Microstructure of saturated bentonites characterized by X-ray CT observations

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A B S T R A C T

Bentonite is a very attractive raw material as a compacted clay liner at general waste disposal sites and also as a buffer material at radioactive waste disposal sites. It has been considered that the permeability of bentonite, the most important index for evaluating barrier materials, is closely related to its microstructure. Recent studies in geological material science show that X-ray computerized tomography (X-ray CT) is a very powerful tool for the study on microstructure and hydro-osmotic phenomena (e.g. Wong and Wibowo [Wong, R., Wibowo, R., 2000. Tomographic evaluation of air and water flow patterns in soil column. Geotech. Test. J. 23, 413–422]). Permeability tests and micro X-ray CT observations of Wyoming bentonite were performed to describe the relationship between microstructure and permeability of the bentonite used as a barrier material. Two types of samples, compacted bentonite-quartz sand mixtures and raw bentonite ores, were used in this study.

The X-ray CT observations of the bentonite-quartz sand mixtures show that ‘vacant pores’ and ‘bentonite-water complexes’ of the bentonite samples after water permeation are distinguishable in X-ray CT images, and that the micro-structural differences are closely relating to the sample permeability, and depend on the mixing and saturation conditions. Permeability tests and X-ray CT observations of the bentonite ore samples show that the permeability and the microstructure are independent to the sedimentary texture developed within the ore samples. In addition, it is characteristic that the bentonite ore samples with micro-cracks show low hydraulic conductivity, comparable to the compacted powder bentonite, implying that cracks in the sample are filled with ‘bentonite-water complexes’ formed after permeation.

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1. Introduction

Many general waste disposal sites in Japan are situated in mountain areas, and require the provision of liner sheets of high sealing ability. For this clay liner sheets in combination with geo-membranes are usually employed at officially controlled landfill sites. For clay liners, bentonite is one of the most commonly used materials at disposal sites and it is well known that the sealing ability of bentonite lining is affected by the mixing with soil during the construction, and/or affected by the properties of leachates after construction (Kenney et al., 1992). To assess the sealing ability of bentonite liners, it is important to understand the microstructure controlling the hydraulic conductivity of the components of the liner compounds.

Bentonite is also commonly considered a candidate for buffers at radioactive waste disposal sites in Japan (e.g. Komine and Ogata, 1999). The use of the bentonite to put as a buffer varies depending on the waste disposal site/type, whether the site is designed for high-level waste (HLW) or low-level waste (LLW) (e.g. JNCTN1400 99-020, 1999). Examples of the manner of construction with the buffer materials is the compacted block method or in-situ compaction as has been proposed in Japan (Chijimatsu et al., 1999), compaction methods that are thought to be better suited for LLW sites. Sato and Suzuki (2003) has shown that the orientation of clay particles is affected by the mixing ratio of the bentonite components (i.e. smectite and quartz). Therefore, it is important to understand the relationship between the microstructure and hydraulic conductivity of the various bentonite components.

Recent studies in geological materials science has shown X-ray computerized tomography (X-ray CT) to be a very powerful tool to

Table 1
Experimental conditions of the X-ray CT.

<table>
<thead>
<tr>
<th>Scan type</th>
<th>Rotating only</th>
</tr>
</thead>
<tbody>
<tr>
<td>Power of the X-ray</td>
<td>90 kV</td>
</tr>
<tr>
<td>Current of the X-ray</td>
<td>89 μA</td>
</tr>
<tr>
<td>X-ray LL</td>
<td>10.16 cm</td>
</tr>
<tr>
<td>Matrix mesh</td>
<td>1024×1024</td>
</tr>
</tbody>
</table>
study microstructure and hydro-osmotic phenomena (e.g. Wong and Wibowo, 2000; Wildenschild et al., 2002), and X-ray CT has been widely used to evaluate the internal structure and swelling properties of bentonite liners (Kozaki et al., 2001; Nishiyama et al., 2004) as well as to provide information on diffusion coefficients and diffusion paths (Nakashima, 2000; Van Geet et al., 2005).

To further develop this line of research, X-ray CT observations of bentonites were made to understand the relationship between microstructure and hydraulic conductivity of the bentonites used both in controlled landfill sites and in LLW disposal sites. At present the microstructure is discussed based on the mixing conditions and the discussion of the saturation conditions uses the differences in the apparent orientations of layers of the bentonites.

2. Materials

2.1. Compacted bentonite–quartz mixture (CBM) sample preparation

Mixtures of 10 wt.% Wyoming bentonite powder (Na-type, Hojun Ben gel A) and 90 wt.% quartz sand (particle size 0.2–0.8 mm, Merck KGaA) were used for the compacted bentonite–quartz mixture (CBM).

This mixing ratio of bentonite is widely adopted in bentonite clay liners of landfills in Japan. After the bentonite powder and quartz sand were dried for 24 h at 110 °C with a temperature-controlled bath and cooled in a desiccator, they were mixed by hand and by a V-type mixing machine to compare the hydraulic conductivities of mixtures made with different mixing methods. After mixing the water content of the mixtures was adjusted to 10 wt.% The mixtures were compacted with a 40 MPa pressure in a 5 cm diameter and 2 cm high mold. These CBM samples were used in the permeability tests described in the next section. After the permeability tests the CBMs were cut into cubes (1 cm × 1 cm × 2 cm) for X-ray CT observations.

2.2. Bentonite ore sample preparation

Block and core samples of Wyoming bentonite ore originated from the Coronie bentonite deposit in the Black Hills, northeastern Wyoming, U.S.A. The Wyoming bentonite deposit was derived from the volcanic ash deposited in shallow marine seas in the Cretaceous period (Thorson, 1997). This bentonite deposit is arranged in gently sloping strata. This bentonite ore exhibits horizontal bedding, and cubic block samples (30 cm × 30 cm × 30 cm) and core samples of φ 3.8 cm were cut from the deposits, 90 cm below the surface of the deposit. The average water content and the average dry density of these samples are 45.5% and 1.13 g/cm³ respectively. Some secondary cracks perpendicular to the bedding plane, possibly arising after sampling, were observed in the block samples.

In the laboratory the bentonite block samples were drilled in the directions parallel and vertical to the bedding plane. The drilled cylindrical samples (φ 4 cm × 5 cm) were used in the permeability test to examine the effect of sedimentary structures like the bedding plane on the hydraulic conductivity of the bentonite block samples. After the permeability tests, cylindrical cores (φ 3.5 cm × 5 cm) of the block samples were used for X-ray CT observation.

The core samples were cut into cylinders (φ 3.8 cm × 5 cm) and used for permeability tests. After the permeability tests, the core samples were further cored to cylinders of φ 3.0 cm × 5 cm, and provided to X-ray CT observations.

3. Experiments

3.1. Permeability tests

The CBMs and bentonite ore samples were examined with a falling head type permeability tester and tested at room temperature. The
permeability tests of the CBMs were performed at atmospheric pressure to understand the effect of sample mixing on the permeability. The permeability tests were performed with distilled water for 1–14 days and 30 days; in the 30 day run the hydraulic head pressure was 0.06 MPa. In the bentonite ore samples, distilled water was infiltrated for about 500 days and the hydraulic head pressure was steadily increased to about 0.3 MPa.

The hydraulic conductivity was calculated by Eq. (1),

$$k = \frac{9.8 \times H \times Q}{\pi \times a^2 \times T \times P \times 10^5} \text{ (cm/s)}$$

where $k$: the hydraulic conductivity, $H$ (cm): the height of the mold, $Q$ (cm$^3$): the flow of the permeating water, $a$ (cm): the radius of the mold, $T$ (s): the penetration time, and $P$ (MPa): the hydraulic head pressure.

3.2. Micro X-ray CT observations

The non-destructive imaging system of X-ray computerized tomography (CT) was developed by Hounsfield (1973). In general, when the X-ray beam passes through a material, the X-ray energy is absorbed in proportion to the density of the material. The spatial characteristics of
the X-ray attenuation energy are obtained by the X-ray beam from various directions. The transmitted X-ray intensities of the material are converted into the projection data by calculation with an algorithm of the filtered back projection method. The X-ray attenuation coefficients of the material are converted into the CT numbers by using Eq. (2).

\[
\text{CT number} = \frac{\mu_i - \mu_w}{\mu_w} \times 1000
\]

(2)

where \(\mu_i\) and \(\mu_w\) are the X-ray attenuation coefficients for the material and water respectively. The X-ray attenuation coefficient depends on the atomic number and the density of the material. The CT numbers of air and water were set as \(-1000\) and 0, respectively.

The X-ray CT observations of the microstructure of the samples were performed using a Micro X-ray CT system with a spatial resolution of 5 \(\mu m\) (TOSCANER 30,000 \(\mu\)hd, TOSHIBA IT control system Co.). The experimental conditions for the X-ray CT observations are shown in Table 1. The X-ray CT scanning was performed on the central cylindrical part of the cubes and cylinders cut from the bentonite samples for about 20 min per one scan. For the X-ray CT scanning of the CBMs, an aluminum filter was positioned into the X-ray beam path to reduce the beam-hardening effect. About 250 2D images were obtained for each bentonite sample to create a 3D image of the sample.

The CT number of the 2D images was analyzed by Image J 1.36b developed by Rasband and Traces V5.1 by Dan Watson Software Team. The 3D images were generated by using the VG Studio MAX 1.2, Volume Graphics software. Petrophysical parameters such as porosity and pore diameter of the samples were quantified by Image-Pro Plus 5.0 by Media Cybernetics.

4. Results

4.1. Preliminary X-ray CT observations of the bentonite samples

Wong and Wibowo (2000) have indicated that it is difficult to distinguish the constituents of rock samples containing moisture. Therefore, the preliminary X-ray CT observations of the simulated CBM sample with moisture (bentonite:water = 2:25) was performed under optimal experimental conditions (Table 1), and the constituents of the sample such as bentonite–water complexes, quartz sand, and vacant pores are clearly distinguishable as shown in Fig. 1, after applying the filtered back projection method on the X-ray CT images. There are three areas showing different brightnesses in the 2D image and in order of decreasing brightness they are: quartz, bentonite–water complexes, and vacant pores (Fig. 1).

4.2. CBM

4.2.1. Hydraulic conductivity

The averaged hydraulic conductivities tested by using distilled water for the CBM mixed with the V-type mixing machine and the CBM mixed by hand are \(2.65 \times 10^{-10}\) m/s and \(5.91 \times 10^{-10}\) m/s respectively (Fig. 2). The hydraulic conductivity of the hand mixed sample is slightly larger than that of the machine mixed sample. The hydraulic conductivities with distilled water permeation are in agreement with Komine et al. (1991).

<table>
<thead>
<tr>
<th>Block samples</th>
<th>Core samples</th>
</tr>
</thead>
<tbody>
<tr>
<td>Direction of the water flow</td>
<td></td>
</tr>
<tr>
<td>Horizontal (parallel to bedding)</td>
<td>Vertical (perpendicular to bedding)</td>
</tr>
<tr>
<td>Averaged hydraulic conductivity (m/s)</td>
<td>(1.12 \times 10^{-14})</td>
</tr>
</tbody>
</table>

4.2.2. X-ray CT observations of the CBM with different mixing conditions

The CT images of the CBMs after water permeation show the differences in microstructure due to mixing conditions as shown in Fig. 3. The CT image of the CBM mixed with the V-type mixing machine shows that the bentonite–water complex is homogeneously distributed (the left image of Fig. 3), more so than the sample mixed by hand (the right image of Fig. 3). In addition the bentonite–water complexes in the hand mixed sample tend to form large mass assemblages. That is, the porosities of the samples mixed by hand and by the V-type mixing machine were 19.0% and 11.9% respectively, and the ranges of pore diameter of the two samples were 0.364–1.216 mm and 0.448–1.015 mm respectively. These petrophysical data suggest that the CBM mixed by hand is more porous than the one mixed by the V-type machine. Thus, the microstructure and the hydraulic conductivity of the CBM can be affected by the mixing conditions, and it indicates that the mixing condition of bentonite with sand must be specified in quality evaluations and quality control of CBMs used in actual construction.

4.2.3. X-ray CT observations of the CBM before and after permeation with distilled water

A comparison of the CT images of the CBMs before and after water permeation shows a visible change in brightness of the images, possibly due to differences in the cumulative areas of ‘vacant pore’, and ‘bentonite–water complex’ (Fig. 4). The CT number histograms of the X-ray scan data indicate differences in the CBMs before and after permeation (Fig. 5). The characteristic peak corresponding to CT numbers around \(-90\) observed in the histogram before permeation disappears in the histogram after permeation, but the peak at about \(-20\) in the histogram before permeation increases after the water permeation. This situation was observed in all the X-ray scan data in this study.

4.3. Bentonite ore samples

4.3.1. Hydraulic conductivity

The hydraulic conductivities of bentonite ore samples are shown in Table 2 (Sato et al., 2007). In the case of block samples, the average hydraulic conductivities of the laminate structure in the horizontal direction after water flow and in the vertical direction after water flow are comparative at \(1.12 \times 10^{-12}\) m/s and \(1.59 \times 10^{-12}\) m/s respectively. This shows that the hydraulic conductivity is not affected by the orientation of the laminate structure.

The average hydraulic conductivity of the core samples is \(2.35 \times 10^{-15}\) m/s, which is slightly different from the block samples, where the
two average hydraulic conductivities (horizontal and vertical) are similar. It may be understood that differences in cutting methods do not have a large impact on the hydraulic conductivity.

4.3.2. X-ray CT observations of the block samples
Images of block samples infiltrated with distilled water are shown in Fig. 6. These images indicate no major differences in the microstructure of the samples permeated in the two different directions relative to the sedimentary structure. Therefore, the block samples can be considered to be homogeneous media, except for cracking. A crack aligned parallel to the water flow is observable in each image: these cracks probably developed during the block sample coring.

4.3.3. X-ray CT observations of the core samples
The images of the core samples after permeation with distilled water are shown in Fig. 7. Crack development in the core samples might be due to the stress relief after core samples were drilled from the bentonite ore deposit and/or to the freezing and thawing process when core samples were transported. Anyway the cracks observed in the core samples might be developed before water permeation. The X-ray CT image of the core samples after permeation shows three different brightnesses in different areas. The cracks have the lowest brightness, bentonite has the highest brightness and there are areas with intermediate brightness (Fig. 7) that partly fill up the cracks and also have spread into the bentonite area after water permeation. This implies that the area with intermediate brightness may be a bentonite-water complex formed after the permeation with distilled water, as also found in the case of CBM. If the cracks were developed during sample preparation for X-ray CT observation after water permeation, they would probably be without ‘bentonite–water complexes’.

5. Discussion
As shown in Fig. 5 there are differences in the microstructures before and after permeation with distilled water. To better understand the differences between the two microstructures, a decomposition of the histograms was carried out. The histograms of the images can be decomposed into three populations having different CT numbers. The decomposition of the histogram was performed with the nonlinear least square method by using the central CT numbers of the three populations in the 2D images (Fig. 8). For the CBM before permeation, the CT numbers for the quartz, ‘bentonite–water complex’ and ‘vacant space’ were 73, −22 and −96, respectively; the CBM after permeation had CT numbers of 68, −23 and −88, respectively. These CT numbers are tentative values and should not be considered absolute. After the decomposition, the peak intensities of the three populations are shown in Table 3. Here the peak area intensity of vacant pore decreases, while that of bentonite–water complexes increases after permeation. These results show that the algorithms mentioned express differences in the microstructure of the CBMs before and after permeation.

<table>
<thead>
<tr>
<th></th>
<th>Before permeation</th>
<th>After permeation</th>
</tr>
</thead>
<tbody>
<tr>
<td>Vacant pores</td>
<td>130,801</td>
<td>89,069</td>
</tr>
<tr>
<td>Bentonite–water complexes</td>
<td>376,532</td>
<td>416,366</td>
</tr>
<tr>
<td>Quartz</td>
<td>314,799</td>
<td>316,923</td>
</tr>
</tbody>
</table>

Table 3
Peak area intensities of the decomposed CBMs.
It is noteworthy that the hydraulic conductivities of the core samples with cracks possibly developed during sample coring and/or sample transport are similar to those of backfill materials composed of compacted MX-80 bentonite as shown in Table 4. The hydraulic conductivities of the core samples of which the void ratio is 2.1, are of the same order as the hydraulic conductivity of a compacted MX-80 bentonite with void ratio of 2.0. The sealing ability of the core samples is not affected by the cracks filled with ‘bentonite–water complexes’. That is, the self-restoring ability has worked due to the smectite swelling within cracks of the bentonite ore during water permeation, similarly as in the case of the compacted bentonite.

The CT images of the core samples mentioned above are quite different from those of the compacted bentonite after saturation, which show a homogeneous area with a nearly constant CT number (Nishiyama et al., 2004). The remaining three different areas in the core samples are postulated to be due to the extremely-low water permeability of the ore bentonite which resided in diagenetic environments for a long geological time after the Cretaceous period. It is known that the cementation of smectite interlayers owing to iron-hydroxide formation may occur in iron-rich bentonite deposits and the cementation may progress by heating to 110 °C experimentally, and this cementation may disturb the swelling of smectite contained in bentonite (Williams et al., 1954). The smectite cementation of the core sample with a relatively high Fe₂O₃ content of ca. 8.0% might occur during diageneis of the bentonite.

To compare the microstructure of the core sample after water permeation with the CBM, the histogram of the X-ray CT image was decomposed with the method for CBM mentioned above. The averaged CT numbers of bentonite, ‘bentonite–water complexes’, and cracks observed in the image are 309, 45 and −131 (Fig. 9, left). The decomposition of the histogram shows that their three different areas are clearly distinguishable like in the decomposition diagram of CBM (Fig. 9). However the peak numbers are not comparable to those of the CBM where the X-ray CT observations used an aluminum board attached to the X-ray tube to control the scattering of X-rays.

On the basis of the scanning electron microscopic observations of saturated bentonites, micro-scale swelling has been shown to play an important role (Komine et al., 1991). In addition, Pusch (2001) reported that the clay matrix forming in saturated bentonites and having a variety of densities controls the bulk properties of bentonites, particularly the hydraulic and gas conductivities. Similarly, the bentonite–water complexes described in this study would be necessary to control the permeability of the saturated bentonites. Further detailed study of the properties and micro-scale behavior of bentonite–water complexes is an important subject to enable bentonite use as barrier and backfill materials under the variety of circumstances encountered in actual landfills.

6. Conclusions

The X-ray CT observations of CBMs showed that ‘vacant pores’ and ‘bentonite–water complexes’ of the CBMs before and after water permeation are distinguishable in X-ray CT images, and that the differences in the microstructure of the CBMs depend on the mixing conditions and sample preparation.

Permeability tests and X-ray CT observations of the bentonite ore samples showed that the permeability and the microstructure are independent of the sedimentary texture shown in the ore samples. Further, X-ray CT observations of saturated ore samples showed self-sealing of micro-cracks with ‘bentonite–water complexes’.

Acknowledgement

We thank Mr. T. Onishi and Mr. Y. Sato of JDC Corporation, and Mr. M. Shishime of Volclay Japan Corporation for offering the bentonite ore samples and their permeability data.

References


Pusch, R., 2001. The microstructure of MX-80 clay with respect to its bulk physical properties under different environmental conditions. Swedish Nuclear Fuel and Waste Management Co., SKB TR-01-08.


Table 4

<table>
<thead>
<tr>
<th>Void ratio</th>
<th>Hydraulic conductivity (m/s)</th>
</tr>
</thead>
<tbody>
<tr>
<td>MX-80 (Borgesson et al., 1995)</td>
<td></td>
</tr>
<tr>
<td>1.0</td>
<td>2.0 × 10⁻¹²</td>
</tr>
<tr>
<td>2.0</td>
<td>6.0 × 10⁻¹²</td>
</tr>
<tr>
<td>5.0</td>
<td>9.0 × 10⁻¹¹</td>
</tr>
<tr>
<td>Core samples</td>
<td>2.1</td>
</tr>
</tbody>
</table>

Fig. 9. CT number histogram transformed from 2D images of the core sample shown in Fig. 7A (left) and the peak decomposition of the CT number histogram (right). 1: cracks; 2: bentonite–water complexes; 3: bentonite; 4: convolution of 1–3.


