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Swelling behavior of unsaturated claystone/ bentonite mixtures

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Abstract. This laboratory experimental program investigated the impact of variations in the expansive mineral content, the grain size distribution of employed bentonite, the initial dry density and the selected hydration path on the water retention characteristics and swelling properties of processed Callovo-Oxfordian claystone and its mixtures with MX80 bentonite. The French reference concept for the disposal of nuclear waste in deep sedimentary rock formations envisages the reemployment of excavated material as backfill material, which is installed in situ by means of conventional compaction techniques. The investigations were of special interest as the major issues involving in situ compacted backfill materials were portrayed. Experiments showed that the impact of variations in the dry density on the water retention characteristics of all materials vanished as suctions exceeded 100 MPa. The negligible impact of variations in the initial dry density on the collapse behavior of claystone/ bentonite mixtures remained questionable.

1 Introduction

The French reference concept considers the disposal of intermediate- and high-level nuclear waste in the deep sedimentary Callovo-Oxfordian rock formation, henceforth referred to as Callovo-Oxfordian claystone (COX-claystone). The nuclear waste is encapsulated in canisters, which are then emplaced in drifts being excavated in the host rock formation [1]. Once the operational phase of the repository is terminated, the drifts and shafts are sealed and backfilled. It is planned to process COX-claystone, which is obtained during repository construction, and to reemploy it as backfill material. Henceforth, crushed and sieved COX-claystone is referred to as COX_c. It is installed in unsaturated conditions and swells upon the wetting front originating in the rock formation intrudes into it. The propagation of the excavation damaged/ disturbed zone (EDZ) is limited and hydraulic conductive voids are closed by material swelling under the constant volume conditions. The hydro-mechanical behavior of materials containing expansive mineral phases, such as COX_c, is generally affected by the environmental conditions and the material properties, especially by the initial dry density. Its manipulation can result in the conformance to defined safety requirements in terms of swelling pressure and hydraulic conductivity.

Conventional compaction techniques, such as vibrating plates, are envisaged to be employed to install the backfill material in situ. Backfill material is thus prepared at the optimum compaction conditions obtained from modified Proctor tests. Difficulties in positioning the machines in situ are expected to cause variations in the compaction energy, which provoke, in turn, an inhomogeneous distribution of dry densities in the cross section of the excavation [2].

Most of the previous studies performed experiments in constant volume conditions to determine the evolution of the swelling pressure of different materials containing expansive mineral phases upon continuous wetting. Many of those also comprised investigations on the impact of variations in the dry density on the maximum swelling pressure. Results revealed that the maximum swelling pressure exponentially increases with increasing initial dry density. Besides, the kinetics of the evolution of the swelling pressure is similar, regardless of the initial dry density.

However, the approach of continuous wetting simplifies the phenomenon of the wetting front. In situ experiments indicated that the material installed in the drift center is likely to remain unsaturated even several hundreds of years after repository closure [2]. Thus, the hydro-mechanical behavior of the backfill is predominantly affected by the reduction of suction. The impact of step-wise wetting on the evolution of the swelling pressure of potential backfill materials in constant volume conditions was hardly investigated [3–5]. Moreover, the transferability of the findings of those studies is questionable as backfill materials compacted in situ are initially characterized by elevated degrees of saturation ($S > 0.8$) and low suctions ($s < 6$ MPa). For instance, the multi-step suction-controlled swelling pressure experiment performed by Lloret et al. [3] included the direct saturation of samples once the swelling pressure stabilized at an imposed suction of 12 MPa. However, such a methodology is not applicable to investigate the kinetics of the swelling pressure of backfill materials compacted in situ. A technique was thus required, which is capable to precisely impose suctions in the range between 12 MPa and quasi-saturation.

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This study investigated the hydro-mechanical behavior of claystone/ bentonite mixtures. The partial replacement of COX_c by a material containing a considerably elevated amount of expansive mineral phases was also aimed at generally enhancing the swelling properties of the material. Previous studies revealed a further enhancement of the mixture performance when reducing the maximum grain size of the bentonite fraction. The impact of the grain size distribution on the water retention characteristics and the unsaturated swelling properties was thus assessed by considering a grain- and powder-mixture. Samples of both mixtures were then compacted to the maximum and reduced dry densities at the optimum water content to respond to the question how variations in the dry density affect the water retention characteristics and the unsaturated swelling properties of potential backfill materials. The osmotic technique was adopted to wet the samples stepwise from their initial suctions to quasi-saturation. The combined impact of dry density and wetting front on the kinetics of the evolving swelling pressures in constant-volume conditions was sufficiently precisely portrayed by this means.

2 Materials and methods

Section 2.1 describes the characteristics of the investigated claystone/ bentonite mixtures with special focus on their water retention characteristics. Information about the employed device for determining the unsaturated swelling pressure in constant volume conditions are given in section 2.2.

2.1 Materials

Callovo-Oxfordian claystone was obtained while excavating drifts of ANDRA URL in Meuse/ Haute-Marne region (France). Excavated drifts are located at a depth of - 500 m. The material was crushed to maximum grain diameters of 2.00 mm a few weeks after the excavation and stored in an air-tight container.

The impact of grain size distribution on mixture behavior was studied by crushing a sodium-bentonite (MX80 bentonite, Wyoming) to maximum grain diameters of 0.28 mm and 2.00 mm. Henceforth, mixtures being composed of COX_c and MX80 bentonite with a maximum grain size of 0.28 mm and 2.00 mm are referred to as powder-mixture and grain-mixture, respectively. The considered mixing ratio was 70% to 30%. Values of physical and compaction properties were taken from [6]. They are compiled in Table 1. Compared to those of COX_c and the grain-mixture, the grain size distribution curve of the powder mixture was generally shifted upwards, as the fraction of fines was significantly increased by replacing COX_c by bentonite powder [6].

Water retention curves were determined by combining the osmotic and vapor equilibrium technique both being extensively employed in the last decades [7,8]. The former technique bases upon the exchange of water molecules through a semipermeable membrane, which separates the pore solution in the samples from a macromolecular solution. The exchange is then driven by a concentration gradient. The latter technique adopts Kelvins equation to relate suction and relative humidity in a closed system. Detailed information about the osmotic and vapor equilibrium technique are given in [7]. Their combined employment allowed the coverage of suctions ranging from quasi-saturated state to several hundred MPa.

Table 1: Physical and compaction properties of COX_c, the grain- and powder-mixture

		COX_c	Grain-mix	Powder-mix
Bentonite fraction	[%]	-	30	30
Max. bentonite GZ$^+$	[mm]	-	2.00	0.28
Initial water content	[%]	5.4	6.4	6.4
Specific gravity	[-]	2.68	2.64	2.64
Liquid limit	[%]	37.5	112.5	112.5
Plastic limit	[%]	24.9	34.7	34.7
Opt. water content*	[%]	13.2	18.2	15.0
Max. dry density*	[Mg/m^3]	1.95	1.72	1.79
+: Maximum bentonite grain size *: obtained in modified Proctor tests				

Samples of grain- and powder-mixture were compacted to the maximum and a reduced dry density at optimum water content, while the samples of COX_c were exclusively compacted to optimum values. This approach emphasized the impact of variations in the dry densities on the water retention characteristics.

The water retention characteristics of materials are depicted in **Fig. 1**. The replacement of COX_c by bentonite caused a significant increase of initial suction (s_{ini}) from 1.5 MPa in the case of COX_c to 4.5 MPa and 5.9 MPa in the cases of the grain- and powder-mixture, respectively. Its increase was attributed to the higher amount of expansive mineral phases. Compared to the grain-mixture, the powder-mixture exhibited an elevated initial suction most likely caused by the slightly lower water content.

The impact of initial dry density on the water retention characteristics generally vanished in ranges of suctions, which were greater than 100 MPa, regardless of the mineralogy and grain size distribution. This finding was in accordance with the literature [3,9] . The negligible impact of the grain size distribution was attributed to the fact that the mineralogy of samples essentially remained the same.

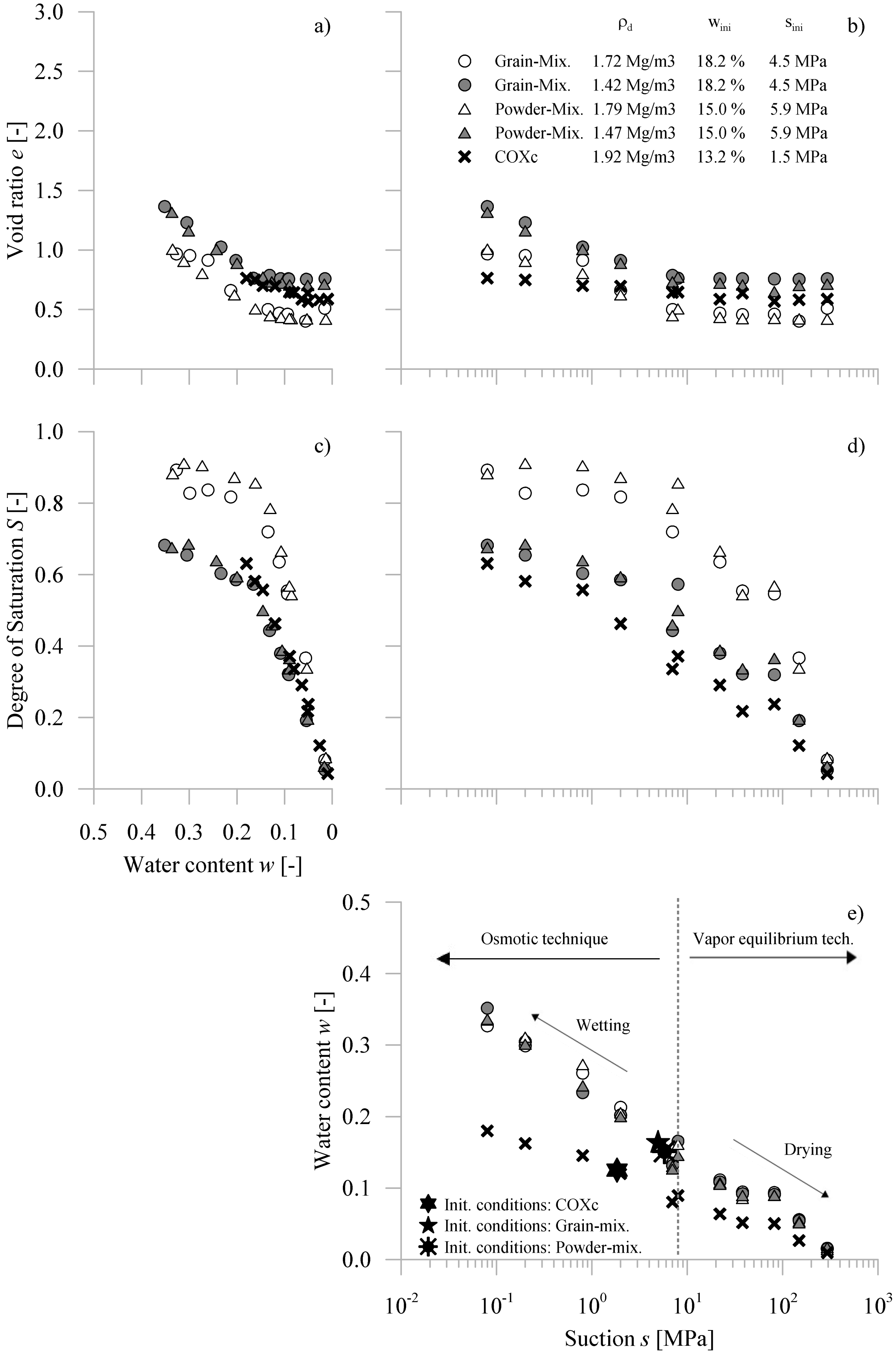

Fig. 1: Obtained water retention characteristics of COX$_c$, the grain- and powder-mixture (Experiments comprised grain- and powder-mixtures compacted to maximum and reduced dry densities at optimum water content

As suction decreased, samples compacted to the reduced dry density exhibited a higher free swell potential than COX_c and those samples being compacted to the maximum dry density (**Fig. 1** a and b). Water can penetrate the material more easily due to their higher macro-porosity. The lower macro-porosity might also cause the higher air-entry values of denser mixtures (**Fig. 1** d).

The looser mixtures and the pure material attained degrees of saturation of less than 0.7. Thus, the water uptake predominantly proceeds at very low suctions in those cases. Experiments including suctions being smaller than 0.08 MPa can complement the water retention curves.

The most important conclusion is that backfill material, which was looser compacted, considerably saturates once suctions were smaller than 0.08 MPa. Conversely, dense samples attained degrees of saturation of higher than 0.9 at a similar suction. Apart from the impact of initial dry density, the impact of the grain size distribution on the water retention characteristics of tested materials was negligible.

2.2 Experiment setup

The employed device adopted the osmotic technique to impose suctions ranging from 0 to 8 MPa. The evolution of the swelling pressure was then measured under constant-volume conditions. More information about the design of the employed cell are given in [5].

A peristaltic pump was used to circulate the macromolecular (Polyethylene-glycol (PEG)) solution through the system. Their reservoirs were positioned in a temperature-controlled water bath, as suctions were expected to fluctuate due to temperature variations. The employed cell was composed of three main parts, namely the bottom plate, the sample ring and the lid. The bottom plate comprised the fluid in- and outflow, the porous disc being made up of stainless steel, and a joint preventing the cell from leakage. The load censor was installed in the lid. Once compacting the sample directly in the sample ring, the bottom plate was bolted to the sample ring. The semi-permeable membrane was positioned in their interspace before. The height to diameter-ratio of samples was about 0.28 in all tests. The assembling was completed by bolting the lid to the sample ring, ensuring the contact of the load sensor with the sample. Evolving pressures were recorded by the data acquisition system. The experiments proceeded as follows: The macromolecular solution enters the cell from the bottom upwards the cell, passes through the porous disc and circulates beneath the semipermeable membrane, which is in contact with the sample. The circulated macro-molecular solution eventually leaves the cell through the outflow.

3 Multi-step Suction-controlled constant-volume swelling pressure experiments

The following subsection describe the adopted experimental approach. The results of the multi-step suction-controlled swelling pressure experiments are presented and discussed in the last subsection.

3.1 Approach

The developed approach of multi-step constant-volume swelling pressure experiments assessed the impact of variations in the dry density on the evolution of the swelling pressure of the grain- and powder-mixtures under stepwise wetting. Imposed suctions ranged from the individual initial suction to quasi-saturated state. The experiment program comprised samples, which were compacted to the maximum dry density at optimum water content. Both values were previously determined in modified Proctor tests. Samples compacted to differently reduced dry densities at optimum water content were additionally considered to emphasize the impact of variations in the dry density (Table 2).

Since the grain- and powder-mixtures were characterized by different initial suction, the normalized suction (s_n) was thus defined as the ratio of imposed suction to initial suction. This approach facilitated the comparison of the swelling pressures. Normalized suctions were decreased in three steps from 60%, to 6% and to 0.6%. The value of 60% was selected to ensure the wetting of samples. According to the considered criteria, suctions were decreased, when the recorded swelling pressure changed less than 10 kPa/day. Swelling pressures were assumed to be stabilized in that case.

Table 2: Characteristics of samples employed in multi-step suction-controlled constant-volume swelling pressure experiments

		Grain-mix	Powder-mix
Maximum bentonite grain size	[mm]	2.00	0.28
Normalized suction path*	[-]	0.6 – 0.06 – 0.006	
Initial dry density	[Mg/m^3]	1.42 – 1.72	1.46 – 1.79
Initial water content	[%]	18.2	15.0
Initial void ratio	[-]	0.54 – 0.88	0.56 – 0.92
Initial degree of saturation	[%]	56.0 – 92.0	46.0 – 80.0
*: Ratio of imposed suction to initial suction of materials			

3.2 Results and discussion

The major results of multi-step suction-controlled swelling pressure experiments, which were performed at the grain- and powder-mixture, are depicted in **Error! Reference source not found.** and **Error! Reference source not found.**. Henceforth, the terms “global” and “local” refer to the complete experiment procedure and to the individual suction stages, respectively.

The maximum swelling pressures of all samples generally increased with increasing their initial dry density.

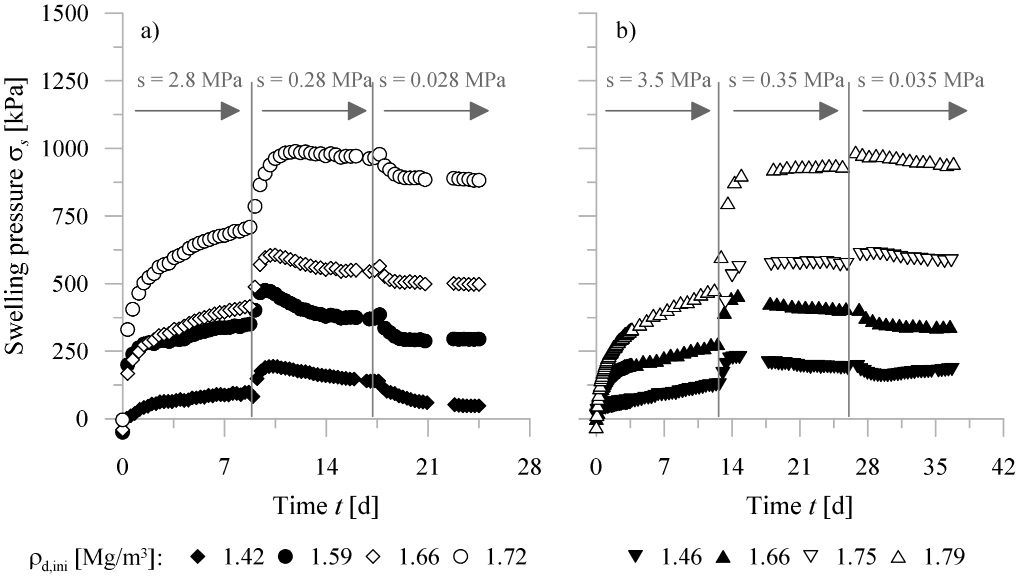

Fig. 2: Evolution of swelling pressure of the grain- and powder-mixture as function of applied suction

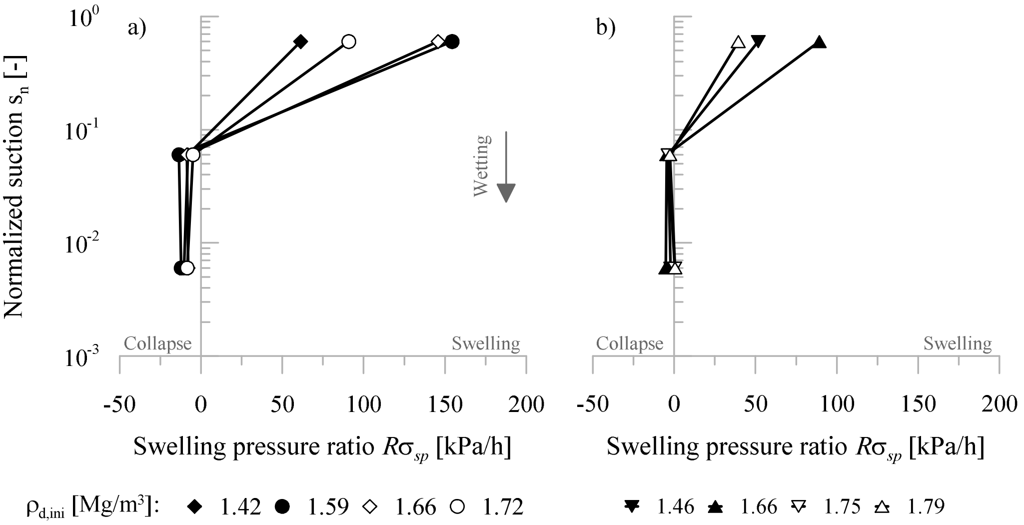

Fig. 3: Calculated swelling pressure ratios of the grain- and powder-mixture in the individual steps of applied suction

Information about the swelling pressure of the powder-mixture compacted to an initial dry density of 1.75 Mg/m³ in the first stage were not available. As depicted in **Error! Reference source not found.** a and b, the global maximum swelling pressures were slightly higher than 1 000 kPa when compacting both materials to their individual maximum dry densities at the optimum water content. The grain- and powder-mixtures attained these maximum values in the beginning of the second and the third stage of wetting, respectively. Most of the samples exhibited a differently pronounced increase of the swelling pressure upon wetting. Especially in the second and third stage of wetting, the evolution of the swelling pressure of the grain-mixture was characterized by its stabilization at lower values after attaining the local maximum swelling pressures. Swelling pressures then decreased in the proceeding of the second and third stage. More information about the increase and decrease of the swelling pressure in each stage can be obtained by considering the time-dependent swelling pressure ratio $R\sigma_{sp}$ (kPa/h):

$$R\sigma_{sp} = \frac{\sigma_{s,max,global} - \sigma_{s,fin.local}}{t_{\sigma_{s,max.global}} - t_{\sigma_{s,fin.local}}} \tag{1}$$

It can be defined as the ratio of the difference between the global maximum swelling pressure ($\sigma_{s,\ max,\ global}$) and the local final swelling pressures ($\sigma_{s,\ fin.\ local}$) to the difference of the times when the global maximum and the local final swelling pressure were recorded. The swelling pressure ratio becomes positive when the swelling pressure increases, whereas a reduction of the swelling pressure is indicated by negative values. As it is depicted in **Error! Reference source not found.** a and b, the powder mixtures tended less to collapse upon passing the global maximum swelling pressure than the grain mixtures. This behavior was indicated by the swelling pressure ratios of the powder mixtures being almost equal to zero. Interestingly, the collapse behavior of both mixtures was hardly affected by the initial dry density of the mixtures.

The observed behavior might be generally attributable to the structural rearrangement in the clay particles and aggregates upon swelling [5,10]. The first stage of wetting resulted in the hydration of exchangeable cations in the interlayer space of clay particles. The consequent expansion of the clay particles in conjunction with the preservation of macro-pores initiated the evolution of the swelling pressure in constant volume conditions. Global maximum swelling pressures can be related to the maximum expansion of clay particles while wetting, and to the load limit of the assembly of clay aggregates at that stage. The decrease of the swelling pressure ensued when macro-pores partially collapsed, and clay particles rearranged in denser particle packing. Stabilized swelling pressures might then indicate the equilibration of micro-structural expansion and macro-structural rearrangement. As mentioned before, this approach of interpretation is generalized and shows limitations with regard to the impact of variations in the dry density. Samples were expected to rearrange more at macro-scale as they were characterized by a higher initial macro-porosity. However, the reduction of the swelling pressure ratios remained essentially comparable, regardless of the initial dry density of the samples.

4 Conclusions

This laboratory experimental program investigated the impact of variations in the expansive mineral content, the grain size distribution, the initial dry density and the suction on the water retention characteristics and the evolving swelling pressures of two different claystone/ bentonite mixtures. Those mixtures are planned to serve as backfill materials in nuclear waste repositories upon installing them directly inside the drifts. The study was of major interest, as information about the combined impact of variations in the dry density and the suction on the swelling pressure of expansive materials were scarcely available. Moreover, there were no studies, which investigated the evolution of the swelling pressure in ranges of suctions between 12 MPa and quasi-saturation.

Generally, the individual impact of the grain size distribution on the water retention characteristics of mixtures was negligible, since both mixtures reacted similarly to variations in the dry density and the suction. The individual impact of variations in dry density on the water retention characteristics of tested materials vanished once suctions exceeded 100 kPa. This finding was in good agreement with the literature. The major water uptake of loose samples proceeded when suctions were less than 0.08 MPa. Multi-step suction-controlled swelling pressure experiments showed that the global maximum swelling pressure generally increased with increasing the initial dry density of samples. The global maximum swelling pressure represented a non-equilibrated stress state, and the swelling pressures finally stabilized at lower values while keeping the imposed suction constant. This observation was generally attributed to the partial collapse of macro-porosity and particle rearrangement. Interestingly, the swelling pressure ratios remained comparable, regardless of the grain size distribution and initial dry density of the samples. The laboratory experimental program thus envisages to complement further multi-step swelling pressure experiments by conducting microstructural analysis (e.g. mercury intrusion porosimetry (MIP)). Their results are expected to improve the interpretation of the swelling pressure experiments and to explain the minor impact of the initial dry density on the collapse behavior.

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