



# HOKKAIDO UNIVERSITY

Title	Analysis of microstructural images of dry and water-saturated compacted bentonite samples observed with X-ray micro CT
Author(s)	Tomioka, Satoshi; Kozaki, Tamotsu; Takamatsu, Hidenori et al.
Citation	Applied Clay Science, 47(1-2), 65-71 <a href="https://doi.org/10.1016/j.clay.2008.09.001">https://doi.org/10.1016/j.clay.2008.09.001</a>
Issue Date	2010-01
Doc URL	<a href="https://hdl.handle.net/2115/42844">https://hdl.handle.net/2115/42844</a>
Type	journal article
File Information	ACS47-1-2_65-71.pdf



# Analysis of microstructural images of dry and water-saturated compacted bentonite samples observed with X-ray micro CT

Satoshi Tomioka <sup>a,\*</sup>, Tamotsu Kozaki <sup>a</sup>, Hidenori Takamatsu <sup>a</sup>,  
Natsuko Noda <sup>a</sup>, Shusuke Nisiyama <sup>a</sup>, Naofumi Kozai <sup>b</sup>,  
Satoru Suzuki <sup>c,1</sup>, and Seichi Sato

<sup>a</sup>*Graduate School of Engineering, Hokkaido University, Kita 13 Nishi 8, Kita-ku,  
Sapporo, 060-8628 Japan*

<sup>b</sup>*Advanced Science Research Center, Japan Atomic Energy Agency (JAEA),  
Tokai, Ibaraki 319-1195, Japan*

<sup>c</sup>*Research Center for Deep Geological Environments, National Institute of  
Advanced Industrial Science and Technology, 1-1-1 Higashi, Tsukuba, Ibaraki,  
305-8567, Japan*

---

## Abstract

Compacted bentonite, of which the major clay mineral is montmorillonite, is a candidate buffer material for geological disposal of high-level radioactive waste. In this study, a microfocus X-ray computed tomography (micro-CT, X-ray microscope), which enables non-destructive, three-dimensional observation of the interior microstructure of a sample with high resolution (several microns), examined compacted montmorillonite samples under dry and water-saturated states. The images thus obtained were analyzed by a computer code developed for this study to obtain

the information on the size and shape of montmorillonite grains in the samples before and after the water saturation. From the results of the image analysis, it can be supposed that the outer montmorillonite sheets of grains swelled and formed a gel, whereas the inner montmorillonite sheets did not change significantly in the water-saturation process.

*Key words:* micro computed tomography, geological disposal, montmorillonite, compacted bentonite.

---

---

\* Corresponding author: S. Tomioka; Graduate School of Engineering, Hokkaido University, Kita 13 Nishi 8, Kita-ku, Sapporo, 060-8628 Japan; E-mail: tom@qe.eng.hokudai.ac.jp, Tel.: +81-11-706-6656, Fax: +81-11-706-7128.

*Email addresses:* tom@qe.eng.hokudai.ac.jp (Satoshi Tomioka),  
kozaki@eng.hokudai.ac.jp (Tamotsu Kozaki).

<sup>1</sup> Present address: Science and Technology Department, Nuclear Waste Management Organization of Japan (NUMO), Mita NN building, 1-23, Shiba 4-chome, Minato-ku, Tokyo, 108-0014, Japan

## 1. Introduction

2 Compacted bentonite (or a mixture of bentonite and sand) is a candidate  
3 buffer material for geological disposal of high-level radioactive waste in Japan  
4 (JNC, 2000). The buffer material plays various important roles in the disposal  
5 system, e.g., to inhibit ground water flow and also to retard the migration  
6 of radionuclides in the region between the waste forms and the surrounding  
7 host rock. Since the properties of the buffer material are closely related to the  
8 microstructure of the bentonite, the study of the microstructure is a key is-  
9 sue for the safety assessment of geological disposal. Therefore, observations of  
10 the microstructure of compacted bentonite have been extensively performed  
11 using transmission electron microscopes (TEMs) and/or scanning electron mi-  
12 croscopes (SEMs). Additionally, theoretical models for the microstructure of  
13 bentonite have been established (Pusch and Yong, 2006). However, further  
14 detailed knowledge of the microstructure of bentonite is still needed.

15 Three-dimensional imaging using X-ray computed tomography is a promising  
16 in-situ observation technique for geological materials (Carlson, 2006; Mooney,  
17 2002; Nakashima, 2003). The advantage of this technique is that the interior  
18 of samples can be non-destructively examined. In addition, 3D imaging with  
19 high spatial resolution (several microns) has been realized with a microfocus  
20 X-ray computed tomography (micro-CT, X-ray microscope), as well as an  
21 X-ray CT using synchrotron radiation. Although there are several reports  
22 available for the X-ray CT observation of geological materials (DeVore et al.,  
23 2006; Nakashima et al., 2004; Van Geet et al., 2001; Van Geet et al., 2005),  
24 few attempts using the X-ray micro CT have been made to observe bentonite  
25 particles before and after water saturation (Kozaki et al., 2001; Liu et al.,

1 2003).

2 In this study, a newly developed X-ray micro CT having a high spatial reso-  
3 lution (about  $0.8 \mu\text{m}$  under ideal conditions) was used to observe bentonite  
4 particles of compacted samples before and after water saturation. Computer  
5 code was newly developed for the analysis of micro-CT images to obtain in-  
6 formation detailing the morphologic changes of bentonite grains before and  
7 after water saturation.

## 8 **2. Experimental**

9 The microfocus X-ray computed tomography used in this study is a SkyScan-  
10 1172 (Skyscan, Belgium). This system is comprised of an X-ray source (100  
11 keV sealed microfocus tube with a spot size less than  $5 \mu\text{m}$ ), an air-cooled  
12 X-ray CCD camera having a resolution of  $4000 \times 2300$  pixels, and a precision  
13 object manipulator. The detectable spatial resolution of this system is about  
14  $0.8 \mu\text{m}$  under ideal conditions.

15 Bentonite used in this study is Na-type montmorillonite, Kunipia-F, which  
16 is commercially available from Kunimine Industries, Japan. This sample con-  
17 tains montmorillonite ( $> 95\text{wt}\%$ ) and minor components (Sato et al., 1992).  
18 The montmorillonite was purified into homoionic  $\text{Na}^+$ -type montmorillonite,  
19 ground by mortar and pestle, and then sieved to obtain a grain size of  $75\text{-}150$   
20  $\mu\text{m}$ , as described elsewhere (Kozaki et al., 1999). The purified montmorillonite  
21 samples were then compacted to a dry density of  $1.0 \text{ Mg m}^{-3}$  in glassy carbon  
22 tube (Tokai Carbon Co., LTD). Each tube had a 5-mm internal diameter and  
23 a 10-mm height. A negligibly small amount (*ca.*  $1.3 \text{ vol.}\%$ ) of glass spheres

1 (100-200  $\mu\text{m}$  in diameter) was included in some montmorillonite samples as a  
2 reference. The montmorillonite samples which were fully saturated with wa-  
3 ter were prepared by contacting the compacted samples with distilled water  
4 through sintered stainless steel filters as illustrated in Fig. 1. After the water  
5 saturation period of more than two weeks, the cells were dismantled to expose  
6 the glassy carbon tubes. Both ends of each glassy carbon tube were sealed with  
7 a plastic plate held in place by adhesive to contain the water-saturated sample  
8 within the tube. The full water-saturations of the samples were confirmed in  
9 another experiment by weighing the samples before and after an oven-dry at  
10 378 K.

11 The microstructures of the samples in the glassy carbon tubes were then ob-  
12 served with the micro-CT technique. The voltage of 50 kV was applied to the  
13 X-ray tube, and an aluminum filter (0.5 mm in thickness) was used in the  
14 observation. The samples were examined while being rotated 360 degrees in  
15 0.1-degree steps. Data acquisition time was approximately 16 hours for each  
16 sample. The images obtained by 3D-reconstruction processes consist of 4000  
17 x 4000 pixels with each pixel having an 8-bit attenuation value (i.e., signal in-  
18 tensity). The image spatial resolution was about 2  $\mu\text{m}$  in this observation. The  
19 attenuation value of the X-ray corresponds to the image brightness at each  
20 pixel. The measurements and the 3D-reconstruction processes for all samples  
21 were carried out under identical conditions.

### 22 **3. Analytical method using computer code**

23 The two-dimensional micro-CT images were analyzed using a computer code  
24 developed for this study. The code employed three analytical steps; noise re-

1 duction, grain boundary analysis, and grain shape analysis. In the first step, a  
2 ring artifact reduction and image noise reduction were applied to the micro-CT  
3 images. In the second step, montmorillonite grains in the image were identi-  
4 fied using a boundary determination algorithm. In the last step, the results  
5 were examined to determine several quantities associated with the montmoril-  
6 lonite particle grain shapes: the cross-sectional area, the length of the contour  
7 (perimeter), the length of the major, minor and mean axes, and the aspect  
8 ratio of each grain. Detailed procedures for each step are described below, to-  
9 gether with the boundary determination algorithm and the parameters used  
10 for the calculations.

### 11 *3.1. Noise reduction*

12 The two-dimensional CT images contained 4000 x 4000 pixels, and each pixel  
13 was comprised of an 8-bit signal. In this study, a set of 256 pieces of two-  
14 dimensional CT images (horizontal plane of 400 x 400 pixels) was precon-  
15 ditioned to remove ring artifacts and random noise, and analyzed to obtain  
16 the grain shape information of the montmorillonite sample. Neither the glass  
17 beads nor any part of the glassy carbon sample holder was involved in the  
18 rectangular parallelepiped region of 400 x 400 x 256 pixels (*ca.* 800 x 800 x  
19 500  $\mu\text{m}$ ).

20 The ring artifacts in the micro-CT images are observed as bright (or dark)  
21 concentric circles, of which the center of each is the same as the rotational  
22 center of the sample. Then, these artifacts were removed by a normalization  
23 in which the averaged intensities of pixels along every circle are assumed to  
24 be same. On the other hand, the random noise reduction procedure involved

1 two steps; spectrum analysis and filtering. The image spectrum contains the  
2 signals of grains in the lower spatial frequency region and widely distributed  
3 noise (Press et al., 1992). Assuming a model that contained the spectrum of  
4 superimposed signals with some Gaussian functions, white noise, and Gaussian  
5 shaped noise having wide spectral width; we determined the nature of each  
6 component of the spectrum using a nonlinear least-squares fit (Tomioka et al.,  
7 2007). The two types of noise (i.e., the white noise and the Gaussian shaped  
8 noise) were removed by a Gaussian spatial filter that had filtering parameters  
9 determined from the result of the nonlinear least-squares fit.

### 10 3.2. Grain boundary analysis

11 Grain boundaries of montmorillonite particles were discriminated by using a  
12 boundary determination algorithm included in the code. The scheme of the  
13 algorithm is shown in Fig. 2. In the algorithm, nodes having specific signal  
14 intensities were introduced in the image. The node was defined as the point  
15 at the center of each pixel, and the signal intensity was the same as that of  
16 the pixel. After a proper discrimination level of signal intensity for the grain  
17 boundary was determined, a point having the same value of signal intensity as  
18 the discrimination level was selected between two adjacent nodes. The position  
19 of this point was determined using linear interpolation. Segments of the grain  
20 boundary can be drawn with lines connecting adjacent points. The connecting  
21 lines were sequentially established in the direction of  $\nabla f \times \mathbf{e}_z$ , where  $f$  is signal  
22 intensity and  $\mathbf{e}_z$  is the unit vector normal to the image plane ( $x$ - $y$  plane). Since  
23 the direction  $\nabla f$  is inward on the contour around the montmorillonite grains,  
24 the contour runs counter-clockwise. Using these steps, both closed and open

1 grain boundaries were obtained. However, only closed boundaries were utilized  
 2 to define grains in this analysis; since an open grain boundary, which signifies  
 3 that some portion of the grain was not in the image range, could lead to  
 4 underestimated sizes and cross-sectional areas of grains.

### 5 3.3. Grain size analysis and shape analysis

6 By applying the above boundary determination algorithm to image data, the  
 7 contours of all montmorillonite grains could be obtained except for grains with  
 8 a portion of the grain not in the image. The circumference of a particular closed  
 9 grain boundary,  $l$ , can be expressed by the following equation;

$$l = \sum_{j=0}^{N-1} |\mathbf{r}_j - \mathbf{r}_{j+1}|, \quad (1)$$

10 where  $N$  is the total number of points belonging to the contour,  $\mathbf{r}_j$  is the  
 11 location of the  $j$ -th point of a boundary line, and  $\mathbf{r}_N$  is identical to  $\mathbf{r}_0$ .

12 On the other hand, the cross-sectional area of a grain can be calculated by  
 13 the contour integral. According to Stokes' integral theorem, one relationship  
 14 between a domain integral and a closed boundary integral can be given as  
 15 follows;

$$\int \nabla \times \mathbf{A} \cdot \mathbf{n} dS = \oint \mathbf{A} \cdot \mathbf{t} dl, \quad (2)$$

16 where  $\mathbf{A}$  is an arbitrary vector,  $\mathbf{n}$  and  $\mathbf{t}$  respectively denote a unit vector  
 17 normal to a cross-sectional area  $dS$  and a unit vector tangential to a boundary  
 18  $dl$ .

19 When the closed boundary is on  $x$ - $y$  plane, taking  $\mathbf{n}$  to be  $\mathbf{e}_z$ , a vector  $x\mathbf{e}_y$  can

1 be chosen as the arbitrary vector  $\mathbf{A}$ , which can satisfy the relation  $\nabla \times \mathbf{A} = \mathbf{e}_z$ ,  
 2 the area of closed grain boundary  $S$  can be given as follows;

$$S = \oint x \mathbf{e}_y \cdot \mathbf{t} dl = \oint x dy = \sum_{j=0}^{N-1} \frac{x_j + x_{j+1}}{2} (y_{j+1} - y_j). \quad (3)$$

3 In this equation,  $S$  of a montmorillonite grain is always positive because the  
 4 signal intensities at any positions in the boundary surrounding the grain are  
 5 larger than the discrimination level, and the contour integral path runs in  
 6 the counter-clockwise direction. Conversely, the contours with a negative  $S$   
 7 value are considered to be internal holes in montmorillonite grains or vacancies  
 8 surrounded by montmorillonite particles. In this study, therefore, the grain  
 9 boundaries with a negative  $S$  value were omitted from the analysis.

10 The major axis of a montmorillonite grain,  $d_L$ , is defined as the longest distance  
 11 between two points among all possible pairs of points on a grain boundary as  
 12 follows;

$$d_L = \max_{i,j} |\mathbf{r}_i - \mathbf{r}_j|, \quad (i, j \in 0, \dots, N-1). \quad (4)$$

13 By assuming the montmorillonite grain to be elliptical, the area,  $S$ , can be  
 14 expressed as follows;

$$S = \pi \frac{d_L}{2} \frac{d_S}{2}, \quad (5)$$

15 where  $d_S$  is the minor axis of the montmorillonite grain. Consequently, the  
 16 minor axis,  $d_S$ , can be given by the following equation;

$$d_S = \frac{4S}{\pi d_L}. \quad (6)$$

1 In addition, the mean diameter of grains,  $d_m$ , can be given as the geometrical  
2 average by the following equation;

$$d_m = \sqrt{d_L d_S} = 2\sqrt{\frac{S}{\pi}}. \quad (7)$$

3 The aspect ratio of grains,  $\alpha$ , can be defined by the following equation;

$$\alpha = \frac{d_S}{d_L} = \frac{4S}{\pi d_L^2}. \quad (8)$$

## 4 4. Results and discussion

### 5 4.1. Microstructure of dry-state compacted montmorillonite

6 The brightness of a CT-image corresponds to the attenuation of the X-rays,  
7 which depends on the atomic numbers of the elements and their densities in a  
8 sample. A typical horizontal-plane, two-dimensional, micro-CT image of a dry  
9 montmorillonite sample at a dry density of  $1.0 \text{ Mg m}^{-3}$  is shown in Fig. 3. In  
10 this figure, montmorillonite particles can be clearly identified as ellipse-shaped  
11 dapples (gray spots) inside the big dark gray ring, which is the glassy carbon  
12 sample holder. On the other hand, the glass spheres introduced as a reference  
13 can be identified as reasonably large, definitely circular dapples (bright spots)  
14 anywhere in the image. This suggests that the CT apparatus used in this study  
15 can precisely reproduce distinct images of masses in a sample with at least a  
16 resolution of several microns.

17 Figure 4 shows the frequency distribution of pixels as a function of signal in-  
18 tensity that was obtained for a dry montmorillonite sample. The frequency

1 varied continuously but has a high peak at a signal intensity of 70 and a lower  
 2 peak at a signal intensity of 140. It seems that pixels at the signal intensity  
 3 of 70 and 140 correspond to air-filled void spaces and montmorillonite parti-  
 4 cles, respectively. In addition, the continuous distribution suggests that there  
 5 are a certain number of pixels in which air and montmorillonite coexist at  
 6 various ratios. These result in pixels with grayish color in the Fig. 3 image.  
 7 The montmorillonite grain boundaries, therefore, should be evaluated with an  
 8 appropriate threshold value (discrimination level). In this study, the discrim-  
 9 ination level was determined by assuming that the numerical ratio of bright  
 10 pixels to the whole pixels was identical to the theoretical volumetric fraction of  
 11 the solid (montmorillonite),  $\eta$ , which can be given by the following equation;

$$\eta = \frac{V^c}{V^{all}} = \frac{\rho - \rho_a}{\rho_c - \rho_a}, \quad (9)$$

12 where  $V^c$  is the volume of montmorillonite,  $V^{all}$  is the total volume of the  
 13 sample,  $\rho_c$  is the partial density of montmorillonite ( $2.88 \text{ Mg m}^{-3}$ ),  $\rho_a$  is the  
 14 absolute density of air ( $\sim 0 \text{ Mg m}^{-3}$ ), and  $\rho$  is the dry density of montmoril-  
 15 lonite (i.e., the packing density in dry condition;  $1.0 \text{ Mg m}^{-3}$ ). In this case, the  
 16 value of  $\eta$  becomes 0.347. For the dry montmorillonite sample, the threshold  
 17 value to discriminate grain boundaries can be determined to be 103.2, which is  
 18 in accordance with the signal intensity at the accumulated frequency of 0.653  
 19 ( $= 1 - 0.347$ ) in Fig. 4.

20 Figure 5 shows grain boundaries determined by the computer code with a  
 21 discrimination level of 103.2 for the image of the dry montmorillonite sample  
 22 at the dry density of  $1.0 \text{ Mg m}^{-3}$ , together with images before and after the  
 23 noise reduction. The dapples drawn by solid lines are montmorillonite particles  
 24 recognized by the code, whereas dapples drawn by dotted lines are closed void

1 spaces surrounded with montmorillonite or portions of montmorillonite parti-  
2 cles that were not in the image range. The total number of montmorillonite  
3 grains identified with the computer code was approximately 24,000.

4 The distribution of the mean diameter of the boundaries of grains analyzed  
5 with the algorithm discussed in the section 3.3 is shown in Fig. 6. The grains,  
6 which had been sieved to 75-150  $\mu\text{m}$  in the sample preparation process, have  
7 wide peaks in the mean diameters around 100  $\mu\text{m}$ . Consequently, it follows  
8 that the images obtained by micro-CT examinations represent the interior mi-  
9 crostructures of the dry montmorillonite. Further, the combination of micro-  
10 CT and the code enables us to evaluate the microstructures of the dry com-  
11 pacted montmorillonite samples with high resolution.

#### 12 4.2. *Microstructure of water-saturated compacted montmorillonite*

13 In the case of water-saturated montmorillonite samples, water in the sample  
14 may attenuate both the intensity and the energy of the X-rays, resulting in  
15 unclear images with artifacts. In addition, the homogenization of the mont-  
16 morillonite caused by the water saturation may make it difficult to determine  
17 a proper discrimination level for grain boundaries during the image analysis.

18 A typical horizontal plane, two-dimensional, micro-CT image of a water-  
19 saturated montmorillonite sample at a dry density of  $1.0 \text{ Mg m}^{-3}$  is shown  
20 in Fig. 7. Montmorillonite particles are unable to be clearly recognized in the  
21 image, although glass spheres still can be identified as circular dapples. There-  
22 fore, water saturation can homogenize the montmorillonite sample, at least in  
23 the scale of this spatial resolution (about 2  $\mu\text{m}$ ). Figure 8 is a plot of the fre-

1 quency distribution of pixels in the image of water-saturated montmorillonite  
2 as a function of signal intensity. This also suggests that the montmorillonite  
3 sample becomes almost homogeneous following water saturation, since there is  
4 only one sharp peak in the figure. However, small patches with slightly higher  
5 brightness seem to be present in the image of the water-saturated montmoril-  
6 lonite shown in Fig. 7. Therefore, the same analysis as for the dry sample was  
7 applied to the water-saturated sample in this study.

8 The spectral analysis detected significant signals in low spatial frequency range  
9 which was not noise. This suggests the presence of objects in the image. The  
10 discrimination level of 141.9 was used for the image of the water-saturated  
11 montmorillonite sample, as shown in Fig. 8. This discrimination level could  
12 produce overestimated values of grain sizes, since the level was determined by  
13 assuming the numerical ratio of bright pixels to the whole pixels was identi-  
14 cal to the theoretical volumetric fraction of the solid, i.e.  $\eta$ ; a fraction of gel  
15 phase was needed to be recognized as solid phase in order to attain the same  
16 numerical ratio as the theoretical volumetric fraction. Figure 9 shows an ex-  
17 ample of grain boundaries of the water-saturated montmorillonite determined  
18 with the computer code using the discrimination level, with the images before  
19 and after the noise reduction. The total number of grains identified with the  
20 computer code was about 12,000, which is almost half of that in the dry con-  
21 dition. This decrease in the total number of grains can be attributed to the  
22 increase in the open grain boundary, which signifies that some portion of the  
23 grain was not in the image, as mentioned above. As compared with Fig. 5(c),  
24 the grains in the water-saturated sample (Fig. 9(c)) seem to be smaller than  
25 those in the dry sample (Fig. 5(c)). To compare them quantitatively, the mean  
26 diameter distribution of the water-saturated montmorillonite grains analyzed

1 with the computer code together with that of the dry sample is shown in Fig.  
2 6. Realizing that the grain sizes could be overestimated in the image of the  
3 water-saturated sample, it is obvious that the grain sizes decreased with the  
4 water saturation.

5 On the other hand, in the X-ray diffraction study of water-saturated, com-  
6 pacted montmorillonite, the diffraction peak of 1.88 nm, which corresponds  
7 to the three-water-layer hydrate state of the montmorillonite interlayer, was  
8 reported to emerge at the dry density of  $1.0 \text{ Mg m}^{-3}$ . However, there was no  
9 peak below this dry density (Kozaki et al., 1998). This fact suggests that the  
10 montmorillonite at this dry density still has dense fragments (indicated by  
11 the diffraction peak) even after the water saturation. It is very likely that the  
12 dense fragments correspond to the grains that were identified in Fig. 9 with  
13 the computer code for the water-saturated montmorillonite.

14 Figure 10 indicates the distributions of the grain aspect ratio (i.e., the ratio  
15 of the minor axis to the major axis) before and after water saturation. No  
16 significant change in grain aspect ratio was found in the images before and  
17 after water saturation. Then, except the region of the open grain boundaries,  
18 it can be supposed that the outer montmorillonite sheets of grains swelled  
19 and formed a gel, whereas the inner sheets did not change significantly in the  
20 water-saturation process, as illustrated in Fig. 11. The gel that formed could  
21 occupy the vacancies between grains. This kind of model for water-saturation  
22 of montmorillonite had been proposed elsewhere (Pusch et al., 2006). However,  
23 in the analysis of the micro-CT image with the computer code in this study,  
24 some portions of gel with slightly higher density could be recognized as a part  
25 of or the whole of montmorillonite particles as illustrated with dark gray color  
26 in Fig. 11, due to the overestimation for the solid phase in the grain boundary

1 determination algorithm. This is a possible reason for the small changes in the  
2 grain aspect ratios before and after the water-saturation.

### 3 **5. Conclusion**

4 The nondestructive, three-dimensional images of the internal microstructures  
5 of compacted montmorillonite samples in dry and water-saturated states were  
6 obtained with the micro-CT, and analyzed with the computer code developed  
7 in this study. The code could determine grain boundaries of montmorillonite in  
8 the images by using appropriate discrimination levels, and provide information  
9 on size and shape of montmorillonite grains, such as the mean diameter and  
10 the aspect ratio of montmorillonite grains. From the results of this analysis,  
11 it is supposed that the outer montmorillonite sheets of grains swelled and  
12 formed a gel, whereas the inner sheets did not change significantly in the  
13 water-saturation process.

### 14 **Acknowledgements**

15 The authors express their appreciation to Mr. T. Ohyagi and Ms. Y. Mizuguchi,  
16 TOYO Corporation, for providing us with the opportunity to use the micro-  
17 CT apparatus and for their technical suggestions regarding these experiments.  
18 The authors wish to thank Professors M. Nakano and R. Pusch for their valu-  
19 able suggestions. Financial support was provided by the Ministry of Education,  
20 Culture, Sports, Science and Technology (MEXT), Japan (a Grant-in-Aid for  
21 Scientific Research (C), No. 17560049), the Japan Atomic Energy Agency  
22 (JAEA), and the Radioactive Waste Management Funding and Research Cen-

1 ter (RWMC).

## 2 **References**

3 Carlson, W.D., 2006. Three-dimensional imaging of earth and planetary ma-  
4 terials. *Earth and Planetary Science Letters* 249, 133-147.

5 DeVore, M.L., Kenrick, P., Pigg, K.B., Ketcham, R.A., 2006. Utility of high  
6 resolution x-ray computed tomography (HRXCT) for paleobotanical stud-  
7 ies: An example using London Clay fruits and seeds. *American Journal of*  
8 *Botany* 93, 1848-1851.

9 Japan Nuclear Cycle Development Institute (JNC), 2000. H12: project to es-  
10 tablish the scientific and technical basis for HLW disposal in Japan, sup-  
11 porting report 3 safety assessment of the geological disposal system. JNC  
12 Technical Report, JNC TN 1400 2000-04.

13 Kozaki, T., Fujishima, A., Sato, S., Ohashi, H., 1998. Self-diffusion of sodium  
14 ions in compacted sodium montmorillonite. *Nuclear Technology* 121, 63-69.

15 Kozaki, T., Sato, Y., Nakajima, M., Kato, H., Sato, S., Ohashi, H., 1999. Effect  
16 of particle size on the diffusion behavior of some radionuclides in compacted  
17 bentonite. *Journal of Nuclear Materials* 270, 265-272.

18 Kozaki, T., Suzuki, S., Kozai, N., Sato, S., Ohashi, H., 2001. Observation of  
19 microstructures of compacted bentonite by microfocus X-ray computerized  
20 tomography (micro-CT). *Journal of Nuclear Science and Technology* 38,  
21 697-699.

22 Liu, J., Kozaki, T., Horiuchi, Y., Sato, S., 2003. Microstructure of montmoril-  
23 lonite/silica sand mixture and its effects on the diffusion of strontium ions.  
24 *Applied Clay Science* 23, 89-95.

25 Mooney, S.J., 2002. Three-dimensional visualization and quantification of soil

1 macroporosity and water flow patterns using computed tomography. *Soil*  
2 *Use and Management* 18, 142-151.

3 Nakashima, Y., 2003. Diffusivity measurement of heavy ions in Wyoming  
4 montmorillonite gels by X-ray computed tomography. *Journal of Contami-*  
5 *nant Hydrology* 61, 147-156.

6 Nakashima, Y., Nakano, T., Nakamura, K., Uesugi, K., Tsuchiyama, A., Ikeda,  
7 S., 2004. Three-dimensional diffusion of non-sorbing species in porous sand-  
8 stone: computer simulation based on X-ray microtomography using syn-  
9 chrotron radiation. *Journal of Contaminant Hydrology* 74, 253-264.

10 Press, W.H., Teukolsky, S.A., Vetterling, W.T., Flannery, B.P., 1992. *Fourier*  
11 *and Spectral Applications. in: Numerical Recipes in Fortran 77: the art*  
12 *of scientific computing, Second ed. Press Syndicate of the University of*  
13 *Cambridge, New York & Melbourne.*

14 Pusch, R., Yong, R.N., 2006. *Microstructure of smectite clays and engineering*  
15 *performance. Taylor & Francis, London & New York.*

16 Sato, H., Ashida, T., Kohara, Y., Yui, M., Sasaki, N., 1992. Effect of dry  
17 density on diffusion of some radionuclides in compacted sodium bentonite.  
18 *Journal of Nuclear Science and Technology* 29, 873-882.

19 Tomioka, S., Nisiyama, S., Enoto, T., 2007. Nonlinear least square regression  
20 by adaptive domain method with multiple genetic algorithms. *IEEE trans-*  
21 *actions on Evolutionary Computation* 11, 1-16.

22 Van Geet, M., Swennen, R., David, P., 2001. Quantitative coal characterisation  
23 by means of microfocuss X-ray computer tomography, colour image analysis  
24 and back-scattered scanning electron microscopy. *International Journal of*  
25 *Coal Geology* 46, 11-25.

26 Van Geet, M., Volckaert, G., Roels, S., 2005. The use of microfocuss X-ray com-  
27 puted tomography in characterising the hydration of a clay pellet/powder

1 mixture. *Applied Clay Science* 29, 73-87.

## List of Figures

Fig. 1 Preparation of the water-saturated sample.

Fig. 2 Schematic of boundary determination algorithm.

Fig. 3 A typical horizontal plane, two-dimensional, micro-CT image of a dry montmorillonite sample (grain size: 75-100  $\mu\text{m}$ ; dry density: 1.0  $\text{Mg m}^{-3}$ ). This sample contained glass spheres having diameters of 100-200  $\mu\text{m}$ .

Fig. 4 Signal intensity distribution of pixels in all images included in a set of CT data of a dry montmorillonite sample (grain size: 75-100  $\mu\text{m}$ ; dry density: 1.0  $\text{Mg m}^{-3}$ ); thin and thick lines represent frequency and accumulated frequency, respectively.

Fig. 5 Image processing of a CT image of a dry montmorillonite sample: (a) original image, (b) preconditioned image by using the ring artifact reduction technique and Gaussian filtering, (c) extracted boundaries from the preconditioned image by using the grain boundary determination algorithm.

Fig. 6 Mean diameter distribution of montmorillonite grains before and after water saturation. Lines with and without square symbols represent accumulated area fraction and area fraction, respectively.

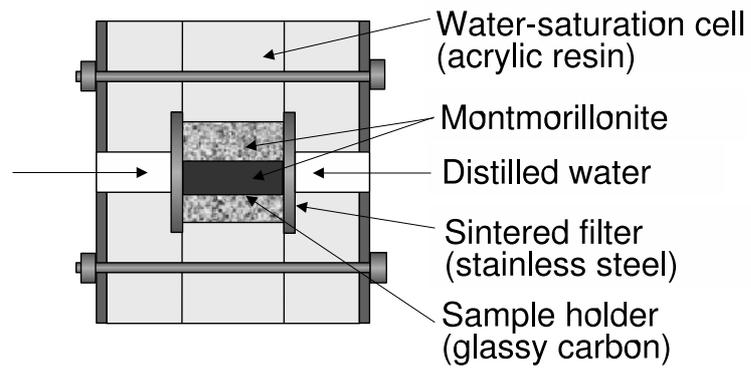
Fig. 7 A typical horizontal plane, two-dimensional, micro-CT image of a water-saturated montmorillonite sample (grain size: 75-100  $\mu\text{m}$ ; dry density: 1.0  $\text{Mg m}^{-3}$ ). This sample contained glass spheres having diameter of 100-200  $\mu\text{m}$ .

Fig. 8 Signal intensity distribution of pixels in all images included in a set of CT data of a water-saturated montmorillonite sample (grain size: 75-100  $\mu\text{m}$ ; dry density: 1.0  $\text{Mg m}^{-3}$ ); thin and thick lines represent frequency and accumulated frequency, respectively.

Fig. 9 Image processing of a CT image of a water-saturated montmorillonite sample: (a) original image, (b) preconditioned image by using the ring artifact reduction technique and Gaussian filtering in which image contrast is enhanced to four times that of the original image, (c) extracted boundaries from the preconditioned image by grain boundary determination algorithm.

Fig. 10 Aspect ratio distribution montmorillonite grains before and after water-saturation. Lines with and without square symbols represent accumulated area fraction and area fraction, respectively.

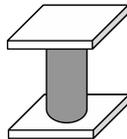
Fig. 11 Schematic of the structures of montmorillonite : (a) in dry state, (b) in water-saturated state.



(a) Water-saturation of montmorillonite



(b) Water-saturated montmorillonite inside of the sample holder



(c) Water-saturated montmorillonite in the sample holder capped with plastic plates for X-ray micro CT observation

Fig. 1. Preparation of the water-saturated sample.

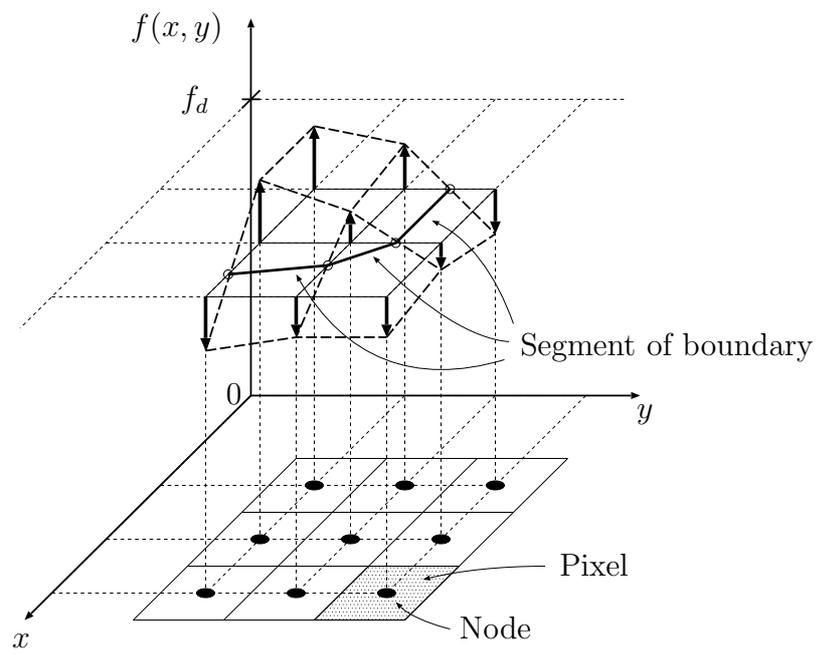


Fig. 2. Schematic of boundary determination algorithm.

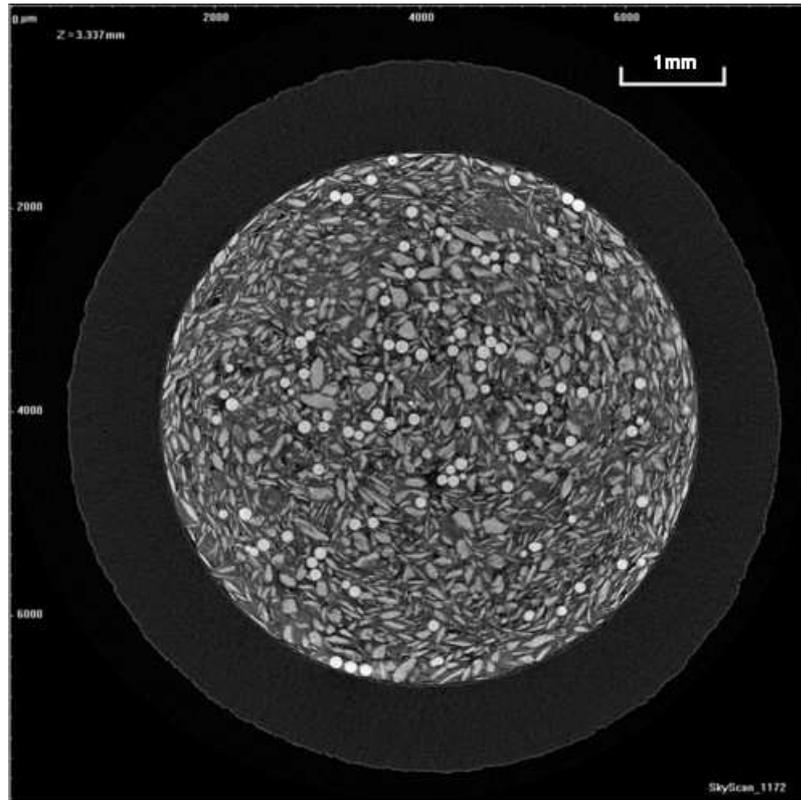


Fig. 3. A typical horizontal plane, two-dimensional, micro-CT image of a dry montmorillonite sample (grain size: 75-100  $\mu\text{m}$ ; dry density: 1.0  $\text{Mg m}^{-3}$ ). This sample contained glass spheres having diameters of 100-200  $\mu\text{m}$ .

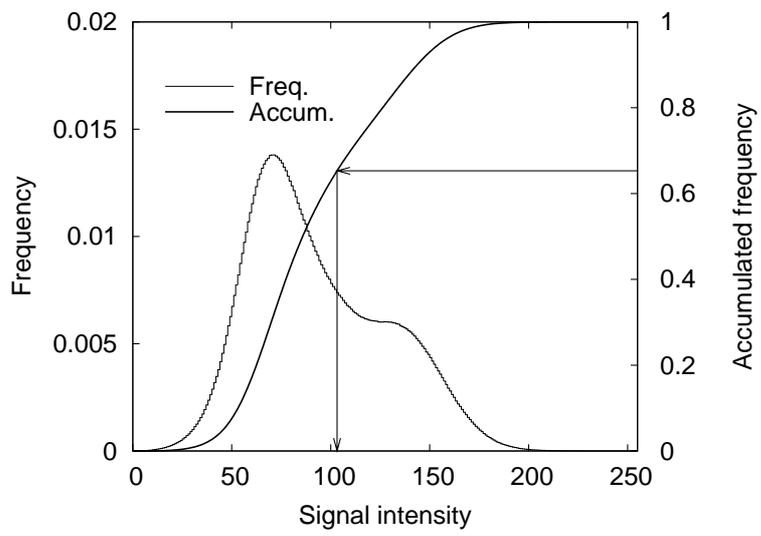


Fig. 4. Signal intensity distribution of pixels in all images included in a set of CT data of a dry montmorillonite sample (grain size: 75-100  $\mu\text{m}$ ; dry density: 1.0  $\text{Mg m}^{-3}$ ); thin and thick lines represent frequency and accumulated frequency, respectively.

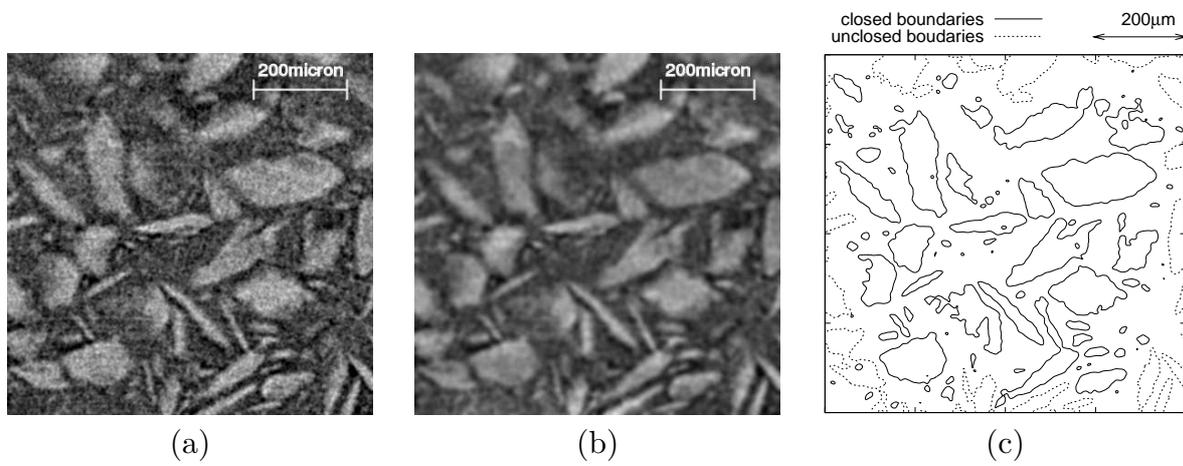


Fig. 5. Image processing of a CT image of a dry montmorillonite sample: (a) original image, (b) preconditioned image by using the ring artifact reduction technique and Gaussian filtering, (c) extracted boundaries from the preconditioned image by using the grain boundary determination algorithm.

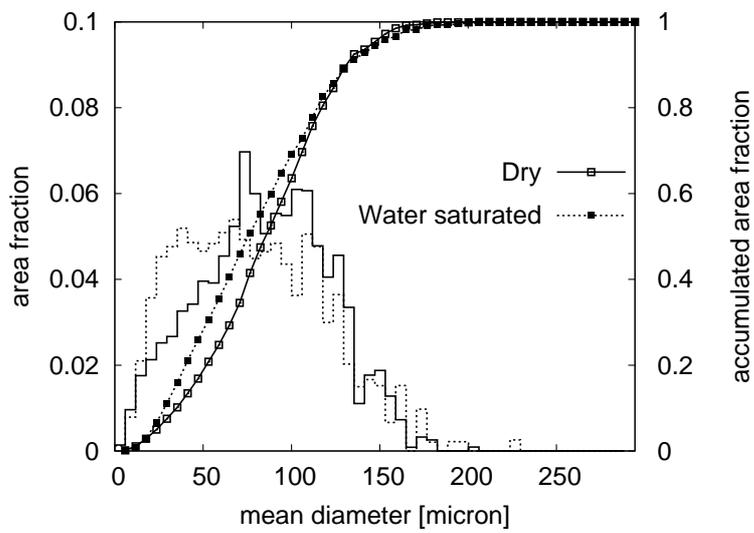


Fig. 6. Mean diameter distribution of montmorillonite grains before and after water saturation. Lines with and without square symbols represent accumulated area fraction and area fraction, respectively.

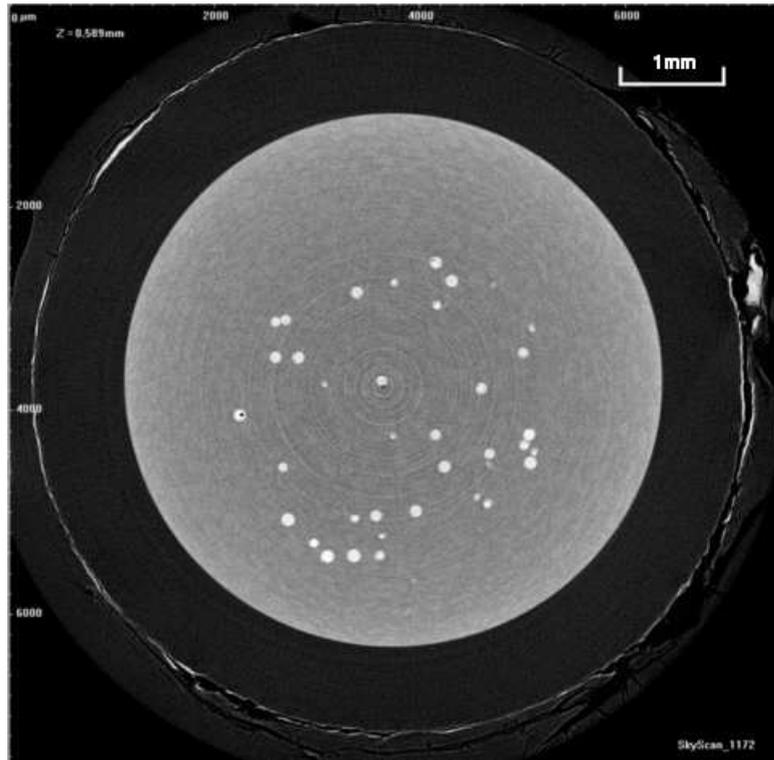


Fig. 7. A typical horizontal plane, two-dimensional, micro-CT image of a water-saturated montmorillonite sample (grain size: 75-100  $\mu\text{m}$ ; dry density: 1.0  $\text{Mg m}^{-3}$ ). This sample contained glass spheres having diameter of 100-200  $\mu\text{m}$ .

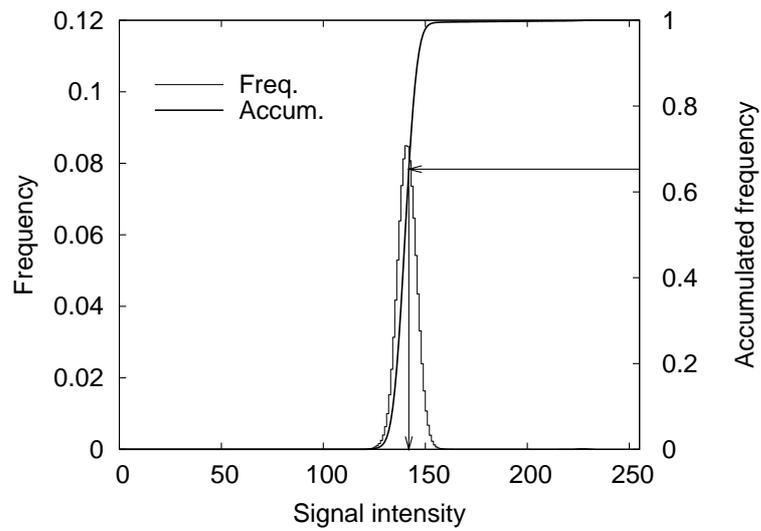


Fig. 8. Signal intensity distribution of pixels in all images included in a set of CT data of a water-saturated montmorillonite sample (grain size: 75-100  $\mu\text{m}$ ; dry density: 1.0  $\text{Mg m}^{-3}$ ); thin and thick lines represent frequency and accumulated frequency, respectively.

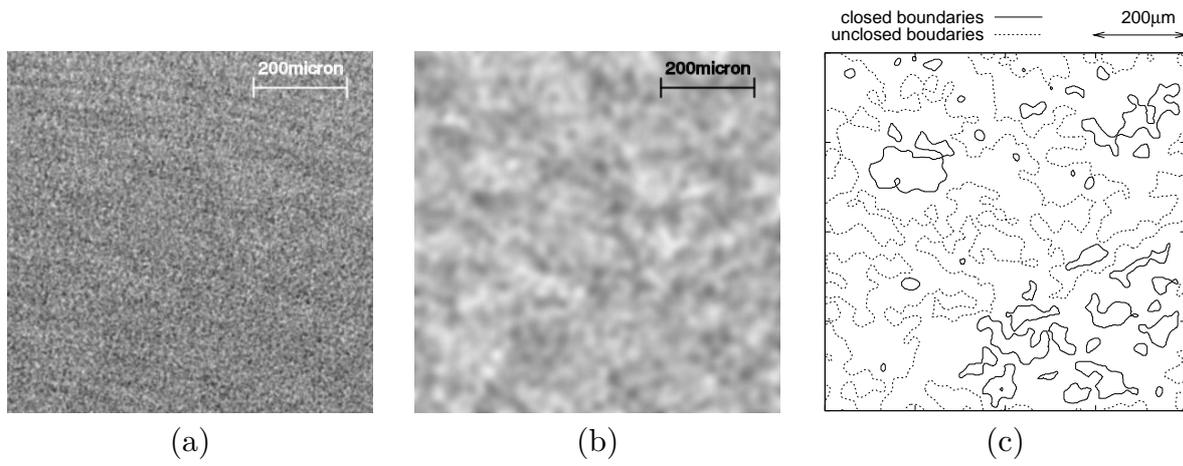


Fig. 9. Image processing of a CT image of a water-saturated montmorillonite sample: (a) original image, (b) preconditioned image by using the ring artifact reduction technique and Gaussian filtering in which image contrast is enhanced to four times that of the original image, (c) extracted boundaries from the preconditioned image by grain boundary determination algorithm.

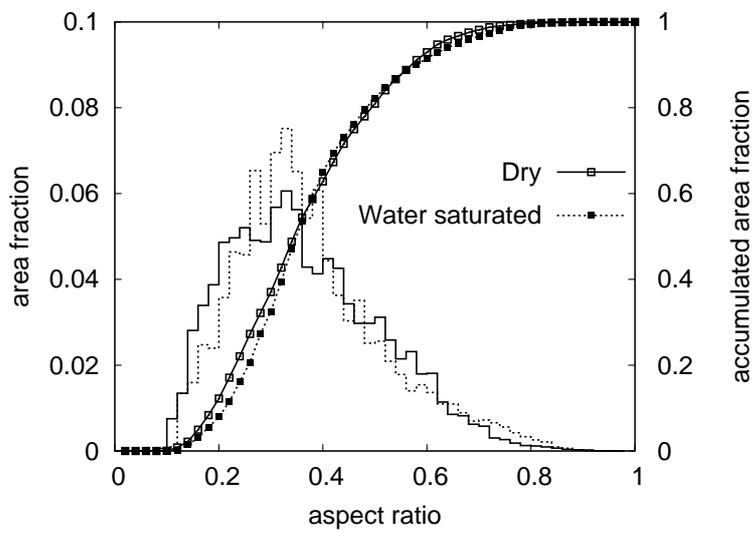
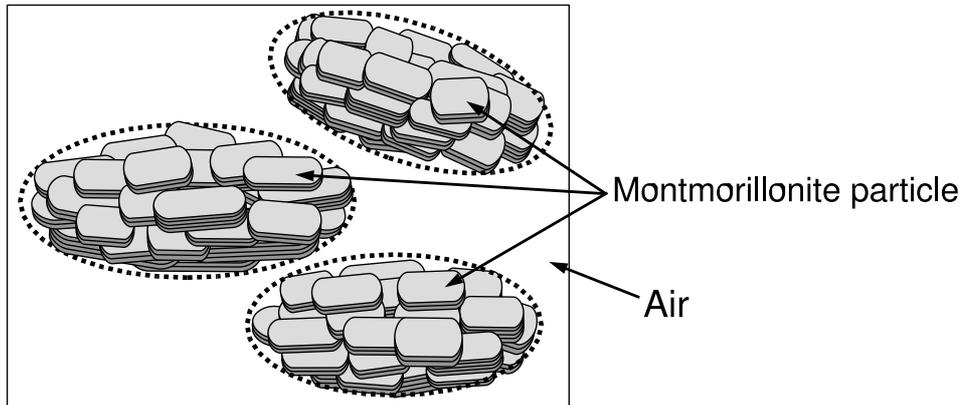
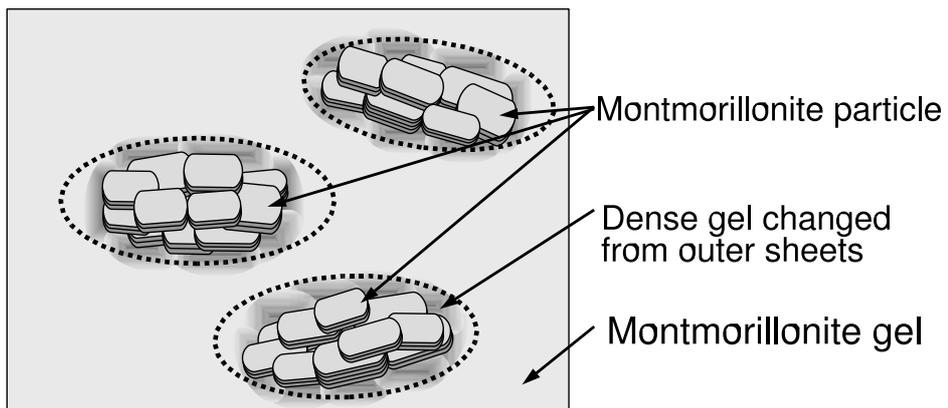


Fig. 10. Aspect ratio distribution montmorillonite grains before and after water-saturation. Lines with and without square symbols represent accumulated area fraction and area fraction, respectively.



(a) Montmorillonite in dry condition



(b) Water-saturated montmorillonite

Fig. 11. Schematic of the structures of montmorillonite : (a) in dry state, (b) in water-saturated state.