

Microstructure formation in bentonite

Formation microstructure en Bentonite

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ABSTRACT

Bentonite, a smectite clay, is widely used in geotechnical engineering as drilling mud, liner material, and it has been proposed as buffer and backfill material for high-level nuclear waste disposal. An understanding of the microstructure formation of bentonite and its hydrophobic properties would enable the development of a methodology capable of predicting its behaviour when exposed to water. This study aims to gain an understanding of the microstructure formation of bentonite, specifically Kunigel-VI, by comparing its behaviour with that of a non-swelling clay, NSF Clay. Three different tests were performed to observe this behaviour: Constant Rate of Strain (CRS) consolidation testing, Mercury Intrusion Testing (MIT), and Scanning Electron Microscopy (SEM). Results indicated that aggregate formation occurred in both clays, but at water contents higher than 50 %, the aggregates in NSF Clay collapsed, while those of Kunigel-VI increased in number and size. An increase in applied pressure resulted in the dissipation of air, and a decrease in size of the intra and inter-aggregate spaces.

RÉSUMÉ

La Bentonite, une argile smectique, est couramment utilisée dans le domaine du génie géotechnique comme fluide de forage, doublure étanche, et a même été proposée comme tampon et matériaux de remplissage pour entrepôt de déchets nucléaire de haut niveau. Comprendre la formation microstructure du Bentonite et ses propriétés hydrophobes, permettrait le développement d'une méthodologie capable de prédire son comportement, lors d'une exposition à l'eau. Cette étude cherche à comprendre la formation microstructure du Bentonite, essentiellement de type Kunigel-VI, en comparant son comportement à celui d'une argile non expansive de type "NSF Clay" (non swelling clay). Trois tests différents ont été réalisés pour observer ce comportement: CRS (Constant Rate of Strain) qui s'agit d'un test de déformation à allure constante voué à évaluer la consolidation; MIT (Mercury Intrusion Testing) test de porosité à l'aide d'un porosimètre à mercure; et MEB Microscopie électronique à balayage (SEM en anglais). Les résultats ont montré qu'une formation granulaire avait lieu dans les deux argiles; par contre à des taux d'humidité supérieur à 50%, le granulat de l'argile NSF s'effondrait, alors que celle de type Kunigel-VI augmentait en nombre et en taille. Une augmentation de la pression appliquée avait comme résultat la dissipation de l'air, et une diminution en taille de l'espace intra- et inter-granulat.

Keywords : bentonite microstructure, CRS testing, mercury intrusion testing (MIT), scanning electron microscopy (SEM)

1 INTRODUCTION

The disposal/storage of radioactive waste is currently being addressed around the world. Facilities are designed to store radioactive waste containers, also known as canisters, several hundreds of meters below ground level in intact rock surrounded by buffer material. The basic function of the buffer material is to keep the containers in place and to create a low permeability layer around the canisters. This allows for the waste to be isolated from the surrounding environment, limiting the access of water to the containers, preventing the migration of waste materials, and transferring the heat from the nuclear waste to the host rock (Tripathy et al. 2004). Engineered clay barriers have been proposed as suitable buffer materials as they provide "low hydraulic conductivity, low ion diffusivity, [and] good self-sealing capacities" (Marcial et al. 2002). Compacted bentonite and bentonite-sand mixtures have been proposed as suitable buffer materials since they not only meet the outlined requirements, but also have the ability to adsorb and prevent the migration of radionuclides.

It is the swelling characteristics of bentonite that make it an attractive choice as a buffer material. With the uptake of water, compacted bentonite/bentonite-sand mixtures are expected to swell and seal the spaces. Additionally, should any cracks form in the host rock, bentonite may migrate into the cracks, sealing them, thus maintaining the low permeability zone around the canisters. The permeability of this zone is determined by its

hydraulic conductivity, which, according to Achari et al. (1999), is "directly influenced by the agglomerates of clay particles." Therefore a comprehensive understanding of the microstructure formation of bentonite would be beneficial in the design of these facilities.

2 EXPERIMENTAL STUDY

The aim of this ongoing study is to gain an understanding of the microstructure formation of bentonite by comparing its behaviour with that of a non-swelling clay. Three different tests have been performed to observe the bentonite's behaviour: Constant Rate of Strain (CRS) consolidation testing, Mercury Intrusion Testing (MIT), and Scanning Electron Microscopy (SEM).

2.1 Materials/specimens

Two types of clay were used in this study. Kunigel-VI is a commercial sodium type bentonite, containing 48 % montmorillonite (smectite mineral) and quartz (non-swelling mineral), from Yamagata Prefecture, Japan (Komine 2004). NSF Clay is a commercial non-swelling clay from Okayama Prefecture, Japan and is composed of illite (clay mineral), and contains traces of quartz and feldspar. The particle size distribution for both clays indicates that approximately 65 % of

Table 1: Material Properties

Property	Kunigel-VI	NSF Clay
G_x (g/cm ³)	2.799	2.760
Natural Water Content	5.0 - 6.2 %	0.2 - 0.5 %
Liquid Limit (LL)	499 %	55 %
Plastic Limit (PL)	39 %	29 %

the particles are smaller than 2 μm . The material properties of both clays are presented in Table 1. Samples were prepared at varying gravimetric water contents, ranging from natural to a water content of 70 %. Distilled water was sprayed onto the soil to create natural aggregates, resulting in non-homogenous samples. The objective of using distilled water in this study was to maintain consistent water chemistry conditions. Samples were prepared 24 hours prior to consolidation testing, to allow for aggregate formation.

2.2 Testing Methodology

The CRS testing assesses the compression behaviour of clay, and the effect of water on its behaviour. The compression behaviour is in turn affected by the microstructure, thus the difference in compression indicates a difference in the microstructure. CRS testing was carried out on samples with an approximate thickness of 20 mm. The samples were loaded under a constant strain ranging from 0.02 to 0.1 mm per minute, and pressures ranging from 200 kPa to 15 MPa. Additional water was introduced into the loading cell following CRS testing in selected samples, resulting in sample submergence. MIT and SEM samples were compressed using the CRS testing apparatus, following which they were freeze-dried and placed in a vacuum for 24 hours prior to testing. MIT was used to determine the pore size distribution in a sample, while SEM allowed viewing of the sample's microstructure.

3 RESULTS AND DISCUSSION

The following describes the results of the testing program. The data discussed here pertains to selected samples from this study. However, extensive testing has been performed on additional samples, which have displayed similar results. Table 2 summarizes water contents for the selected samples tested for this study.

3.1 Constant rate of strain (CRS) consolidation

Figure 1 shows selected CRS test results for both Kunigel-VI and NSF Clay; with samples selected to show the general trends observed in this study. The difference in initial void ratio between samples with similar water contents was due to the initial sample conditions; namely whether or not the sample was manually compressed in the loading cell prior to testing. Therefore, different initial conditions were tested to determine whether or not it influenced the final compression behaviour of the clay.

Figure 1a shows the effects of water on the compression behaviour of Kunigel-VI. Increasing the sample water content resulted in an increase in the void ratio, and a greater difference in the initial void ratio within the same range of water content. The addition of water to dry samples resulted in the formation of microstructures surrounding the water molecules. Aggregate formation also resulted in the introduction of air into the system. Aggregates formed surrounding the water molecules, and air filling the spaces between the aggregates resulted in the creation of micro- and macro-pores. The higher the water content, the larger the number and size of aggregates formed, while swelling of the aggregates may also occur. An increase in water content also resulted in a decrease in the pre-consolidation pressure, with values ranging from 150 kPa at lower water contents to 10 kPa at 70 % water content.

Table 2: Sample Data

Sample Name	w_i (%)	w_f (%)
B-natural - 1	6.74	12.89
B-natural - 2	5.97	12.69
B-50% - 1	54.60	57.96
B-50% - 2	55.37	43.72
B-70% - 1	89.49	75.00
B-70% - 2	78.82	90.00
NSF - natural	0.32	0.71
NSF - 30% - 1	33.00	37.44
NSF - 30% - 2	32.30	32.64
NSF 50%	47.94	40.65

*where 2 samples exist, the first is the one with the larger void ratio

Increasing the pressure resulted in a decrease in void ratio. The formation of an S-shaped curve, with a higher slope, or compression index was observed in samples containing higher water contents. Applying pressure causes the collapse of structures formed within the clay, seen as the crushing of the aggregates. This compression behaviour results in the dissipation of air from both the macro- and micro-pores; however, the water is not dissipated, but rather moved from the macro- to the micro-pores. At approximately 150 kPa, it is believed that the macro aggregate samples with water content of 50% and 70 % collapsed. However, the water present in the micro-pores limited the amount of compression possible, which was observed in the decrease of the consolidation rate.

Figure 1b presents similar results for dry NSF Clay samples as were observed for dry Kunigel-VI samples. Increasing the water content to 30 % resulted in an initial void ratio increase due to the formation of aggregates around water molecules, and a decrease in the pre-consolidation pressure to approximately 20 kPa. However, unlike Kunigel-VI, increasing the water content to 50 % resulted in a decrease in the initial void ratio. This initial void ratio for 50% water content decreased below the void ratio observed for dry samples; however, the curve trend resembled that of the dry sample. This increase in water content resulted in the collapse of the aggregates, rearrangement of particles, and formation of a more homogenous and anisotropic sample. A saturated NSF Clay sample was tested, and the same trends described above were observed, suggesting that the 50 % water content sample is at or near saturation.

The CRS testing performed allows water to drain out of the sample. The permeability values of Kunigel-VI are on the order of $\sim 10^{-11} - 10^{-12}$ cm/s, much lower than the permeability values of NSF Clay, which are on the order of $\sim 10^{-6}$ cm/s. Therefore water drainage in Kunigel-VI samples may occur at an appreciably slower rate than NSF clay, and may not be observed in the testing performed. During compression most of the air is squeezed out; however, it is possible that small amounts of air might have existed in the microstructures which could not be expelled without the application of a larger pressure. No water dissipation was observed in Kunigel-VI samples, as evidenced by the negligible difference in initial and final water contents shown in Table 2. The variations between the initial and final water contents can be due to sample non-homogeneity, which resulted in an unequal water distribution within a sample. NSF Clay samples on the other hand experienced no sharp decreases in void ratio, and during testing, water dissipated from the sample and gathered on the surface. Additionally, water dissipation was evident by the large variations in water content in the 50 % water content sample in Table 2.

Figure 2 shows the maximum compression index (C_c) normalized according to Terzaghi's equation for remoulded clays (1) versus the initial water content (w_i), normalized by the liquid limit (LL), for Kunigel-VI and NSF Clay samples.

$$C_c = 0.009(LL - 10) \quad (1)$$

From Figure 2, it can be observed that increasing the water content resulted in an increase in C_c for both clays. This suggests that at higher water contents larger aggregates are formed, from where air is more readily expelled, and the aggregates collapse and/or are deformed when compressed. Increasing the water content to 50 % ($w_i/LL \approx 0.008$) resulted in a decrease in C_c for NSF Clay, suggesting that at these higher water contents the structures are smaller or non-existent due to saturation, and the particles become horizontally oriented, thus decreasing the compression index. At higher water contents than those tested to date, a similar decrease in the compression index is expected to be observed in Kunigel-VI as was observed in NSF Clay.

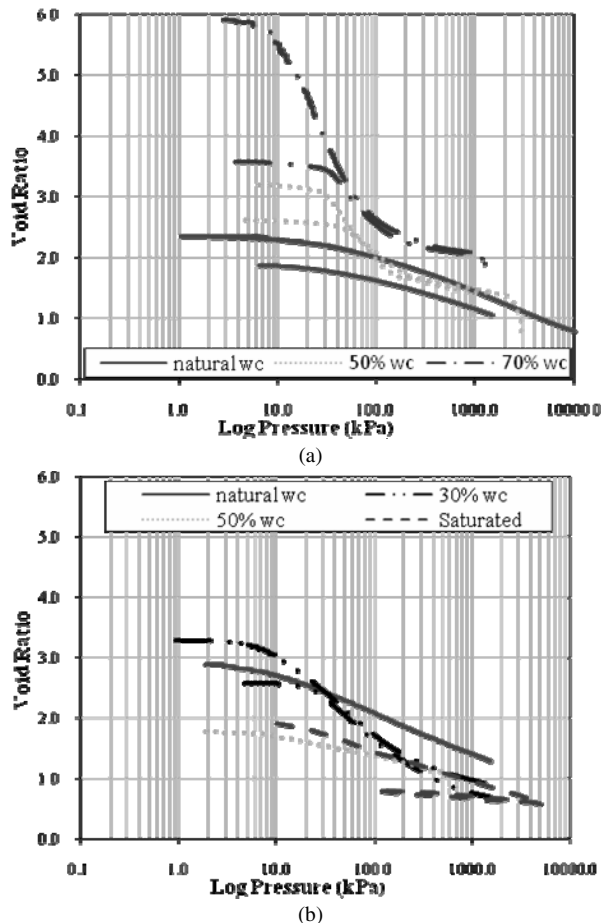


Figure 1: CRS testing results for a) Kunigel-VI and b) NSF Clay

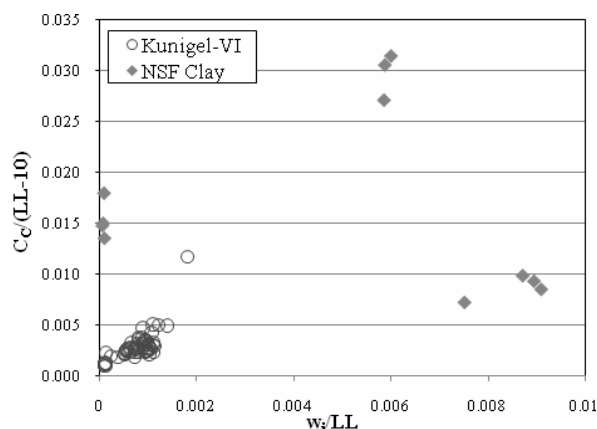


Figure 2: Compression Index (C_c) vs. Initial Water Content (w_i)

3.2 Mercury Intrusion Testing (MIT)

Figure 3 shows the MIT results for Kunigel-VI and NSF Clay, respectively. At low pressure (200 kPa) a higher volume of larger sized pores was observed in Kunigel-VI (Figure 3a), with the amount of voids and pore entrance diameters being larger for 50 % samples than for 30 % samples, corresponding to the CRS test results. An increase in pressure resulted in a decrease in pores as seen by the bimodal curve centered at 0.5 μm and 0.8 μm for 30 % water content samples and 0.3 μm and 0.5 μm for 50 % water content samples. Delage et al. (2006) stated that for compacted soils, a bimodal curve indicates the presence of an aggregate microstructure, with the modes showing the intra and inter-aggregate pore spaces. The presence of the semi-bimodal curves in this study suggested that the difference between the intra and inter-aggregate pore spaces was not as large as expected or could not be observed for the specimens in this study. Submergence of the samples caused a rearrangement of the particles, resulting in a smaller quantity of smaller sized pores, as seen by the shift in the MIT curves for both 30 % and 50 % water content at 200 kPa. It is believed that once in contact with the water molecules, the particles rearrange themselves, resulting in smaller pores, contributing to the difference in curves between the 30 % and 50 % samples at 200 kPa, and the submerged samples at 200 kPa. Both types of samples underwent the same CRS testing procedure; however, water was introduced into the loading cell following CRS testing for the submerged samples.

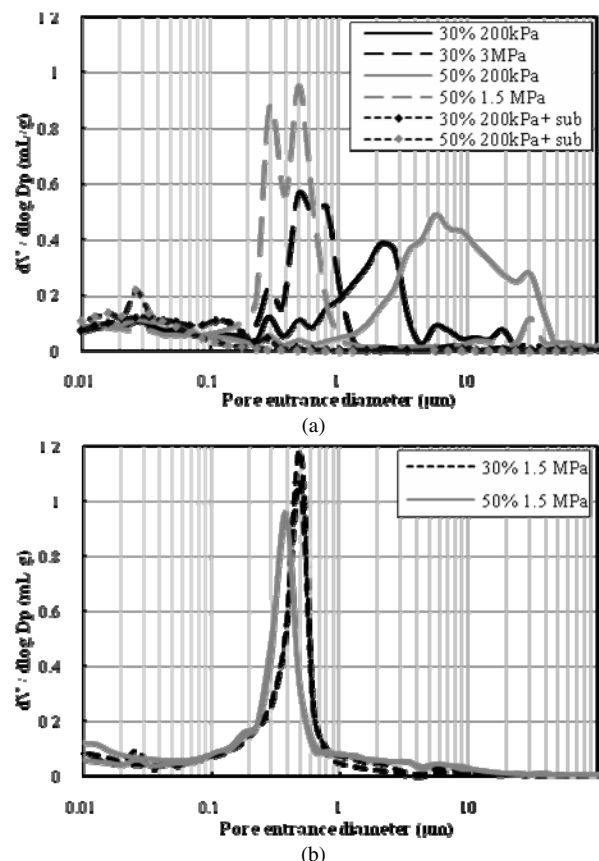


Figure 3: MIT Results for a) Kunigel-VI b) NSF Clay

NSF Clay shows a unimodal curve centered at 0.35 μm and 0.5 μm for samples at 50 % and 30 % water contents at 1.5 MPa respectively as seen in Figure 3b. No other pressures have been tested to date. The unimodal curves suggest the presence of similarly sized voids, which are smaller in size and total volume at higher water contents, correlating to the lower

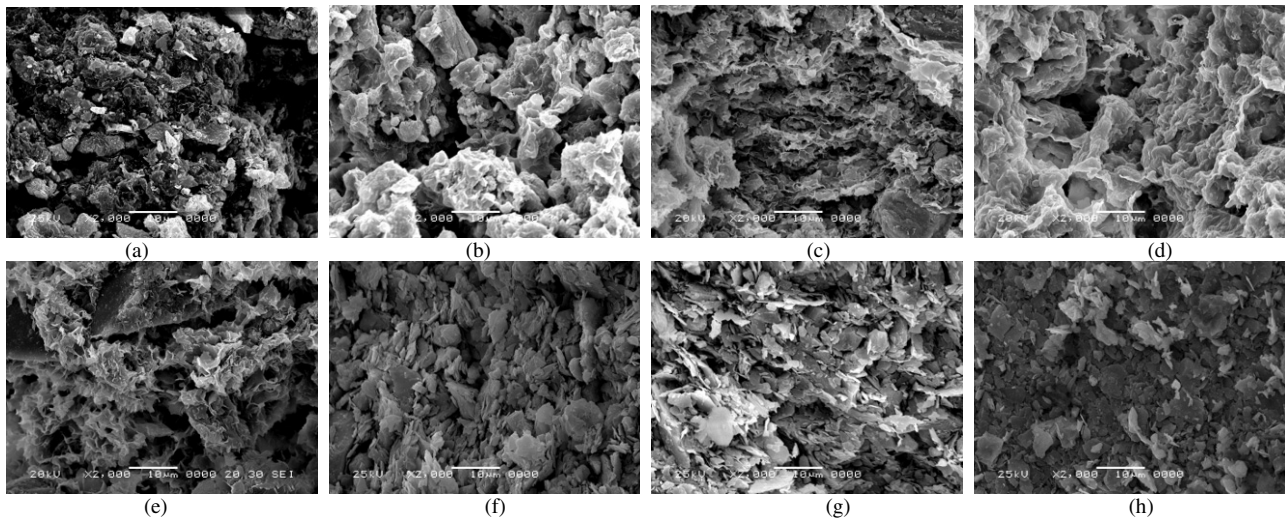


Figure 4: SEM results for Kunigel-VI. a) dry, 10MPa b) 30%, 0.2MPa c) 30%, 3MPa d) 50%, 0.2MPa e) 50%, 3MPa and for NSF Clay f) dry, 15MPa g) 30%, 1.5 MPa h) 50%, 1.5 MPa

initial void ratio for CRS testing for samples at 50 % water content.

3.3 Scanning Electron Microscopy (SEM)

Figure 4 presents the SEM results for Kunigel-VI and NSF Clay. Unfortunately wet samples could not be viewed in the SEM, thus samples had to be freeze-dried. All samples have the same magnification, with the white bar indicating a scale of 10 μm . The Kunigel-VI sample results are shown in Figure 4a through f. Figure 4a shows a sample at natural water content and at 10 MPa pressure. From the image it can be seen that at a natural water content the particles are aggregated. An increase in water content causes a change in the aggregates, which is believed to be a result of both increased aggregation, and an increase in the swelling of existing aggregates. This change can be seen by the particle alignment and voids present in Figure 4b and d. An increase in pressure at higher water contents resulted in the realignment of particles and the collapse of the aggregates as shown in Figure 4c and e. Particle alignment can especially be observed in Figure 4c, with a decrease in size of pores observed in both figures.

NSF Clay samples are shown in Figure 4f to h. As with Kunigel-VI, the dry sample shown in Figure 4f shows that at a natural water content NSF Clay is aggregated. An increase in water content to 30 % results in an increase in aggregation, as seen by the presence of voids and the alignment of particles in Figure 4g. A further increase in water content to 50 % produces samples that are near or at saturation, and that are more homogenous in nature, resulting in the parallel alignment of particles shown in Figure 4h, where void spaces were still observed but at a smaller scale and number.

4 SUMMARY AND CONCLUSIONS

Bentonite microstructure was observed in this study through a series of laboratory tests on Kunigel-VI and NSF Clay, swelling and non-swelling clays, respectively.

In natural conditions, both Kunigel-VI and NSF Clay were observed to be in an aggregated state, as seen by SEM imaging shown in Figure 4a and f. Increasing the water content resulted in an increase in aggregate size, as observed by the initial water content increase in the CRS results (Figure 1a and b), and the SEM imaging in Figure 4a, b, and c for Kunigel-VI and Figure 4f to h for NSF Clay. An increase in water content to 50 % for NSF Clay resulted in homogenous samples, and a decrease in the initial void ratio (Figure 1b) and compression index (Figure

2). Particle rearrangement was also observed at 50 % water content, and can be seen in SEM imaging, Figure 4h. The submergence of Kunigel-VI samples resulted in the rearrangement of particles into a horizontal orientation. This can be observed by the shift in MIT curves for samples containing large volume of large sized pores (200 kPa), to samples containing small volume of small sized pores (200 kPa + sub) as presented in Figure 3a.

An increase in pressure caused a rearrangement of the particles and a collapse of the aggregates, resulting in smaller pores. This can be seen by the decrease in void ratio with an increase in pressure from the CRS test results shown in Figure 1a and b, and a decrease in the size and total volume of pores in MIT testing shown in Figure 3a. SEM imaging also showed particle realignment and a decrease in pore size with an increase in pressure in Kunigel-VI samples (Figure 4c and e).

Achari et al. (1999) have shown the influence of aggregates on the hydraulic conductivity of clays. A collapse of aggregates can result in a decrease in permeability, as the pores decrease in size and number. This study has shown that an increase in water content resulted in an increase in aggregation, and the swelling of existing aggregates. This caused an increase in size and total volume of pores, which can result in an increase in permeability. Further increase in water content to the point of saturation, in the form of submergence in Kunigel-VI samples, resulted in particle realignment, which decreases both the size and total volume of pores, decreasing the sample's permeability. An increase in pressure resulted in the collapse of aggregates, reducing the pore volume and size, also reducing the permeability. Thus it is believed that an understanding of the microstructure in bentonite would allow for a better prediction of its behaviour when used as a buffer material in nuclear waste disposal, specifically the effects of water and pressure.

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REFERENCES

- Achari, G. Joshi, R.C. Bentley, L.R. & Chatterji, S. 1999. Prediction of the hydraulic conductivity of clays using electric double layer theory. *Canadian Geotechnical Journal* 36:789-792.

- Delage, P. Marcial, D. Cui, Y.J. & Ruiz, X. 2006. Ageing effects in a compacted bentonite: a microstructure approach. *Géotechnique* 56(5): 291-304.
- Komine, H. 2004. Simplified evaluation for swelling characteristics of bentonites. *Engineering Geology* 71: 265-279.
- Komine, H. & Ogata, N. 1996. Prediction of swelling characteristics of compacted bentonite. *Canadian Geotechnical Journal* 33: 11-22.
- Marcial, D. Delage, P. & Cui, Y.J. 2002. On the high stress compression of bentonites. *Canadian Geotechnical Journal* 39: 812-820.
- Tripathy, S. Sridharan, A. & Schanz, T. 2004. Swelling pressures of compacted bentonites from diffuse double layer theory. *Canadian Geotechnical Journal* 41: 437-450.