Graduate School of Creative Science and Engineering Waseda University

博士論文概要 Doctoral Dissertation Synopsis

論 文 題 目 Dissertation Title

Evaluation of underground environment effects on hydraulic properties and swelling pressure of bentonites in HLW disposal

高レベル放射性廃棄物処分におけるベントナイトの浸透特性と膨潤圧に 及ぼす地下環境の影響評価

> 申 請 者 (Applicant Name) Kunlin RUAN 阮 坤林

Department of Civil and Environmental Engineering, Research on Geotechnical Engineering

Handling high-level radioactive waste from nuclear power industry had been an urgent issue. Deep geological disposal method, which tries to seal waste deeply underground, was adopted by many countries to treat nuclide waste. In a Japanese deep geological disposal project, a multi-barrier system, including vitrificated waste, stainless container, buffer material and natural rock, would be constructed more than 300m underground. Bentonite was chosen as buffer material due to its low permeability. Bentonite has high self-healing capacity, which can fill cracks in multi-barrier system after contacting water. Swelling pressure and hydraulic properties, including water diffusivity and hydraulic conductivity, were adopted to evaluate self-healing capacity and permeability of bentonite, respectively.

It is necessary to assess swelling pressure and hydraulic properties under the effect of underground environment. Hundreds or even thousands of years are needed for high-level radioactive waste to decay fully. During such a long period, ground water would intrude consistently into buffer material. A great amount of heat would be emitted until radioactive waste decays fully. Japanese deep geological project was discussed to be cited in coastal area because of the feasibility of transportation by ship. This thesis focused on the water chemistry and temperature effects on swelling pressure and hydraulic properties of bentonites in saturation process. Additionally, X-ray diffraction tests were carried out to make sense the microstructural changes under different influencing factors.

This thesis was divided into 11 chapters.

In Chapter 1, Chapter 2 and Chapter 3, backgrounds, materials and testing devices were carefully interpreted, respectively;

In Chapter 4, swelling pressures and water content distributions of bentonites during saturation process were achieved with a developed swelling pressure device and a new multi-ring. Basal spacings were got by sending slices from multi-ring to X-ray diffraction tests. By bringing the relation between volumetric water content and new parameter χ into traditional Darcy equation, an easy equation for calculating water diffusivity was proposed. With the relation between water diffusivity and hydraulic conductivity, hydraulic conductivity during saturation process was obtained. It was found that, water diffusivity decreases consistently as the volumetric water content increases. With the variation of volumetric water content, hydraulic conductivity exhibits a "∩" shape. This phenomenon may because of the montmorillonite microstructure evolution during hydration process;

In Chapter 5, three dry densities for bentonites were adopted to investigate the effect of dry density on swelling pressure, hydraulic properties and basal spacing during saturation. Swelling pressures and water diffusivities during saturation

process were discovered increasing with the rise of dry density. Hydraulic conductivity and basal spacing under wetting were found decreasing as dry density increases. Smaller basal spacing in higher dry density condition induces larger DDL repulsive force, thereby leading larger swelling pressure. Water flowing channel outside inter-layer space would be compressed as basal spacing rises, giving smaller hydraulic conductivity in high dry density specimen;

In Chapter 6, NaCl solutions with different concentrations were used as saturation liquids for researching the concentration effect on sodium type bentonite swelling pressure, water diffusivity and basal spacing during saturation process. It was discovered that, swelling pressure and basal spacing decrease as concentration rises. Water diffusivity was found increasing as concentration rises. Smaller basal spacing gives greater water flow channel in higher concentration condition, leading greater water diffusivity. Negative effect from larger osmotic suction conquers positive effect from smaller basal, bringing smaller swelling pressures in higher concentration condition;

In Chapter 7, movements of Na⁺ and water were studied by infiltering bentonites with NaCl solutions. The results showed that the movement of Na⁺ has a certain lag to the movement of water in compacted sodium bentonite. This may be due to the fact that there is no medium for Na⁺ movement until water totally occupies inter-layer space;

In Chapter 8: NaCl and CaCl₂ solutions were used as saturation liquids for seeking the saturation liquid cation effect on swelling pressure, water diffusivity and basal spacing during saturation. For sodium type bentonites, CaCl₂ saturated specimen has larger swelling pressure, water diffusivity and basal spacing than NaCl saturated ones. The cation exchange between the Ca²⁺ in saturation liquid and the Na⁺ in montmorillonite mineral may give positive impact on swelling pressure, water diffusivity and basal spacing. In case of calcium type bentonite, NaCl has larger swelling pressure, water diffusivity and basal spacing instead of CaCl₂. At the same concentration, greater osmotic suction from CaCl₂ would bring negative impact on swelling pressure, water diffusivity and basal spacing;

In Chapter 9, swelling pressures and water diffusivities during saturation of bentonites were obtained by putting swelling apparatus into ovens with different temperatures. In case of powder bentonites, the decrease of basal spacing in higher temperature condition would bring larger DDL repulsive force, thereby inducing larger swelling pressures in higher temperature. For particle bentonite, back stress from the expansion of free water conquers the growth of DDL repulsive force from reducing basal spacing, bringing smaller swelling pressures as temperature rises;

In Chapter 10, bentonite with different water contents were achieved by spraying water and controlling the relative humidity. Effects of initial water content and specimen preparation method on swelling pressures were researched. Another individual set of X-ray diffraction test was conducted to make sense the effects of initial water content and specimen preparation method on microstructure of bentonite. The investigations indicated that initial water content and specimen preparation method have no effect on equilibrium swelling pressure. By XRD investigations, it can be easily concluded that, initial water content has no effect on basal spacing of saturated specimen, which may indicate similar microstructure in saturate state. Equilibrium swelling pressure would be similar at different initial water contents due to the similar saturated microstructure. The mutual compensation of swellings between layers and particles results in the similarity equilibrium swelling pressure from two-specimen preparation methods;

In Chapter 11, conclusions and suggestions for the application of bentonite by the testing results were illustrated. High dry density compacted bentonite should be applied in multi-barrier system for obtaining enough swelling pressure and low hydraulic conductivity. It is better to use calcium type bentonite as buffer material for reducing negative impact on swelling pressure saturated by Na⁺ dominating liquids. Powder bentonite should be used instead of particle bentonite for avoiding the decrease of swelling pressure as temperature rises.

Observations from this thesis can be concluded as the followings:

- 1. Swelling pressure increases as dry density increases, since smaller basal spacing brings larger DDL repulsive force. Hydraulic conductivity decreases as dry density increases, as water flowing channel is compressed as dry density rises;
- 2. Ca²⁺ saturated sodium bentonites have larger swelling pressures than Na⁺, because positive effect from cation exchange conquers reducing DDL repulsive force from greater basal spacing. For calcium bentonite, Na⁺ obtains larger swelling pressure than Ca²⁺, as Ca²⁺ gives larger osmotic suction;
- 3. Swelling pressures of powder bentonites rise as temperature grows and swelling pressures of particle bentonite decrease as temperature rises. This phenomenon may be due to the back stress from free water volume expansion. Larger water diffusivity was found in higher temperature condition as increasing water viscosity;
- 4. High dry density compacted bentonite is recommended as buffer material for obtaining enough swelling pressure and lower hydraulic conductivity. From the aspect of swelling pressures, experimental results from this study gives reference database for the design of Japanese deep geological disposal project.

List of research achievements for application of Doctor of Engineering, Waseda University

Full Name: 阮 坤林 seal or signature

	Date Submitted(yyyy/mm/dd): 2022/12/2
種類別	題名、 発表・発行掲載誌名、 発表・発行年月、 連名者(申請者含む)
(By Type)	(theme, journal name, date & year of publication, name of authors inc. yourself)
Paper O	Kunlin Ruan, Hideo Komine, Hailong WANG, Daichi Ito. 2022. "Experimental study of initial water content and specimen preparation method effects on Kunigel-V1 bentonite swelling pressures", Canadian Geotechnical JournalIn press, https://doi.org/10.1139/cgj-2021-0532
0	Kunlin Ruan, Hailong WANG, Hideo Komine, Daichi Ito. 2022. "Experimental study for temperature effect on swelling pressures during saturation of bentonites", Soils and Foundations, 62(6): 101245.
О	Kunlin Ruan, Hideo Komine, Hailong WANG, Daichi Ito, Takahiro Gotoh. 2022. "Microstructural changes and swelling pressure of low dry density compacted bentonites during saturation combining X-ray diffraction", Canadian Geotechnical Journal. In press, https://doi.org/10.1139/cgj-2020-0342
О	Kunlin Ruan, Hideo Komine, Daichi Ito, Kentaro Miyoshi, Takahiro Gotoh. 2022. "Hydraulic Conductivity and X-ray Diffraction Tests of Unsaturated Bentonites with a Multi-ring and Their Predictions by Pores Distributions", Engineering Geology, 306: 106738
О	Kunlin Ruan, Xianlei Fu. 2022 "A Modified Kozeny-Carman Equation for Predicting Saturated Hydraulic Conductivity of Compacted Bentonite in Confined Condition", Journal of Rock Mechanics and Geotechnical Engineering, 14 (3): 984-993.
О	Hailong WANG, <u>Kunlin Ruan</u> , Satoru Harasaki, Hideo Komine. 2021. "Effects of specimen thickness on apparent swelling pressure evolution of compacted bentonite", Soils and Foundations, 61(1): 101099.
О	Hailong WANG, Daichi Ito, Takumi Shirakawabe, <u>Kunlin Ruan</u> , Hideo Komine. 2022. "On swelling behaviors of a bentonite under different water contents", Geotechnique, Ahead of Print. https://doi.org/10.1680/jgeot.21.00312
Conferences	
0	Kunlin Ruan, Dean Sun, Xianlei Fu, Hailong Wang, Hideo Komine, Daichi Ito. 2021/10. "Reversibility of micro and macrostructure in compacted bentonite under chemo-mechanical couplings". 3rd International Symposium on Coupled Phenomena in Environmental Geotechnics. Kyoto university, Kyoto, JAPAN.
О	Kunlin Ruan, Dean Sun, Hailong Wang, Hideo Komine, Daichi Ito. 2020. "Effects of Calcium Carbonate on hydro-mechanical properties of Bentonite". The 55th annual meeting of the Japan national conference on geotechnical engineering. Kyoto, JAPAN. [22-1-3-05]
О	Kunlin Ruan, Daichi Ito, Hideo Komine, Hailong Wang. 2021. "Tests for pores of Kunigel-V1 compacted bentonite during saturation". The 56th annual meeting of the Japan national conference on geotechnical engineering. Yamagata, JAPAN. [13-4-4-02]
О	Kunlin Ruan, Daichi Ito, Hideo Komine, Hailong Wang. 2021. "Swelling pressure and water diffusivity of sodium bentonite saturated by different solutions under confined wetting". 76 th Annual Meeting of the Japan Society of Civil Engineers. Kanagawa, JAPAN. [CS2-38]
Research fund	
О	Kunlin Ruan. "Effects of ions on swelling pressures of compacted bentonites combining X-ray diffraction", 2021-2022, B2R101231001, W-Spring, Japan Science and Technology Agency (JST).
	<u>l</u>