Cementation Effect on Swelling and Permeability Properties of Bentonite Considering Microscopic Structural Evaluation

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ABSTRACT: Bentonite buffer material in the geological disposal of high-level radioactive waste would be subjected to high temperatures and pressures for a long period, which may cause alteration through cementation and changes of swelling and permeability characteristics. In this study, bentonite ore was used to simulate the cemented buffer material, and effects of cementation on swelling and permeability properties were evaluated. As results, swelling pressure of undisturbed specimens was about half that of reconstituted specimens, while hydraulic conductivity was almost in the same order. To elucidate mechanism from microstructural viewpoint, SEM and XRD was utilized. SEM observations showed the undisturbed specimens had more complex structure at same dry density. XRD analysis indicated that undisturbed specimens had smaller montmorillonite basal spacing at same water content. Therefore, cementation may reduce permeability by complexity of structure, and inhibit water absorption and swelling of montmorillonite interlayers, resulting in almost no change in hydraulic conductivity.

1 BACKGROUND

In the geological disposal of high-level radioactive waste, bentonite is planned to be used as a buffer material surrounding the waste in many countries (Pusch, 1992; Sellin & Leupin, 2013). Bentonite has high swelling capacity and low permeability, so it can delay contact between radionuclides and groundwater. The properties of bentonite at the initial condition of disposal have been investigated experimentally and theoretically in many countries (Alonso & Gens, 1999; Komine & Ogata, 2004; Garitte et al., 2017), and repository design proposals have been established (Ogata et al., 1999). On the other hand, because of the long half-life of radionuclides, buffer material is required to maintain their integrity for tens of thousands of years. However, because the buffer material may be subjected to high temperatures and pressures and inflow of groundwater containing salts, there is concern that cementation would occur and properties such as swelling and low permeability will deteriorate. It is difficult to reproduce and evaluate such long-term alteration phenomena only by laboratory experiments. In this study, we focused on natural analogues, a geological research method, and used raw ore from bentonite deposits that has undergone diagenesis in the natural ground as a simulated buffer material.

In this paper, to quantitatively evaluate the effect of cementation on swelling and permeability properties of bentonite, measuring swelling pressure, and hydraulic conductivity experiments were conducted, and microscopic structural evaluation was performed using SEM and XRD.

2 SAMPLE USED IN THIS STUDY AND TESTING APPARATUSES

2.1 *Fundamental properties of bentonite ore and specimen preparation*

Na-bentonite ore from the Tsukinuno Mine in Yamagata, Japan (Tsukinuno ore) was used. Fundamental characteristics of the ore are shown in Table 1. The bentonite ore used in this study is shown in Figure 1.

To evaluate the cementation effects on swelling and permeability properties, undisturbed and reconstituted specimens were prepared by the following method. The undisturbed specimens were formed from ore using cutter rings, trimmers, knives, and were made into specimen sizes (28 mm in diameter × 10 mm in height for swelling pressure test and 28 mm in diameter × 2 mm in height for permeability experiment). The reconstituted specimens were prepared using a crusher with grain sizes of ~0.425 mm, 0.850 to 2 mm, and 2.00 to 4.75 mm, and then compacted to an arbitrary dry density by applying static load. All grain sizes were used in the swelling experiments, while only specimens with grain diameters of ~0.425 mm was used in the permeability experiments.

Table 1. Fundamental characteristics of bentonite ore.

Soil particle density (Mg/m^3)	2.77
Liquid limit (%)	419.1
Plastic limit (%)	29.2
Plasticity index	389.9
Montmorillonite content (%)	44.7
Leached cation (Na^+) (cmol(+)/kg)	43.7
Leached cation (Ca^+) (cmol(+)/kg)	5.6
Leached cation (K^+) (cmol(+)/kg)	Lower than detection limit
Leached cation (Mg^{2+}) (cmol(+)/kg)	0.9
Accessory minerals	Quartz, Plagioclase
Approximate geological age	Ten million years

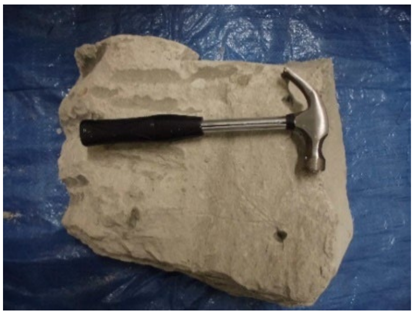

Figure 1. Appearance of bentonite ore.

2.2 *Testing devices of swelling pressure and hydraulic conductivity test*

Figure 2 shows the experimental apparatus. Lateral deformation of the specimen was restrained by a stainless ring, and vertical deformation was controlled by tightening the clamp knob in the swelling pressure experiment. Evaluation is based on the relationship between the value obtained when the increase in swelling pressure converges after a certain period (maximum swelling pressure) and the dry density at that time.

Figure 3 shows the hydraulic conductivity apparatus. This apparatus is for falling head hydraulic conductivity test, and burettes are connected to both the inlet and outlet sides and air pressure is applied to add hydraulic gradient. Details of the experimental setup and procedures are described in Ito et al., 2022. Tests were conducted under conditions of a hydraulic gradient of 4000-20000 during the test period, and in this range of hydraulic gradients, proportional relationship between flow flux and hydraulic gradient was observed which follows Darcy's law. Average hydraulic conductivity was taken for each measurement period in a similar way as Ito et al. (2022a).

3 TESTING RESULTS ON SWELLING AND HYDRAULIC PROPERTIES

Figure 4 shows relation between maximum swelling pressure and dry density of specimen. For the reconstituted specimens, maximum swelling pressure tended to be higher with higher dry density, and the effect of grain size before compaction was small. On the other hand, the undisturbed specimens showed a variation in pressure values, but all of them were only about half of those of the reconstituted specimens of similar dry density. These results indicate that swelling pressure of undisturbed specimen is reduced by cementation, although the effect of cementation might be small when the samples are crushed before compaction.

Figure 5 shows the relation between hydraulic conductivity and dry density of specimens, as well as theoretical calculation using equations proposed by Komine (2008). Hydraulic conductivities of the undisturbed and reconstituted specimens were all in the order of 10^{-12} to 10^{-13}, which are similar to the calculation results.

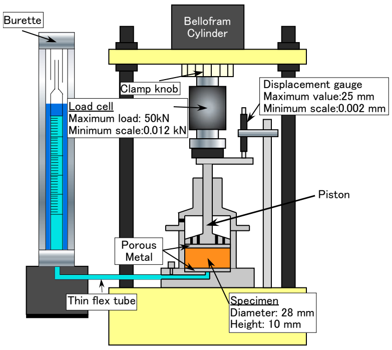

Figure 2. Swelling pressure test apparatus.

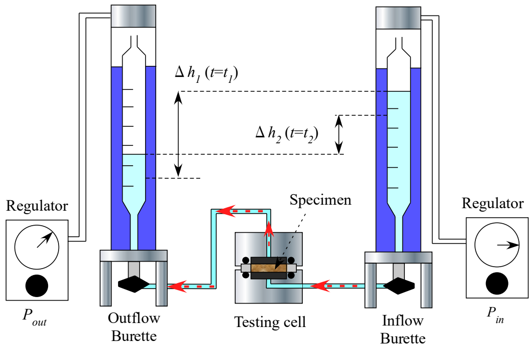

Figure 3. Hydraulic conductivity apparatus.

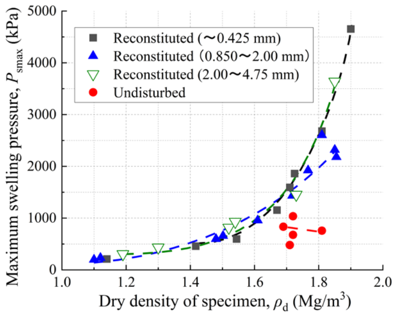

Figure 4. Relation between maximum swelling pressure and dry density.

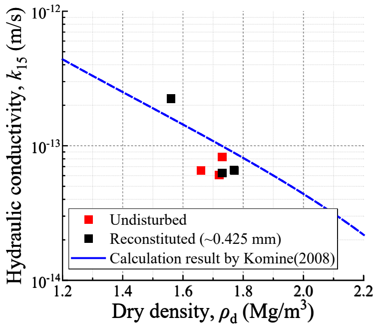

Figure 5. Relation between hydraulic conductivity and dry density.

4 MICROSTRUCTURAL OBSERVATION RESULTS BY SEM AND XRD

The differences in the shape and skeleton of the soil particles in the undisturbed and reconstructed specimens were observed using SEM (S-4500, Hitachi). When making observed sample, undisturbed and reconstituted specimens (Ø=28 mm × h=10 mm) of similar dry density (ρ_d=1.7 Mg/m^3), and then the specimens were broken to expose fresh internal sections and formed to a size of 5 mm × 5 mm. Figure 6 shows appearance of SEM observation sample.

Figure 7 and Figure 8 show observation results of undisturbed and reconstituted samples, respectively. Undisturbed sample had a complex soil particle shape, and reconstituted sample had larger pores that were not seen in undisturbed sample. From these results, the more complex soil skeleton because of cementation in undisturbed specimens might affect lower water permeation.

Also, XRD measurements were performed on the undisturbed and reconstituted specimens after water absorption and swelling pressure measurements. Montmorillonite basal spacing and adsorption state of water molecules were evaluated from the peak shapes obtained by XRD. Details of this measurement and devices are described in Ito et al. (2022b).

Figure 9 shows peak shape comparison of undisturbed and reconstituted specimen at water content of 17.0% and 18.8%. From these results, in reconstituted specimen, peak of water molecule three-layer hydration (2θ=4.7 deg) is dominant, whereas in undisturbed specimen, two-layer hydration peak (2θ=5.8 deg) remains beside the three-layer hydration peak. Undisturbed specimen has fewer water molecules between montmorillonite crystalline layers because of cementation. In undisturbed specimens, pore outside montmorillonite interlayer remains, rather than only

Figure 6. SEM observation sample.

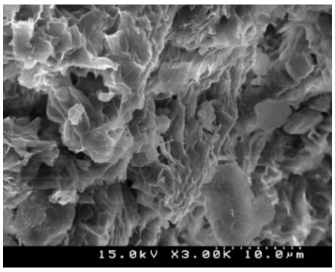

Figure 7. Observation on undisturbed sample (×3000).

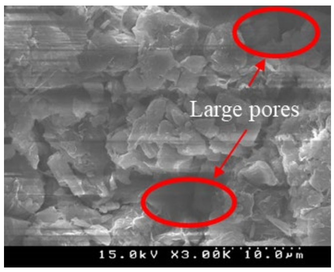

Figure 8. Observation on reconstituted sample (×3000).

between the montmorillonite layers which dominate the low permeability of bentonite. The combination of two factors, which complexity of skeleton and inhibition of swelling of montmorillonite layers, is assumed to be the reason why the permeabilities of the undisturbed and reconstituted specimens were not significantly different.

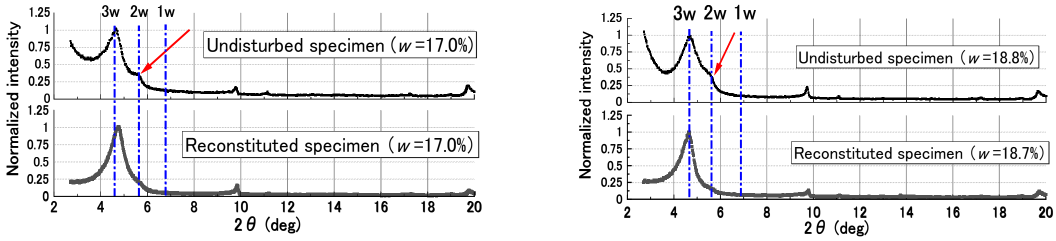

Figure 9. XRD peak shape comparison of undisturbed and reconstituted specimen at water content of 17% and 18.8%.

5 CONCLUSION

1. Because of cementation, swelling pressure of undisturbed specimen was almost half of that of reconstituted specimen, on the other hand, hydraulic conductivity was in the same order.
2. From SEM observation, undisturbed specimen has more complex soil particle structure. From XRD analysis, montmorillonite interlayer in undisturbed specimen is inhibited swelling and has less water molecules. From these factors, while undisturbed specimens decrease in swelling, the hydraulic conductivity remained almost the same.

ACKNOWLEDGMENTS

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