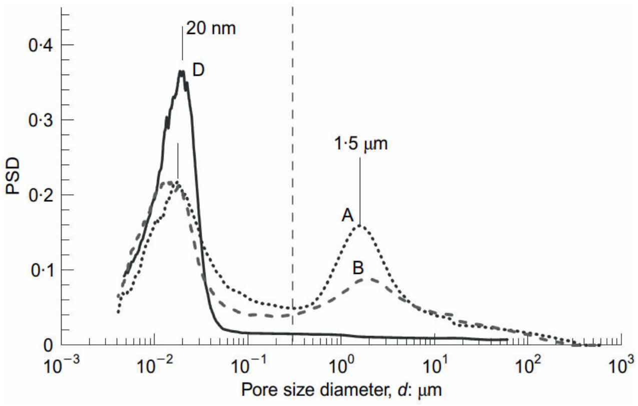
Figure 5. PSD of compacted Boom clay subjected to different hydro mechanical paths, as compacted sample (continuous line), vertical loading (dash line), constant volume (bold cont. line) and free swell condition (bold dash line) [23].

Considering the effect of different hydraulic path in terms of wetting and drying cycles, Seiphoori et al. (2014) observed an irreversible modification of pore size distribution curve, when the material approached a fully saturated state during the first wetting, and a new hydraulic domain was consequently created (Figure 6). The samples E and B show completely distinct PSD, although having the same degree of saturation, whereas the sample D, E and G show similar PSD having different degree of saturation. This permanent structure modification seems to occur after completion of first wetting path (D) and will remains permanent during the subsequent wetting and drying cycles. Considering the material in as compacted state with bimodal PSD and the state after first wetting with uni-modal PSD will modify the water retention capacity.

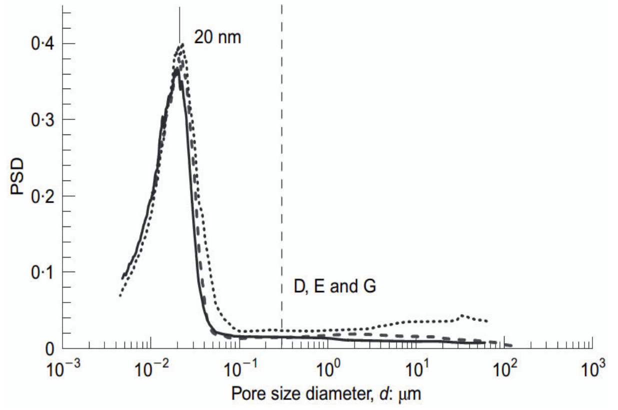
The changes in PSD domains in terms of micro to macroporosity fraction are highly sensitive to the involved physico-chemical processes as a function of applied loading paths and sequences, especially in the frame of couple THM processes at nuclear waste repository. Hence the phenomenological experiments provide additional key inputs related to understand the complex material behaviour.



(a) Water retention behaviour and cyclic variation in total suction.



(b) PSD of as compacted sample (A), sample with higher degree of saturation (B) and at saturation stage (D).



(c) PSD of fully saturated sample (D), end of first drying (E) and end of second drying (G).

Figure 6. Microstructure evolution for compacted MX-80 granular bentonite during different phases of wetting and drying [21].

4 Column experiment setup at RUB

As compared to field scale tests, the small scale phenomenological tests are more economical and have precise control over applied boundary conditions. Column type experiment devices have been extensively used for such proto type tests [24-30]. In column experimental studies, the transient hydration process in compacted buffer is simulated under repository relevant loading paths and the material response is observed by monitoring the key THM parameters like temperature, relative humidity and swelling pressure etc.

A unique column experiment facility is designed at Ruhr Universität Bochum as a forerunner of field scale test. The device is unique in terms of having proficiency to capture the transient material response under various possible repository relevant loading paths with higher precision level by monitor the key parameters like temperature, soil total suction, water content and axial & radial swelling pressure at three different sections along the total length of 30 cm. In general, a larger spectrum of loading paths / scenarios, which may arise in the nuclear repository, can be covered precisely with this devise.

4.1 Description of Column Testing Device

The entire device (Figure 7) can be assembled with five main components namely, (a) boundary control units (top & bottom), (b) PTFE rings for sample (middle), (c) monitoring sensors, (d) outer confining cell, and (e) rigid frame structure.

4.1.1 Top and bottom boundary control units

These units are encapsulated in 10 cm thick PTFE rings to ensure the one dimensional heat flow along the length of sample. Temperature is controlled by external units for heating and cooling separately. Silicon oil is used as a heat transport medium, which is pumped continuously in a closed circuit from external control unit to a small stainless steel (SS) chamber located at top and bottom of soil sample. For the hydration, water is supplied externally via. a SS pipe and SS porous stone, this porous stone is kept in contact with soil sample. The chamber, pipe and porous stone are designed and installed as a single unit to insure the thermal equilibrium in between the externally supplied water and the heating or cooling chambers.

The target temperature for heating (maximum 80°C) and cooling (10°C) can be controlled with precise rate using external units. The temperature of circulating Silicon oil is monitored constantly and adjusted according to target temperature. Both heating and cooling control units have three Pt100 sensors to perform the above task, which are installed in porous stone, inside the chamber and in Silicon oil reservoirs of external control units. The overall dimensions of top and bottom boundary control units are 35 cm as outer diameter and 20 cm of height.

4.1.2 PTFE rings for sample (middle section)

The middle section consists 03 PTFE rings having thermal conductivity as low as 0.23 W/mK. The rings have inner diameter as 15 cm, outer diameter as 35 cm and height as 10 cm. Each compacted clay sample (total

03) has the identical dimensions (diameter 15 cm and height 10cm) to fit into corresponding PTFE ring. The individual sample is compacted under uni-axial vertical pressure of 20-30 MPa in specially designed compaction mould with target initial dry density in the range of 1.8 to 2 g/cc for 50:50 bentonite sand mixture with 8-9% initial water content.

After compaction, the mould having compacted sample is placed over the bottom most PTFE ring. The sample is extruded from mould and pushed directly into the ring using a hydraulic jack. The same procedure is followed to push other two samples (middle and top) over each other, the provision of peripheral O-ring in between these rings ensure the air and water tight joint. The tightening screws made up by PTFE are further deployed to make the proper connection in between rings. Once all the sample rings are stacked over each other, the compacted soil sample has overall height of 30cm with 15 cm diameter. The top boundary control unit is placed over the stacked sample rings with O-ring and PTFE tightening screws. After assembling the sample rings in between top and bottom boundary control units, the confining structure is placed around the assembled column testing units to make sure constant volume condition. Finally the monitoring sensors are installed in each sample ring.

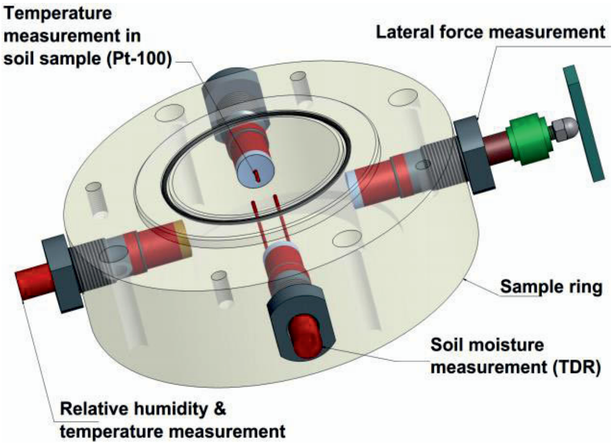
4.1.3 Monitoring sensors

To monitor the transient material behavior, each sample ring is having four sensor installation points perpendicular to each other at the same horizontal level as shown in Figure 8a. Thus we have individual sensor to monitor four key parameters as temperature, relative humidity, water content and radial swelling pressure at the same level. Each sensor is installed with its specially designed PTFE adaptor. These PTFE adaptors with O-rings are designed to ensure the air and water tight joints with minimum disturbance to sample condition. The detailed description of individual sensor is not discussed here. Additionally, the axial swelling pressure will be measured at top and bottom using load cells.

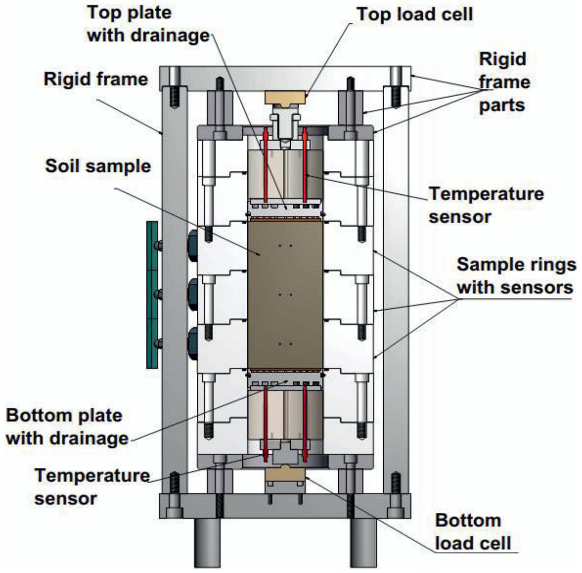
To capture the effect of transient boundary conditions during hydration, the swelling pressure will be measured by total pressure transducers in between two sample interfaces. These total pressure transducers will be embedded in the center of compacted sample between first and second; second and third sample rings. To get the transient profile of total suction along the sample length with Kelvin's equation using transient RH profiles and temperature profiles, one must be ensure about the proper thermal equilibrium between location of measuring temperature and relative humidity. The schematic view of soil sample ring with sensor installation locations are shown in Figure 8.

5 Discussion

Transient hydration process of compacted buffer material under coupled THM processes at nuclear waste repository indicates the transformation of its initial heterogeneity into homogeneity as final state. This process is identical to the process of getting more stable lower energy state by dissipating the initial high energy. In transient hydration process, the initial higher energy can be correlated with initial higher total suction, which reduces slowly with elapsed time.



(a) PTFE sample ring with installed sensors.



(b) Overall schematic view

Figure 7. Schematic view of column testing device at RUB, (a) individual PTFE ring, and (b) overall column device.

The micro structure evolution drives the homogenization process, which involves the shift of initial bi-modal pore size distribution into uni-modal nature. The changes in PSD domains in terms of micro to macroporosity fraction are highly sensitive to the involved physico-chemical processes as a function of applied loading paths, especially in the frame of couple THM processes at nuclear waste repository. Hence the phenomenological experiments provide additional key inputs related to understand the complex soil water retention and volume change behaviour. The SWRC perform in laboratory under unconfined condition using axis translation and vapour equilibrium technique are not directly applicable for phenomenological investigations due to difference initial soil state, different fabric arrangement and transient boundary conditions. Similarly the swelling pressure test results under steady state boundary conditions like constant volume or free swell conditions will differ from the swelling pressure under transient boundary conditions in small scale phenomenological investigations. This ambiguity in material behaviour under transient boundary conditions is a major challenge for researchers, which can be removed by defining a suitable state parameter, which essentially describe the path dependent microstructure evolution and its interaction with macrostructure.

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