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Swelling pressure and permeability of laminated bentonite–sand mixtures with different exchangeable cation composition

Y. Watanabe[1], S. Yokoyama[2], and E. Yoshikawa[3]

[1]Dr., Central Research Institute of Electric Power Industry, Chiba, Japan, email: yasutaka@criepi.denken.or.jp

[2]Dr., Central Research Institute of Electric Power Industry, Chiba, Japan, email: shingo@criepi.denken.or.jp

[3]Dr., Central Research Institute of Electric Power Industry, Chiba, Japan, email: yoshikawa3829@criepi.denken.or.jp

ABSTRACT

Changes in the swelling pressure and permeability of laminated compacted bentonite–sand mixtures with different exchangeable cation compositions were investigated using a permeating salt solution. A laminated specimen was produced by stacking two compacted bentonite–sand mixtures: a 7% Na-bentonite–sand mixture on the upper side and a 30% Ca-bentonite–sand mixture on the lower side. The laminated specimen was infiltrated using 0.1 and 1.0 mol/L Na_2SO_4 solutions. The vertical pressure, hydraulic conductivity, and ion concentration in each effluent solution were measured. Ca-bentonite (upstream) discharged Ca ions by ion exchange, whereas Na-bentonite (downstream) exhibited ion exchange from Na to Ca ions. The equivalent hydraulic conductivity slightly differed from that of the laminated specimen at 0.1 mol/L. As lower concentrations cause osmotic swelling, it is essential to estimate the hydraulic conductivity of the downstream material using a complex ion-exchange process to obtain precise equivalent hydraulic conductivity. After changing the solution to deionized water, the vertical pressure increased, whereas the hydraulic conductivity decreased. The estimation of swelling pressure is expected to be useful in setting the overburden pressure to prevent swelling deformation and maintain the low permeability of the engineered barrier in radioactive waste disposal.

Keywords: expansive clay, montmorillonite, swelling, hydraulic conductivity, ion exchange, salt solution

1   INTRODUCTION

Compacted bentonite–sand mixtures are used as engineered barriers at radioactive waste disposal sites to delay nuclide migration. A low-level radioactive waste facility in Japan comprises concrete, a low-permeability layer, bottom cover soil, and top cover soil above the waste packages (Figure 1). In the preliminary design (JNFL, 2021), Na-bentonite or Ca-bentonite is used by mixing sandy soil as the low-permeability layer, and a bentonite–sand mixture using Na-bentonite or Ca-bentonite with different bentonite mixture ratio is also used as the bottom cover soil. This indicates that bentonites with different exchangeable cation compositions can be used for lamination.

Salt solutions produced by waste and groundwater infiltrate the bentonite–sand mixture. Therefore, montmorillonite ion exchange may occur affecting the properties of bentonite–sand mixtures (Watanabe & Yokoyama, 2018; Zeng et al., 2022). Laminations of bentonite–sand mixtures with different chemical compositions exhibit complex ion-exchange reactions and hydromechanical responses. However, a lack of experimental studies has hindered the advancement in understanding the occurrence of this phenomenon in lamination systems.

Therefore, in this study, we investigated changes in the swelling pressure and permeability of laminated compacted bentonite–sand mixtures with different exchangeable cation compositions, using the permeation of salt solutions. To produce each laminated specimen, two types of compacted bentonite–sand mixtures were stacked. The hydraulic conductivities of the laminated bentonite–sand mixtures were compared with the equivalent hydraulic conductivities calculated using the measured values for each layer. Furthermore, assuming a gradual decrease in groundwater salinity over a long period, changes in the vertical pressure and permeability of the bentonite–sand mixture after the ion exchange of montmorillonite were measured by replacing the influent solution from a salt solution to deionized

water. Based on the analysis of the interlayer cation composition, the hydromechanical response to an ion-exchange reaction in the lamination system is discussed.
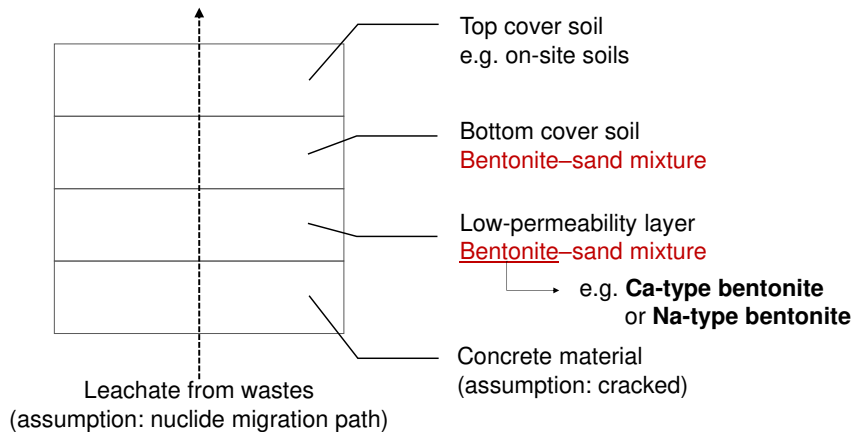

Figure 1. *Conceptual image of the engineered barrier and cover soils using different type of bentonites.*

2   EXPERIMENTAL PROCEDURE

2.1   Materials and methods

The bentonites used in this study were Na-bentonite, Kunigel V1, and Ca-bentonite, Kunibond (Kunimine Industries Co., Ltd., Japan). The leachable cations of the bentonites washed with an alcohol solution were identified by leaching with a 6% benzyl trimethylammonium solution (Shinoki et al., 2009). The leachable cations in each sample were representative of the exchangeable cations in that sample. The amount of methylene blue (MB) adsorbed onto the bentonites was measured according to the Japanese Industrial Standard (JIS Z 2451, 2019). Specimens were created by mixing the bentonites with sand. The grain-size distribution of the sand was measured using the sieving method provided in JIS A 1204 (2009).

2.2   Specimen production

A specimen with a diameter of 60 mm and total height of 20 mm was produced using two compacted bentonite–sand mixtures, each with a thickness of 10 mm. Two types of bentonite–sand mixtures were compacted separately within 10 mm-thick rings. These rings were then stacked, as illustrated in Figure 2. Provisional requirements state that the bottom cover soil has a hydraulic conductivity lower than 1×10^{-8} m/s (JNFL, 2021). In this study, to satisfy this requirement, the bentonite mixture ratio of the Na-bentonite–sand mixture in the upper layer was 7%. The low-permeability layer was assumed to be a 30% Ca-bentonite–sand mixture (JNFL, 2021). Therefore, in this study, the bentonite ratio of the Ca-bentonite–sand mixture in the lower layer was 30%. The target dry density of the mixtures was 90% of the maximum dry density. The specimen conditions are presented in Table 1.

Table 1. *Conditions of specimens and experimental program.*

	Specimen location	Material	Molding water content (%)	Dry density (Mg/m^3)	Influent solution	Exchange of solution to deionized water
Case 1	Upper layer	7% Na-bentonite–sand mixture	11.7	1.89	0.1 mol/L-Na$_2$SO$_4$	No
	Lower layer	30% Ca-bentonite–sand mixture	15.1	1.58		
Case 2	Upper layer	7% Na-bentonite–sand mixture	11.8	1.89	1.0 mol/L-Na$_2$SO$_4$	Yes
	Lower layer	30% Ca-bentonite–sand mixture	14.3	1.60		

2.3 Swelling–permeability test

The laminated specimens were placed in a permeameter with load cells at the top and bottom (Figure 2). The porous end piece was previously dried. The test apparatus was assembled, and a vertical pressure of less than 20 kPa was measured, confirming contact between the specimen and porous end piece. After vacuuming the specimen and the tubes, the specimen was saturated from the bottom with the influent solution. The experimental program is summarized in Table 1. The component of the leachate from the waste was assumed to be Na_2SO_4 (JNFL, 2021). In this study, Na_2SO_4 solution (0.1 and 1.0 mol/L) was supplied from the bottom. The hydraulic gradient was less than 20. The effluent solution was then connected to a burette and the flow rate was measured. At each interval, the effluent solution was collected in a vessel to measure its chemical composition. The vertical pressure, hydraulic conductivity, and ion concentration of each effluent solution were continuously measured during the experiment.

In Case 2, after the Na-ion concentration in the effluent reached the same value as that of the influent solution, the influent solution was changed to deionized water. This study aimed to investigate the changes in vertical pressure and hydraulic conductivity after the cation exchange of montmorillonite due to decrease in ionic strength of the pore fluid, in the variable environmental conditions in radioactive waste disposal facilities. Each experiment was terminated when the vertical pressure and hydraulic conductivity stabilized. In both cases, following the procedure described above, the specimens were divided into 1 mm intervals and washed with alcohol. Leachable cations were measured using the method described in Section 2.1.

In addition, a monolayer permeability test was conducted in which the hydraulic conductivities of the compacted Na-bentonite– and Ca-bentonite–sand mixtures were measured using Na_2SO_4 solutions with the same specifications as those of the laminated specimens. The equivalent hydraulic conductivity k_{eq} was calculated using the following equation:

$$k_{eq} = H / (h_1 / k_1 + h_2 / k_2)$$
(1)

where H, h_1, and h_2 are the thicknesses of the entire laminated, upper-side, and lower-side specimens (m), respectively. k_1 and k_2 are the hydraulic conductivities (m/s) of the upper and lower sides, respectively, as measured using the monolayer permeability test.
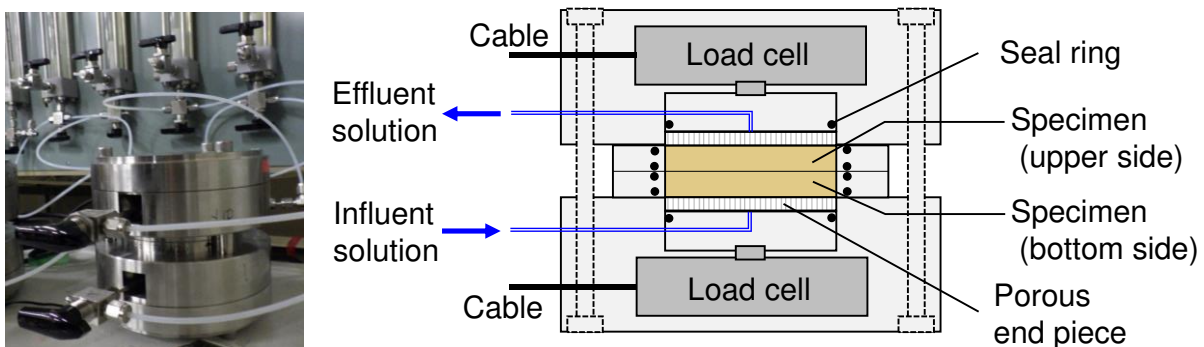

Figure 2. *Photograph and schematic diagram of the swelling–permeability test apparatus.*

3 RESULTS AND DISCUSSION

3.1 Physicochemical properties of bentonites

The physicochemical properties of the bentonite and sand used in this study are listed in Table 2. For Kunigel V1 and Kunibond, the amount of leachable Na and Ca ions was 81% and 71% of the total value, respectively. Therefore, this study confirmed that they are Na- and Ca-type bentonites. Based on the pure montmorillonite MB adsorption values of 140.0 mmol/100 g and 150.5 mmol/100 g for Na-bentonite and the Ca-bentonite, respectively (Watanabe and Yokoyama, 2022), the montmorillonite content was

estimated to be 54.3% for Na-bentonite and 75.1% for Ca-bentonite. The 50% particle diameter of the sand was approximately 0.2 mm.

Table 2. *Physicochemical properties of bentonites and sand used in this study.*

		Na-bentonite (Kunigel V1)	Ca-bentonite (Kunibond)	Sand
Particle density of soil (Mg/m^3)		2.744	2.680	2.659
Amount of MB adsorbed (mmol/100 g)		76	113	Not measured
Montmorillonite content (%)		54.3	75.1	Not measured
Amount of leachable cation (meq/100 g)	**Na$^+$**	51.3	12.3	Not measured
	Ca^{2+}	10.7	58.8	Not measured
	K$^+$	0.4	1.4	Not measured
	Mg^{2+}	1.0	10.0	Not measured
	Total	63.4	82.5	Not measured

3.2 Vertical pressure and hydraulic conductivity during permeation of Na$_2$SO$_4$ solutions

The vertical pressure and hydraulic conductivity measured in the swelling–permeability test using the Na$_2$SO$_4$ solution are shown in Figure 3. In both cases, the vertical pressure at the lower side increased immediately after the Na$_2$SO$_4$ solution was supplied. Subsequently, the pressure on the upper side increased by 0.5 h. The vertical pressure on the lower side was higher than that on the upper side. The vertical pressure continued to increase under 0.1 mol/L condition, but remained constant under 1.0 mol/L condition after the peak was attained. The vertical pressure tended to be higher in the 0.1 mol/L condition than in the 1.0 mol/L condition.

Notably, infiltration to the upper side was delayed because the solution was supplied from the bottom. The vertical pressures were measured almost simultaneously because the swelling of the lower side presumably pushed the upper side. The lower vertical pressure in Case 2 was most likely caused by the higher ionic strength of the pore fluid, which reduced the swelling properties of montmorillonite. This was consistent with the results of previous studies (Zhu et al., 2013). The slight increase in the vertical pressure might be caused by swelling of the upper layer under 0.1 mol/L condition, whereas swelling could not be detected because of the small swelling property under 1.0 mol/L condition. Furthermore, the difference in vertical pressure between the upper and lower sides could be attributed to the variations in specimen conditions, friction between the specimen and test ring, and direction of infiltration.

The hydraulic conductivity gradually decreased from 2×10^{-9} to 3×10^{-10} m/s under 0.1 mol/L condition (Figure 3 (a)). The hydraulic conductivity was maintained at approximately 6×10^{-9} m/s during Na$_2$SO$_4$ permeation under 1.0 mol/L condition (Figure 3 (b)). In both cases, the Na ion concentration of the effluent solution gradually increased and reached the same value as that of the influent solution after the pore volume ratio of the effluent solution exceeded five.

The increase in Na ion concentration indicated that Na ions were primarily consumed by montmorillonite ion exchange. As depicted in Figure 4, the ratio of leachable Ca ions decreased and that of leachable Na ions increased on the lower side, compared to those of the initial Ca-bentonite. On the upper side, the ratio of leachable Ca ions increased compared to that of the initial Na-bentonite. This indicated that the ion exchange of montmorillonite from Ca to Na ions proceeded and released Ca ions on the lower side. On the upper side, the ion exchange of montmorillonite from Na to Ca ions proceeded using the Ca ions provided from the lower side. Rao et al. (2013) reported that crystalline swelling was dominant, considering that the diffuse ion layer growth was curtailed in NaCl solutions with a concentration higher than 0.4 mol/L. Na-montmorillonite has a higher expansivity, whereas the Ca-montmorillonite interlayer distance cannot expand more than the three-water layer hydrate state (Fukushima, 1984). In the 1.0 mol/L condition, it was presumed that the vertical pressure and hydraulic conductivity were not affected by the ion exchange because the osmotic swelling induced by the diffuse ion layer was not substantially operative. In the 0.1 mol/L condition, the gradual increase in the interlayer Na ions on the lower side might enhance the swelling pressure by the osmotic effect and resulted in a gradual decrease in permeability (Figure 3 (a)).
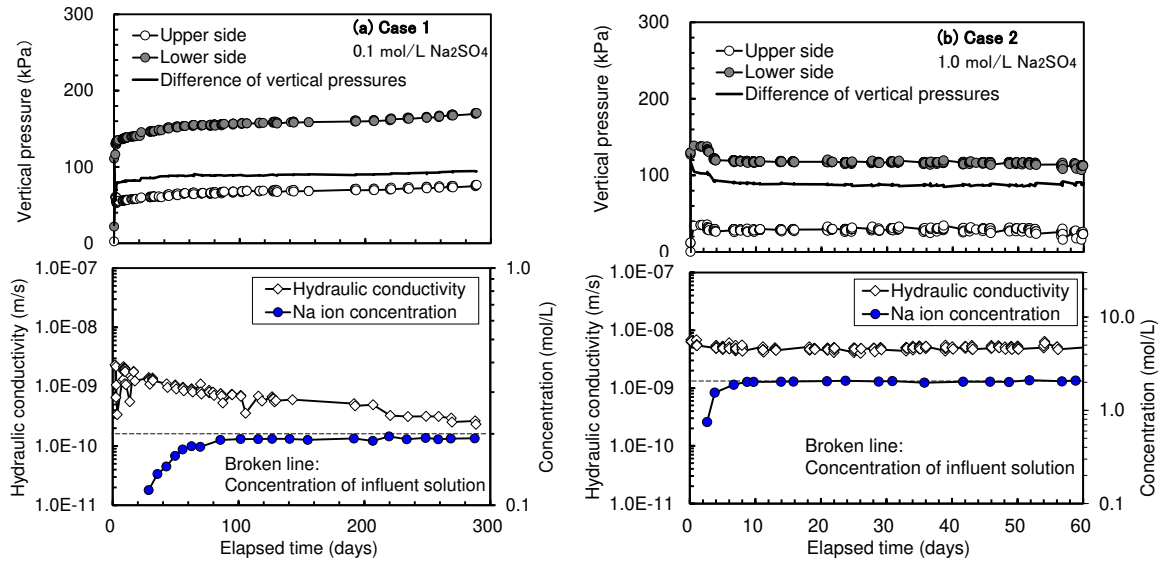
Figure 3. *Vertical pressure and hydraulic conductivity for (a) 0.1 mol/L and (b) 1.0 mol/L Na$_2$SO$_4$.*
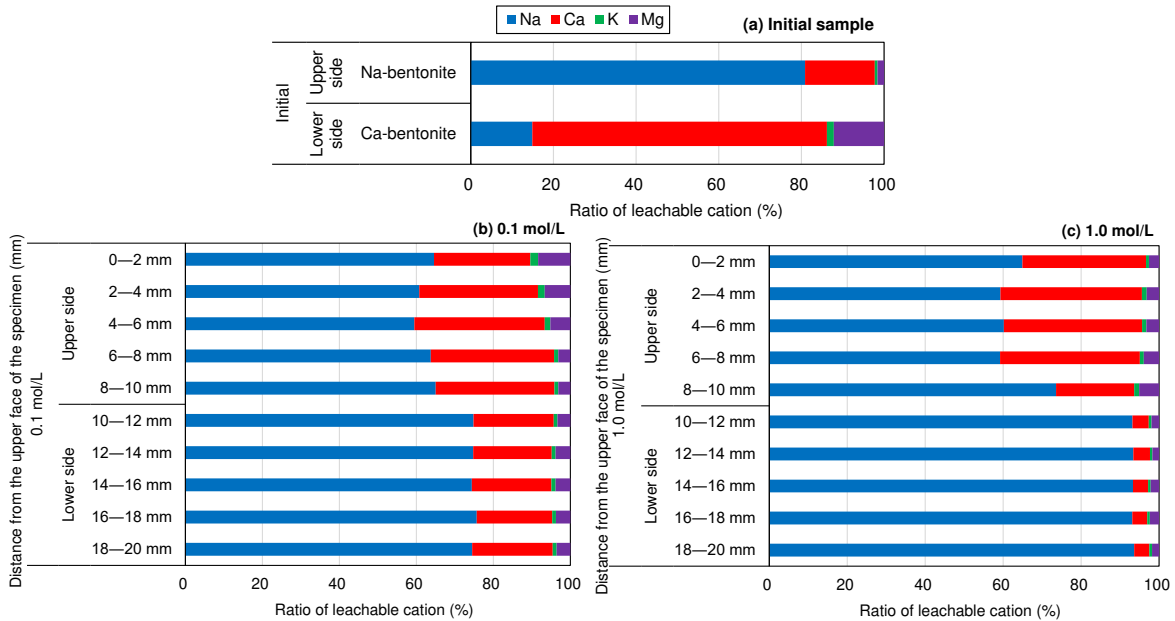

Figure 4. *Ratio of leachable cation of each specimen after the permeability test.*

3.3 Equivalent hydraulic conductivity

As shown in Figure 4, spatial variation in the composition of leachable cations was observed in the specimen, particularly on the upper side. Therefore, it is important to investigate the influence of the heterogeneous state of the specimen on the evaluation using equivalent hydraulic conductivity. The hydraulic conductivities of the laminated specimen and the equivalent hydraulic conductivities calculated by the monolayer permeability test are shown in Figure 5. The hydraulic conductivity of each material increased with the increasing Na$_2$SO$_4$ concentration. The hydraulic conductivity of the laminated specimen was compared with the equivalent hydraulic conductivity, which was consistent with 1.0 mol/L Na$_2$SO$_4$ solution. The hydraulic conductivity of the laminated specimen was twice than the equivalent hydraulic conductivity under 0.1 mol/L condition.

According to the experimental results (Figure 4), the upper side reacted with the pore fluid containing Ca ions, which were released from the lower side. In the monolayer permeability test, the Na-bentonite–sand mixture directly interacted with the Na$_2$SO$_4$ solution. Compared with the laminated system, ion exchange from Na to Ca ions in the Na-bentonite–sand mixture could not be simulated in the monolayer

permeability test. Therefore, the measured hydraulic conductivity could be lower in the monolayer permeability test, resulting in a marginally lower equivalent hydraulic conductivity under 0.1 mol/L condition. Under 1.0 mol/L condition, the osmotic effect was not operative. Then, the difference in the interlayer cation composition was insignificant when comparing the hydraulic conductivity of the laminated specimen with the equivalent hydraulic conductivity.

The ion exchange of montmorillonite exerted a greater effect on the permeability of compacted bentonite–sand mixtures at lower ion concentrations because it caused osmotic swelling. Thus, it was important to estimate the realistic hydraulic conductivity of each layer, particularly in downstream materials with complex ion-exchange processes, for a more precise evaluation of equivalent hydraulic conductivity.
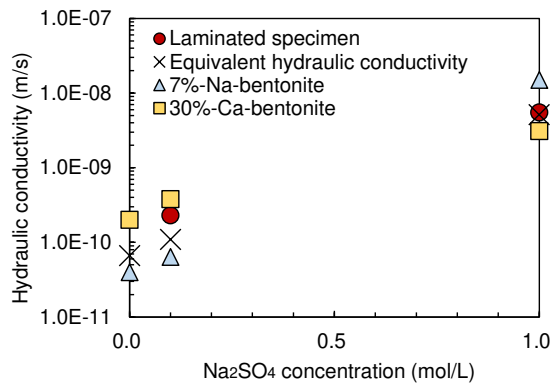

Figure 5. *Hydraulic conductivities of the laminated specimen and equivalent hydraulic conductivities calculated using the results of monolayer permeability test.*

3.4   Influence of change in pore fluid chemistry on swelling and permeability

The vertical pressure and hydraulic conductivity after changing the influent solution from 1.0 mol/L-Na_2SO_4 solution to deionized water are shown in Figure 6. The vertical pressure increased significantly after the permeation of the deionized water. The vertical pressure measured at the bottom increased more than that measured at the upper side. Because the ratio of leachable Na ions increased on the lower side, it was presumably caused by the swelling pressure increase after ion exchange under the pore fluid concentration diluted with deionized water. The hydraulic conductivity decreased from 6×10^{-9} m/s to 4×10^{-11} m/s after deionized water permeation. As the swelling pressure increased, it was assumed that larger pores were filled by swollen montmorillonite in the specimen, resulting in low permeability.

From a practical perspective, bentonite-based materials, such as the low-permeability layer and bottom cover soil, include the possibility of interaction with groundwater having low ionic strength after salt solution infiltration. In the lamination systems of different bentonites, the change in the interlayer cation composition of montmorillonite was more complex downstream and influenced by the ion-exchange reaction in the upstream layer. The effect of montmorillonite on ion exchange was evident when groundwater concentrations were reduced, and the swelling of Na-montmorillonite was predominant. An increase in the swelling pressure suggested that the swelling deformation occurred even under a higher confining stress. In the future, the estimation of the swelling pressure through experimental work, as shown in Figure 6, will be valuable for setting the overburden pressure to prevent swelling deformation (dry density reduction) considering the variable pore fluid chemistry. If the swelling deformation could be controlled, then the reduction in hydraulic conductivity induced by ion exchange from Ca to Na ions in montmorillonite will be advantageous in delaying radionuclide migration when the ion concentration decreases over the long term.
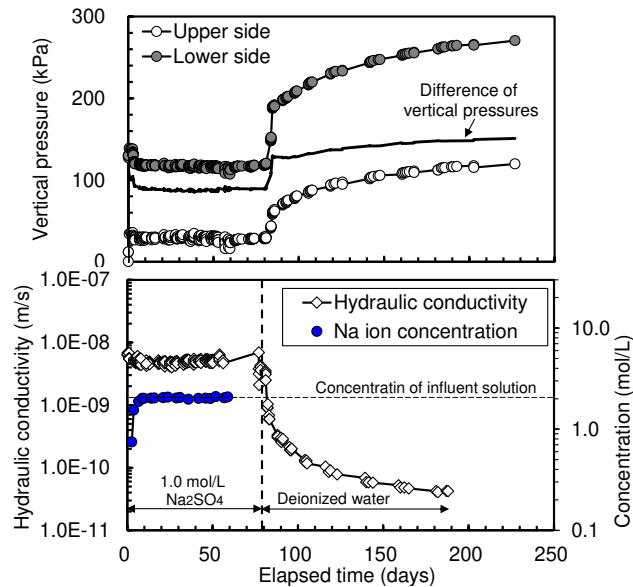
Figure 6. *Changes in vertical pressure and hydraulic conductivity by changing the influent solution.*

4   CONCLUSIONS

To understand the hydromechanical responses of the composite structures made up of different bentonites, this study investigated the changes in the swelling pressure and permeability of laminated compacted bentonite–sand mixtures with different exchangeable cation compositions using the permeation of Na_2SO_4 solutions.

When the laminated Na- and Ca-type bentonites were infiltrated with the Na_2SO_4 solution, our experiments indicated that the ion-exchange reaction from Ca to Na ions in the Ca-bentonite–sand mixture layer occurred on the upstream side, and the released Ca ions were consumed in the following ion-exchange reaction in the Na-bentonite–sand mixture layer occurring on the downstream side. Due to osmotic swelling being operational in the lower ionic strength of the pore fluid, the precise prediction of the changes in the interlayer cation composition of montmorillonite in the downstream parts through experimental or analytical work is important to evaluate the equivalent hydraulic conductivity of the laminated layer.

Furthermore, as shown in this study, the swelling pressure might increase following the ion exchange of montmorillonite (e.g., Ca to Na ions) and a reduction in the pore fluid ion concentration. This prediction may assist in the design of cover soils to control swelling deformation by the overburden pressure, and subsequently satisfying the long-term low-permeability of the engineered barrier.

REFERENCES

Fukushima, Y. (1984). X-ray diffraction study of aqueous montmorillonite emulsions. Clays and Clay Minerals. 32, 320-326.

JNFL. (2021). Compliance with permit criteria rules for waste disposal facilities (Haikibutsumaisetsushisetsu niokeru kyokakijunkisoku heno tekigousei nitsuite). Article 10, Item (iv). Japan Nuclear Fuel Limited (in Japanese).

JIS A 1204. (2009). Test method for particle size distribution of soils. Japanese Industrial Standard Committee, Tokyo, Japan.

JIS Z 2451. (2019). Test method for methylene blue adsorption on bentonite and acid clay. Japanese Industrial Standard Committee, Tokyo, Japan.

Rao, S. M., Thyagaraj, T., & Rao, P. R. (2013). Crystalline and osmotic swelling of an expansive clay inundated with sodium chloride solutions. Geotechnical and Geological Engineering. 31, 1399-1404.

Shinoki, S., Yamada, T., Kurosaka, K., & Ito, H. (2009). Measurement of leached cation of bentonite using benzyltrimethylammonium chloride solution. Proceedings of the 53rd Clay Science Symposium. Clay Science Society of Japan, Iwate, Japan, 53, 73 (in Japanese).

Watanabe, Y., & Yokoyama, S. (2018). Changes in swelling pressure and permeability of bentonite caused by ion exchange of montmorillonite. Proceedings of the 8th International Congress on Environmental Geotechnics. International Society for Soil Mechanics and Geotechnical Engineering, China, 523-528.

Watanabe, Y., Yokoyama, S., Shimbashi, M., Yamamoto, Y. & Goto, T. (2022). Saturated hydraulic conductivity of compacted bentonite–sand mixtures before and after gas migration in artificial seawater. Journal of Rock Mechanics and Geotechnical Engineering. in press, https://doi.org/10.1016/j.jrmge.2022.01.015.

Zeng, Z., Cui, Y. J., & Talandier, J. (2022). Effect of water chemistry on the hydro-mechanical behaviour of compacted mixtures of claystone and Na/Ca-bentonites for deep geological repositories. Journal of Rock Mechanics and Geotechnical Engineering. 14, 527-536.

Zhu, C. M., Ye, W. M., Chen, Y. G., Chen, B., & Cui, Y. J. (2013). Influence of salt solutions on the swelling pressure and hydraulic conductivity of compacted GMZ01 bentonite. Engineering Geology. 166, 74-80.

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