

Article



Swelling Pressure and Permeability of Compacted Bentonite from 10th Khutor Deposit (Russia)

Artur Yu. Meleshyn¹, Sergey V. Zakusin^{2,3} and Victoria V. Krupskaya^{2,4,*}

- ¹ Gesellschaft für Anlagen- und Reaktorsicherheit (GRS) gGmbH, 38122 Braunschweig, Germany; artur.meleshyn@grs.de
- ² Institute of Ore Geology, Petrography, Mineralogy and Geochemistry, Russian Academy of Science (IGEM RAS), 119017 Moscow, Russia; zakusinsergey@gmail.com
- ³ Department of Engineering and Environmental Geology, Lomonosov Moscow State University, 119991 Moscow, Russia
- ⁴ Nuclear Safety Institute, Russian Academy of Science (IBRAE RAS), 115191 Moscow, Russia
- * Correspondence: krupskaya@ruclay.com; Tel.: +007-926-819-6398

Abstract: Bentonites from the 10th Khutor deposit (Republic of Khakassia, Russia) are considered a potential buffer material for isolation of radioactive waste in the crystalline rocks of Yeniseyskiy site (Krasnoyarskiy region). This study presents the results of a series of permeameter experiments with bentonite compacted to dry densities of 1.4, 1.6, and 1.8 g/cm³, saturated and permeated by the artificial groundwater from Yeniseyskiy Site. Permeation was conducted at hydraulic gradients of 180–80,000 m/m to simulate potential hydraulic conditions in the early post-closure phase of a deep geological repository (DGR). The respective swelling pressures of 0.8 ± 0.3, 2.2 ± 0.6, and 6.3 ± 0.3 MPa and permeabilities of (27 ± 15) × 10⁻²⁰, (3.4 ± 0.8) × 10⁻²⁰, and (0.96 ± 0.26) × 10⁻²⁰ m² were observed for the hydraulic gradient of 2000 m/m, which is recommended for the determination of undisturbed swelling pressures and permeabilities in permeameter experiments. Upon incremental increases in the hydraulic gradient, swelling pressures at all densities and permeability at the density of 1.8 g/cm³ remained unchanged, whereas permeabilities at 1.4 and 1.6 g/cm³ decreased overall by a factor of approximately 5 and 1.7, respectively. Seepage-induced consolidation and/or reorganisation of bentonite microstructure are considered possible reasons for these decreases.

Keywords: radioactive waste; bentonite; swelling pressure; permeability; hydraulic gradient; engineered barriers; geological repository

1. Introduction

In the Russian Federation, a concept is being developed to isolate class 1 and 2 high-level radioactive waste and spent nuclear fuel in crystalline rocks (Yeniseyskiy Site, Krasnoyarskiy region) at depths of about 500–700 m [1–4]. In this case, the safety function of radioactive waste disposal is provided by a passive multi-barrier system that ensures the isolation of radioactive waste from the biosphere [5–7].

The current Russian sealing concept of a deep geological radioactive waste disposal facility (DGR) in crystalline rocks foresees the use of compacted bentonite as a buffer material enclosing containers with radioactive waste [1–4] due to the stability of its properties and ability to prevent the mass transfer of radionuclides from advection and diffusion mechanisms [2–7]. The 10th Khutor bentonite deposit is located relatively close to the area of the deep geological disposal site and considered as a source for the potential buffer material in study [4] based on its favourable sealing properties and deposit resources. Swelling pressure and permeability (or proportional to that hydraulic conductivity) of compacted bentonite are key parameters in determining the sealing performance of bentonite-based geotechnical barriers. A direct determination of these parameters for a realistic buffer



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Copyright: © 2021 by the authors. Licensee MDPI, Basel, Switzerland. This article is an open access article distributed under the terms and conditions of the Creative Commons Attribution (CC BY) license (https:// creativecommons.org/licenses/by/ 4.0/). configuration under in situ conditions of DGR would require conducting sophisticated, long-term, and expensive experiments in an Underground Research Laboratory. Laboratory experiments with small specimens of compacted bentonite provide a feasible way to obtain estimates of the parameters necessary to conduct preliminary performance assessments of bentonite-based geotechnical barriers and facilitate the planning and optimisation in situ studies, possibly reducing their extent.

During the construction of geotechnical barriers, unsaturated bentonites will experience high fluid pressure differences—up to several MPa—which may result across the buffer in the course of establishing hydraulic conditions characteristic of the host formation. Therefore, fluid pressure differences of up to 8 MPa across the compacted specimens were imposed on the present study in order to test the sealing performance of a bentonite sample from the 10th Khutor deposit (described as "10X" or "10X bentonite"). Swelling pressure and hydraulic conductivity of 10X were estimated in a series of preliminary laboratory experiments with distilled water [4]. The present study provides an independent laboratory estimation of swelling pressure and permeability for 10X upon saturation and permeation with the synthetic groundwater of crystalline rocks at the Yeniseyskiy site.

2. Materials and Methods

2.1. Geological Site and Composition of 10X

The 10th Khutor deposit is located in the steppe area 8 km southwest of Chernogorsk city (Republic of Khakassia). It has reserves of about 3–4 million tons and belongs to a group of deposits with total reserves of approximately 13 million [8]. The deposit has a Carboniferous age (C_2) expressed in several bentonite layers interbedded with clay rocks and carbonaceous layers [8–10]. Its genesis is considered volcanogenic-sedimentary.

In the process of studying bentonite powder with specified technical conditions, obtained characteristics were required to justify the use of clay material as a component of engineering safety barriers. This material was assigned a commercial name,10X.

The study of particle size distribution was conducted using laser deflation on an Analysette-22 (Fritsch GmbH, Idar-Oberstein, Germany) particle analyser in suspension for the measurement range of 0.1–300 μ m in five different positions of the measuring cell and 100 measurements in each position. The results are shown in Table 1.

a—Particle Size Distribution												
Mods, µm		Me	Median, µm			Particle Size Distribution, % 0.1–1 μm 1–10 μm					:5 μm	
1.79			2.90			15.6		72.6			11.8	
b—Mineral Composition (%)												
Smectite (Montmorillonite)		e) Chlorite	Quartz		Feldspars (Microcline and Albite)		Calcite			Anatase	Gypsum	
73.0		0.9	13.9		10.8		0.7			0.3	0.4	
c—Chemical Composition												
Na2O	MgO	A12O3	SiO2	K2O	CaO	TiO2	Fe2O3	FeO	MnO	LOI (105	–1000 °C)	
0.107	2.67	18.84	51.87	0.48	6.58	0.70	3.37	0.33	0.01	7	.64	

Table 1. Basic characteristics of bentonite powder 10X.

The mineral composition of the 10X bentonite [10,11] was estimated by X-ray diffraction using an Ultima-IV (Rigaku, Tokyo, Japan) X-ray diffractometer (Cu-K_{α} radiation, detector—D/Tex-Ultra, scan range 3–65° 2 θ). The quantitative mineral analysis (Table 1) was conducted using the Rietveld method [12] with PROFEX GUI ver. 3.1.3, (Nicola Doeblin, Solothurn, Switzerland) software for BGMN [13].

The chemical composition (Table 1) was determined using X-ray fluorescence analysis using an Axios mAX (PANalytical, Almelo, the Netherlands) according to standard meth-

ods [11]. The hygroscopic water content of 8.94% was determined upon drying at 105 °C. The suction of the 10X bentonite was not determined.

The density of the solid phase was measured using the pycnometer method with kerosene following [14,15] and amounted to 2.8 g/cm^3 .

The value of the cation exchange capacity was determined from the adsorption of the Cu(II)-trien-complex using the spectrophotometric method [16–18] and amounted to 73.5 meq/100g. The composition of the exchangeable complex is mainly Na-Mg, and comprises Na—23.6, Ca—16.6, Mg—30.3, and K—1.2 meq/100g, measured with atomic absorption spectrometry (AAS) after repeated extraction with ammonium acetate solution.

2.2. Experiments and Conditions

For the research, triplicate experiments of 10X bentonite at the dry densities of 1.4, 1.6, and 1.8 g/cm³ were conducted using nine constant-head, rigid-wall permeameter cells made of titanium (Figure 1a). Compacted specimens were prepared by pressing directly into permeameter cells. Bentonite powder (Table 1a) was filled into a permeameter between two sintered steel filters (GKN SIKA-R 20 AX, a porosity of 43%, an average pore diameter cells—of $(3.4 \pm 0.5) \times 10^{-14} \text{ m}^2)$ and, after installing a load-transfer ram and a press flange above the top sintered filter, it was statically compacted (without adding any water) to a pellet specimen with a diameter of 5 cm, a height of 1 cm, and a dry density of 1.4, 1.6, and 1.8 g/cm³. The cap nut of the press flange was then tightened without adding axial stress. Based on the density of soil solids (2.80 g/cm³), porosity of specimens can be estimated to equal 0.50, 0.43, and 0.36, pore volumes to 9.8, 8.4, and 7.0 cm³, and saturation to 25%, 33%, and 45%, respectively. Respective bulk densities equal 2.02, 2.17, and 2.32 g/cm³.



Figure 1. A construction drawing of experiments; (**a**); supply of SGW to inlets at cell bases by a piston pump; (**b**) permeability experiments (note that burettes connected to outlets at cell tops to collect percolating solutions).

The swelling pressure of the specimens in the permeameter cells was measured continuously by force transducers FKA613 (Ahlborn Mess- und Regelungstechnik GmbH, Holzkirchen, Germany). It should be noted that for the specimen in the ninth cell (at 1.8 g/cm^3 , the leftmost cell in Figure 1b), a behaviour strongly deviating from the other two specimens at 1.8 g/cm^3 was observed, as its swelling pressure exceeded the measuring capacity of force transducers at 10.2 MPa within two days of starting the experiment. Two restarts and additional tests suggested a permeameter malfunction as the reason for this deviating behaviour; therefore, the experiment with the ninth cell was discontinued.

Since no reliable data currently exists on the composition of the groundwater of the Yeniseyskiy Site at the target depth, synthetic groundwater (hereafter referred to as SGW) was used in the experiments and developed based on available hydrogeological data and data in [19], which is characterized by a pH of 8.1, ionic strength of 6.5 mmol/L, and ionic composition: Na⁺—0.760, Mg²⁺—0.494, Ca²⁺—1.217, K⁺—0.115, Cl⁻—2.549, SO₄^{2–}—0.494, and HCO₃⁻—0.76 mmol/L.

Compacted bentonite specimens were allowed to saturate by absorbing SGW from burettes connected to inlets at cell bases to let trapped air escape through outlets at cell tops for 28 days, then to inlets at cell tops for 87 days to reach the most homogeneous saturation of bentonite possible (Figure 1b). The duration of 28 days for the first saturation stage was chosen based on a preceding study involving 15 different bentonites [20,21]. According to the experimental protocol established in that study, the second saturation stage was continued until the swelling pressure of the specimens was judged to level off. The fluid pressure difference across the specimens was equal to 0.018 MPa during this stage.

After disconnecting the burettes, permeation tests were started on the 133rd day (after a delay due to a malfunction and temporary unavailability of the pump) by injecting SGW through inlets at cell bases using a piston pump (BESTA HD 2-200, BESTA-Technik GmbH, Wilhelmsfeld, Germany) and a bypass (Figure 1b) to prevent uncontrolled fluid injection pressure surges observed in a preceding study discussed in [20,21]. Outlets at cell tops were connected to burettes for measuring volumes of percolating SGW used to calculate permeability. Swelling of compacted bentonites upon saturation was assumed to seal off possible flow pathways along the bentonite-cell interface with the solution percolating only through bentonite pores during permeability measurements, which was confirmed by the observed low permeabilities of specimens. The fluid pressure difference of 0.018 MPa maintained across the specimens for 132 days by the water column in the burettes and was then incrementally increased to 0.2, 0.5, 2.0, and 8.0 MPa using the pump. These fluid pressure differences correspond to hydraulic gradients of 180 up to 80,000 m/m across the compacted bentonite specimens. For the first stage of the permeation tests at a hydraulic gradient of 2000 m/m, levelling off of permeabilities while observing for the levelling off of swelling pressures, if reasonably possible, was used as a termination criterion. During this stage, the pore volume of specimens was replaced on average 13.7, 2.2, and 0.5 times at a dry density of 1.4, 1.6, and 1.8 g/cm³, respectively. For the following stages, a sufficient number of measurement points to capture the permeability trend was aimed, and the pore volume of specimens was replaced at a respective dry density of 1.4, 1.6, and 1.8 g/cm^3 on average 0.9, 0.1, and 0.02 times at a hydraulic gradient of 5000 m/m up to 6.9, 4.0, and 1.8 times at a hydraulic gradient of 80,000 m/m. Room temperature and relative humidity during the entire experiment were 18.1 ± 0.4 °C and $48\% \pm 7\%$, respectively. A dynamic viscosity of $1.014 \pm 0.009 \,\mu\text{Pa}\cdot\text{s}$ was determined for SGW from eight replicate measurements at room temperature.

Apart from the specimens for the swelling and permeation experiments described above, but from the same batch of 10X, further specimens were compacted at hygroscopic humidity for studies using the method of computed microtomography (μ CT, using Yamato TDM1000H-2) and qualitative analyses of the microstructure by scanning electron microscopy (SEM) using LEO 1450 VP (Zeiss, Jena, Germany) with a guaranteed resolution of 5 nm). These specimens were prepared in the same manner as the swelling and permeation experiments with distilled water described in [4]. To study changes in the microstructure of 10X during swelling, SEM analyses were conducted with specimens after permeation with distilled water [4].

3. Results and Discussion

The μ CT study of the structure of compacted 10X showed that the specimen at a density of 1.4 g/cm³ is characterized by an uneven distribution of moisture (Figure 2), and areas of non-compacted material are no more than 0.25 mm in size. The specimen at a density of 1.6 g/cm³ is characterized by greater homogeneity, although a small amount of a non-compacted material (up to 0.1 mm on average, some areas with 0.5 mm) and some moisture inhomogeneity are observed. When compacted to a density of 1.8 g/cm³, the

obtained specimen does not contain any non-compacted parts, and no inhomogeneous distribution of moisture between the formed aggregates was observed. The appearance of cracks at the edges of the specimen at a density of 1.8 g/cm^3 (Figure 2c) was attributed to a relaxation of the 10X's fabric upon taking the specimen out of the compaction mold for the μ CT study.



Figure 2. μ CT images of 10X specimens compacted to dry densities of (a) 1.4 g/cm³; (b) 1.6 g/cm³; (c) 1.8 g/cm³.

The initial stage of bentonite swelling upon saturation with solutions of low salinity [22,23] and—for some bentonites—with brines [20,21] is characterized by a "doublepeak" pattern as a result of (i) swelling of aggregates (the first peak), (ii) their subsequent partial decomposition and a collapse of the inter-aggregate pores (the depression), and (iii) swelling of the reorganised aggregates (the second peak). This pattern is also observed for 10X specimens at the studied densities (graphs on left-side in Figure 3). The variation of this pattern with changing density further exemplifies that the collapse of the inter-aggregate pores is more strongly pronounced at the higher porosity so that the second peak is much lower than the first peak at 1.4 g/cm^3 . This effect diminishes with decreasing porosity so that the second peak becomes higher than the first peak within 1 to 4 days at 1.8 g/cm^3 . The higher second peak was observed previously for low- and high-salinity solutions ([20–25] and references therein), whereas the lower second peak was observed for high-salinity solutions [20,21]. By combining the latter observation with the present one, it can be suggested that the collapse of the inter-aggregate pores may depend on the electrostatic repulsion between diffuse-double layers (DDL) of single montmorillonite particles or their aggregates, as high salinity is known to depress their DDL [26] and increase pore volume available for their reorganisation accordingly.



Figure 3. Total pressure at tops of permeameter cells for 10X specimens compacted to dry densities of 1.4 g/cm³ (cells A1–A3); 1.6 g/cm³ (cells B1–B3); and 1.8 g/cm³ (cells C2, C3), saturated and permeated by SGW. Graphs on left-side represent x- and *y*-axis close-ups of the data measured within the first four days of saturation. Vertical dotted lines mark onsets of solution supply by burettes through cell tops (28th day) and by the pump at hydraulic gradients of 2000 m/m (133rd day), 5000 m/m (246th day), and 20,000 m/m (261st day) (note that corresponding labels in the 2nd graph on right-side).

It can be further seen in Figure 3 that despite the identical preparation protocol of specimens, the method used did not eliminate the inherent inhomogeneity of the microstructure of compacted bentonites. As a result of this inhomogeneity, measured swelling pressures and the time span of development for the second peak of the swelling pattern show significant variation at a given density and are otherwise similar to initial swelling patterns. This variation would not disappear (and even became larger at the densities of 1.4 and 1.6 g/cm^3) upon switching the solution supply from cell bases to cell tops on the 28th day. Possible reasons for this behaviour can be better understood considering the solution volumes absorbed by specimens and channels of permeameter cells from burettes within the first 115 days of the experiment (Figure 4). Firstly, these data reveal that the bentonite saturation was not accomplished within this stage, as the solution uptake did not level off. This occurrence is a well-known drawback of permeameter tests performed without a back pressure high enough to ensure full saturation of compacted specimens [27]. Secondly, for specimens with estimated pore volumes of 7.0–9.8 cm³ at a given density, a variation in the absorbed solution volumes of up to 10.7 cm³ was observed, which clarifies that the air was still present in the channels of some permeameters or even in the specimens. Hence, a reliable determination of the solution uptake by specimens upon saturation from burettes in the experimental setup used seems only possible through a test termination, dismantling of permeameters, and weighing the specimens.



Figure 4. Uptake of SGW by permeameter cells with 10X specimens compacted to dry densities of 1.4 g/cm³ (A1–A3); 1.6 g/cm³ (B1–B3); and 1.8 g/cm³ (C2, C3), upon saturation from burettes connected to inlets at cell bases for 27 days and cell tops afterwards.

The subsequent application of the hydraulic gradient of 2000 m/m led to a significant gradual increase of swelling pressure in all specimens, presumably, as a result of displacement of residual air from specimens by the injected solution (as discussed below to permeabilities) and/or microstructural reorganisation (as discussed below). During this stage and the preceding saturation stage using burettes, the total pressure measured at cell tops was equal to the swelling pressure of specimens. Starting from the 246th day, however, it was equal to the sum of the swelling pressure of the specimens and the fluid pressure established at cell tops upon imposing the hydraulic gradient of \geq 5000 m/m across the specimens. This can be seen in steep increases of the total pressure at top of all cells on the 246th, 261st (Figure 3), and 281st day (Figure 5) upon increasing the hydraulic gradient to 5000; 20,000, and 80,000 m/m, respectively. It also results from its steep decrease to the values measured before the 246th day upon the intermediate and final switching off the pump on the 294th and 297th day, respectively (Figure 5). Therefore, the swelling pressure of 10X compacted to the densities of 1.4, 1.6, and 1.8 g/cm³ can be concluded to equal to $0.8\pm0.3, 2.2\pm0.6$, and 6.3 ± 0.3 MPa, respectively, measured at the hydraulic gradient of 2000 m/m (Table 2).



Figure 5. Total pressure at tops of permeameter cells for 10X specimens compacted to dry densities of 1.4 g/cm³ (A1–A3), 1.6 g/cm³ (B1–B3), and 1.8 g/cm³ (C2, C3) and permeated by SGW at a hydraulic gradient of 80,000 m/m. Real fluid injection pressure imposed by the pump is shown in the upper graph (note that the abrupt fluid injection pressure changes were caused by short switching off the pump before weekends to disconnect cells A1–A3, preventing an overflow of burettes connected to them and after weekends, reconnecting them).

.o, and 1.0 g/ cm , saturated and permeated by 5000.											
•		σ , MPa		κ , 10 ⁻²⁰ m ²							
<i>₁_H,</i> m/m	1.4 g/cm ³	1.6 g/cm ³	1.8 g/cm ³	1.4 g/cm ³	1.6 g/cm ³	1.8 g/cm ³					
180	0.7 ± 0.2	1.5 ± 0.6	5.3 ± 0.9	-	-	-					
2000	0.8 ± 0.3	2.2 ± 0.6	6.3 ± 0.3	27 ± 15	3.4 ± 0.8	0.96 ± 0.26					
5000	0.9 ± 0.3	2.3 ± 0.6	6.4 ± 0.3	25 ± 14	3.2 ± 0.4	0.54 ± 0.10					
20,000	2.1 ± 0.2	3.1 ± 0.6	6.9 ± 0.3	11 ± 4	2.9 ± 0.4	0.68 ± 0.02					
80,000	7.6 ± 0.1	8.3 ± 0.8	9.7 ± 0.3	5.4 ± 1.5	2.0 ± 0.2	0.67 ± 0.07					

Table 2. Total pressures at the top (σ) and permeabilities (κ) averaged over the last two weeks of measurement at a given hydraulic gradient (i_H) of 10X specimens compacted to dry densities of 1.4, 1.6, and 1.8 g/cm³; saturated and permeated by SGW.

Based on these values, the fluid pressure at cell tops can be estimated to equal 1.3, 0.9, and 0.6 MPa for the fluid pressure of 2.0 MPa at cell base (hydraulic gradient of 20,000 m/m) and 6.8, 6.1, and 3.4 MPa for the fluid pressure of 8.0 MPa at cell base (hydraulic gradient of 80,000 m/m) at dry densities of 1.4, 1.6, and 1.8 g/cm³, respectively. The decrease of fluid pressure from cell bases to cell tops is in line with previous observations of the non-linear variation of the excess fluid pressure across specimens in constant-head experiments with compacted clays [28]. A decrease in these values with increasing density further indicates that the degree of non-linearity in this variation increases with the decreasing porosity of the specimens (0.50, 0.43, and 0.36, respectively).

In the preliminary 7-day experiments [4] with 10X and distilled water using permeameters similar to those of the present study, for dry densities in the range of 1.44–1.82 g/cm³, swelling pressures in the range of 0.52–9.39 MPa and a relationship of the form $P = \exp(6.4\rho_d - 9.403)$ between swelling pressure P (MPa) and dry density ρ_d (g/cm³) were obtained. At the hydraulic gradient of 2000 m/m in the present study, a somewhat different relationship $P = \exp(5.16\rho_d - 7.445)$ was obtained (Figure 6). The difference might be due to the use of only three data points for obtaining the relationship in the present study, or the difference between SGW and distilled water, or for other reasons, which cannot be identified in view of the limited information on parameters of the experiments and statistical weight of fitted data in the study [4]. In the study by Wang et al. [29], relationships of the form $P = \exp(6.75\rho_d - 8.634)$, $P = \exp(6.85\rho_d - 9.231)$, and $P = \exp(3.32\rho_d - 5.608)$ were provided for MX-80 (75–90% smectite), FEBEX (92% smectite), and Kunigel V1 (48% smectite) bentonites, respectively. Based on the smectite content of 73 mass% of 10X [4], it may be speculated that the value of the coefficient preced-

ing the density ought to be in a range from 3.3 to 6.8. Although the values obtained in [4] and in the present study agree with this range, establishing values that are characteristic of 10X would require additional experimental data.



Figure 6. Swelling pressure and permeability (m²) of 10X specimens permeated by SGW as a function of dry density ρ_d . Circles and solid lines represent measured values and fitted curves of the present study, whereas dashed lines represent fitted curves of the study [4] (hydraulic conductivities reported in [4] were converted to permeabilities for the purpose of comparison).

Permeabilities measured for 10X in the present study show a gradual decrease at all densities during the initial experimental stage at the hydraulic gradient of 2000 m/m (Figure 7). The gradual decrease of permeabilities suggests that the residual air, if any, was largely displaced from the specimens at the very beginning of this stage since air removal would otherwise lead to a non-transient increase of permeability [27], which was not observed later on. At densities of 1.4 and 1.6 g/cm³, a double linear decrease was observed with a steeper initial decrease during the first 10 days at 1.4 g/cm³ and 40 days at 1.6 g/cm³ followed by a flatter one. Shortly after further increasing the hydraulic gradients at these densities, considerably higher transient permeabilities were measured (data points on the vertical dotted lines in Figure 7, which should not be treated as outliers but rather attributed to the additional amount of water squeezed out of specimens by seepage-induced (local) consolidation under the pressure gradient [28,30]. According to [30], the initial transient response preceding the steady-state flow condition can take up to a few hours for clay specimens due to time-dependent changes in the volume or distribution of pore space in a specimen.



Figure 7. Permeability (m²) of 10X specimens compacted to dry densities of 1.4 g/cm³ (A1–A3); 1.6 g/cm³ (B1–B3); 1.8 g/cm³ (C2, C3), and permeated by SGW. Vertical dotted lines mark increases of hydraulic gradient to 5000 m/m (246th day); 20,000 m/m (261st day), and 80,000 m/m (281st day).

As a result of hydraulic gradient increases above 2000 m/m, steady-state permeability showed two major decreases by a factor of ~2–2.5 at a density of 1.4 g/cm^3 and one major decrease by a factor of ~1.5 at 1.6 g/cm³, whereas it stabilized at ~6 × 10⁻²¹ m² at 1.8 g/cm³ (Figure 7, Table 2). The seepage-induced consolidation, which reduces the specimen's pore volume, was proposed as a reason for permeability decrease with increasing pressure gradient [28,30]. This reason cannot be discarded for the present experiment, as the observed dependence of the extent of permeability decrease on the density would be in line with a larger seepage-induced consolidation for less dense specimens. If this proposal is valid, a larger hydraulic gradient than those applied in the present study can be expected to cause a major decrease of steady-state permeability of 10X specimen at 1.8 g/cm³ as well.

Alternatively, the hydraulic conductivity of expansive materials was observed to decrease after full saturation due to reorganisation of microstructure in the course of a water potential re-equilibration and corresponding water redistribution between the inter-aggregate and intra-aggregate pores [31]. Accordingly, it can be assumed that such microstructural reorganisation may have occurred to a larger extent for 10X specimen at a density of 1.4 g/cm³ and to a lesser extent at 1.6 g/cm³, whereas it did not take place at 1.8 g/cm³, at least for hydraulic gradients \geq 5000 m/m. This assumption would be in line with the decreasing pore volume available for a microstructural reorganisation with increasing density.

A change in the pore space during the SGW saturation was detected by scanning electron microscopy. The microstructure of the original compacted 10X is characterized by the predominance of fine interparticle micropores and large inter-micro-aggregate pores. During saturation with the SGW, the number and size of pores are decreased (Figure 8), inter-micro-aggregate and small inter-micro-aggregate micropores predominate, and an increase in the size and isometricity of large inter-micro-aggregate pores is also noted. There is also a decrease in the degree of orientation of structural elements (clay aggregates of different sizes) in the plane orthogonal to the pressing direction, acquired during the process of compacting the specimen, in which the microstructure becomes more uniform.



Figure 8. SEM images of the compacted 10X, fraction <0.25 mm, density 1.65 g/cm³, at 32,000×: (**a**) natural micro-aggregates of clay particles after an air-drying; (**b**) partially swelled micro-aggregate of clay particles. Sidebars on the top corners: structural model of clay swelling with different moisture (W) content: (**a**) $W \le Wmg$; (**b**) Wp < W < Wsw (Wmg—maximal hygroscopic moisture content; Wp—moisture of the plastic limit; Wsw—moisture of swelling). 1—clay particle; 2—adsorbed water; 3—osmotic water.

This uniformity in microstructure is explained by the processes of hydration of highly hydrophilic clay particles upon saturation of the specimens and accompanying swelling of microaggregates, closure of fine pores, and opening of inter-ultra-micro-aggregate (inter-micro-aggregate) micropores. As a result, the microstructure of the specimens after permeation becomes more homogeneous, intra- and inter-micro-aggregate pores become predominant since they open significantly during the swelling stage but are not affected by the fluid pressure during the permeation stage. This effect can also be associated with the swelling of the specimens during saturation and with the so-called "hinge-joint effect" of clay particles in a micro-aggregate during hydration [32].

The swelling of clays is often characterized by the effect of disordering the microstructure. According to the results of experiments and calculations given in the article [33], this effect is associated with the distribution of forces acting on particles and microaggregates, the result of which leads to the appearance of the hinge-joint effect. Thus, as a result of the wedging pressure of the hydration shells around the particles, their mutual repulsion occurs, while the forces of attraction at their edges prevent complete separation, resulting in the formation of a honeycomb structure with higher porosity and a lower degree of microstructure uniformity (Figure 8, sidebars).

A displacement of particles, which are not directly involved in the load-carrying of specimen's fabric downward the flow direction, resulting in clogging of specimen's pores was proposed as a further possible reason for seepage-induced changes in the permeability [30]. In the study by Al-Taie et al. [34], clogging of sintered filters by this mechanism was proposed as an alternative explanation for permeability decreases observed upon increases of hydraulic gradients above 1000 m/m in experiments with a smectite-rich clay and distilled water. However, in the infiltration tests of hydraulic gradients up to ~64,000 m/m [29], no density changes along the profile of a bentonite specimen, which would provide evidence of such displacement, were found. Furthermore, the initial permeability of compacted 10X was ~10⁻¹⁸ m² at 1.4 g/cm³ (Figure 7), whereas that of sintered filters was $(3.4 \pm 0.5) \times 10^{-14}$ m², so that clogging of pore space would be more probable than that of sintered filters at 1.4 g/cm³ and even more so at 1.8 g/cm³. Since, no major decreases of steady-state permeability occurred at this density, clogging of pore space or sintered filters due to displacement of particles within the studied specimens was not a factor contributing significantly to the observed permeability decreases. Indeed, the permeability of

uncleaned sintered filters after the experiment was measured to equal $(1.9 \pm 1.2) \times 10^{-14}$, $(1.5 \pm 0.6) \times 10^{-14}$, and $(1.2 \pm 0.2) \times 10^{-14}$ m² at a dry density of 1.4, 1.6, and 1.8 g/cm³, respectively, or a factor of 2 to 3 lower than that before the experiment. Thus, though some clogging of sintered filters occurred during the experiment, its contribution to the observed permeability decreases can be neglected.

In view of remaining uncertainty about the mechanism behind the permeability decreases observed at higher hydraulic gradients, permeability values of $(27 \pm 15) \times 10^{-20}$, $(3.4 \pm 0.8) \times 10^{-20}$, and $(0.96 \pm 0.26) \times 10^{-20}$ m² (Table 2) obtained at a hydraulic gradient of 2000 m/m were suggested as conservative estimates of permeabilities of 10X for SGW at a temperature of 18.1 ± 0.4 °C and dry densities of 1.4, 1.6, and 1.8 g/cm³, respectively. When assessing the performance of the bentonite in the early post-closure phase of DGR at hydraulic gradients higher than 2000 m/m, corresponding permeability estimates given in Table 2 may be used.

In the experiments with seawater, permeability κ (expressed in millidarcies, 9.869233 × 10^{-19} m²) of pure smectite and kaolinite was found to be related to bulk density ρ_{bulk} (g/cm³) by respective equations $\ln \rho_{\text{bulk}} = -0.037 \ln \kappa + 0.27$ and $\ln \rho_{\text{bulk}} = -0.074 \ln \kappa + 0.40$ [35]. Notably, as can be seen from the equations derived in the present study (Figure 9), the coefficient preceding $\ln \kappa$ at hydraulic gradients of 2000 and 5000 m/m (0.040 ± 0.005 and 0.035 ± 0.001 , respectively) is close to the value obtained in study [35] for pure smectite, whereas at 80,000 m/m it is close to that for a mixture of 20% smectite and 80% kaolinite.



Figure 9. Permeability (mD) of 10X permeated by SGW at hydraulic gradients of 2000–80,000 m/m as a function of bulk density. Circles and solid lines represent measured values and fitted curves.

The equation $\ln \rho_{\text{bulk}} = -0.040 \ln \kappa_{(\text{mD})} + 0.47$ (Figure 9), obtained in the present study at the hydraulic gradient of 2000 m/m, corresponds to $\ln \kappa_{(\text{m}^2)} = -13.4 \ln \rho_{\text{dry}} - 38.3$ (Figure 6), with κ being a power function of ρ_d . In this form it can be compared with the relationship $\ln \kappa_{(\text{m}^2)} = -9.03\rho_d - 14.778$ obtained in the experiments with distilled water [4] where κ is an exponential function of ρ_d . In the semi-logarithmic plot (Figure 6), the latter relationship has the linear form whereas that used in the present study and the study by Mondol et al. [35] does not. In view of this difference, it appears that establishing the form of the relationship between permeability and density that is characteristic of 10X requires additional experimental data.

The comparison of the fitted curves in Figure 6 further reveals that permeabilities obtained in [4] at hydraulic gradients of 22,000 up to 92,000 m/m are about 2.5 times smaller than those in the present study obtained at a hydraulic gradient of 2000 m/m. Upon

increasing the hydraulic gradient to 20,000 m/m in the present study, the permeability decreased by a factor of 2.5 at 1.4 g/cm³ and of 1.5 at 1.8 g/cm³ (Table 2), whereas at 1.6 g/cm³ it decreased by a factor of 1.7 only upon an increase of hydraulic gradient to 80,000 m/m. Therefore, the difference between the permeabilities measured in the present study and in [4] may be due to hydraulic gradients higher than 2000 m/m in the latter study. A contribution of other reasons to this difference cannot be excluded and needs to be verified in additional experiments.

4. Conclusions

Differences in the swelling pressures and permeabilities observed in the present study for 10X bentonite at given dry densities suggests that because of inherent inhomogeneity in the microstructure of compacted bentonites, at least duplicate, preferably triplicate, experiments should be conducted when using the present method to estimate swelling pressures or permeabilities. In this regard, a saturation of compacted bentonite specimens and measurements of their swelling pressure and permeability are recommended to be conducted at a hydraulic gradient of 2000 m/m. As higher hydraulic gradients cause modifications of compacted bentonite microstructure, which are still not well understood, the estimates obtained with those should be used with care, and their applicability may be restricted to the early post-closure phase of DGR.

To obtain a reliable estimate of the dependence of swelling pressure and permeability of compacted bentonite on its density, a series of triplicate experiments for at least 7–8 densities, preferably with an even distribution over a broader density range than that of 1.4–1.8 g/cm³ studied in work [4] and the present study, is needed. The dataset for 10X bentonite should be accordingly extended.

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