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Visualization of microstructure and measurement of mass transport parameters for granulated bentonite mixtures

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ABSTRACT

Bentonite is often considered as one of the key components of the Engineered Barrier System (EBS) for the radioactive waste disposal; mechanical, thermal, and hydraulic behaviors of bentonite should be thoroughly studied to ensure the emplaced quality and to control the performance of EBS. This study examined the effects of dry density and relative humidity (RH) on microstructure and mass transport characteristics of Granulated Bentonite Mixtures (GBM). The tested GBM samples were prepared by sieving and grading Japanese bentonite (trade name: OK Bentonite, Kunimine Industries, Japan). For dry density-controlled conditions, air-dried GBM samples (RH 60%, water content of 6.38% by weight) were packed at six different dry densities ranging from 1.25 to 1.75 g/cm³. The samples for humidity-controlled conditions were prepared at the dry density of 1.45 g/cm³ and stored at RH 90% (0 to 12 weeks). A microfocus X-ray computerized tomography (MFXCT) scanning apparatus was used to visualize the microstructures of packed tested samples. For each tested sample, the mass transport parameters of gas diffusivity ($D_p/D_o$), air permeability ($k_a$), thermal conductivity ($\lambda$), and volumetric heat capacity ($C$) were measured. The results indicated that dry density greatly affected the CT brightness values of GBM; the histogram of CT brightness well captured the internal distribution of density and the CT brightness homogenized more with increasing in dry density. The measured $D_p/D_o$ and $k_a$ were mainly controlled by the dry density. On the other hand, the thermal properties such as $\lambda$ and $C$ were governed both dry density and RH.

Keywords: Engineered Barrier System (EBS), granulated bentonite mixtures (GBM), microstructure, mass transport parameters, dry density, relative humidity

1 INTRODUCTION

The design concept for the disposal of high-level radioactive waste in deep geological environment includes the construction of an engineered barrier system (EBS) around the waste canisters composed of a backfilling/buffer material (Villator and Lloret, 2008). Bentonite has been selected as a backfilling/buffer material in EBS of high-level waste repositories due to its favorable physical and chemical properties (Arthur et al., 2004) including high swelling capacity, low permeability, high radionuclide retardation capacities (Yong et al., 1986; Arthur et al., 2004; Wersin et al., 2007; Villar and Lloret, 2008), micro-porous structure (Wersin et al., 2007), plasticity (Arthur et al., 2004; Wersin et al., 2007) and thermal conductivity (Arthur et al., 2004). Additionally, granulated bentonite mixtures (GBM) comprising granules (highly compressed pellets) and powders of bentonite have been proposed and opted as candidate buffer/backfill material in many recent concepts of EBS (Salo and Kukkola, 1989) including NAGRA’s concept demonstrated in the Engineered Barrier (EB) experiment (Mayor et al., 2005; Karnland et al., 2008) and Full-scale Emplacement (FE) experiment (Müller et al., 2017) at the Mont Terri underground laboratory in Switzerland. The GBM exhibit good compaction properties and are preferred owing to the operational advantages including easy transportation and in-situ placement/backfilling as well as an appropriate material to minimize the gaps between the rock and seal (Alonso et al., 2011; Guerra et al., 2017; Guerra et al., 2018).

After the emplacement in the repository, the GBM will be subjected to complex thermo-hydro-mechanical (THM) settings including heat dissipation from the waste canister and hydration processes resulting from the infiltration of water from the host rock as well as generation of gases as a result of metal corrosion or by the degradation of organic wastes. Therefore, the emplaced GBM in the EBS should be capable to develop a sufficiently high swelling pressure to maintain a good contact between the host rock and EBS, and to ensure rapid dissipation of radiogenic heat.
as well as hold enough permeability to allow the transport of gases without cracking and rupturing of the EBS (Arthur et al., 2004). It is well documented that the THM behavior of bentonite is related to its microstructure changes (Delage, 2007; Romero and Simms, 2008). Alonso et al., 2011 also reported that the macroscopic behavior of the compacted granular bentonite mixtures is related to its microstructure. Therefore, a good understanding of the behavior of GBM is essential to design an efficient EBS and to achieve optimum performance during the operation of repository. The objective of this study is to investigate the microstructural features using a microfocus X-ray computerized tomography (MFXCT) apparatus and mass transport characteristics (with a focus on gas and heat transport) of the GBM under the varied dry density and relative humidity-controlled conditions.

2 MATERIAL AND METHODS

2.1 Material and sample preparation

The material used in this study was prepared from granulated bentonite (trade name: OK bentonite, Kunimine Industries, Japan). The GBM samples were prepared by crushing and sieving the original bentonite sample having the grain size ranges between 0.075 mm and 10 mm. The particle size distribution curve was adjusted to follow the fuller grading curve (Mayor et al., 2005). Fig. 1 shows the adopted particle size distribution curve for tested GBM in this study. The tested GBM samples were prepared to study the effect of packing density and relative humidity on the microstructural features and mass transport characteristics. For measurements under density effects, air-dried (A.D.) GBM samples having water content (w.c.) of 6.38% by weight (at RH 60%) were packed in a cylindrical core of 100 cm$^3$ at six different dry densities (DD) ranging from 1.25 to 1.75 g/cm$^3$. On the other hand, the samples for humidity-controlled conditions were prepared at 1.45 g/cm$^3$ dry density (RH = 60% and 6.38% w.c.) and stored under controlled humidity at RH = 90 % to carry out measurements over a time period of twelve (12) weeks. Triplicate samples were prepared for both DD-controlled and RH-controlled measurements.

![Fig. 1. Particle size distribution curve for OK-GBM.](image)

2.2 Microstructural visualization

For visualizing the microstructure, a microfocus X-ray computerized tomography (MFXCT) apparatus (inspeXio SMX-90CT, Shimadzu Co Ltd. Japan) was used to scan the packed GBM samples. The samples were scanned at a resolution of 20 µm/voxel. The scanned slices were reconstructed to create a Multiplanar Reconstruction (MPR) image. The MPR images were visualized in three dimensions using an analysis software ExFact VR 2.1 (Nihon Visual Science Inc., Japan).

2.3 Measurement of mass transport parameters

After the MFXCT scanning, the GBM samples were used to measure the gas and heat transport parameters; gas diffusion coefficient ($D_g$), air permeability ($k_a$), thermal conductivity ($\lambda$) and volumetric heat capacity (C). The $D_g$ was measured by a diffusion chamber method (Currie, 1960; Rolston and Moldrup, 2002). The oxygen was used as a tracer gas and measured as a function of time in the diffusion chamber. In order to calculate the gas diffusivity ($D_g/D_0$), the gas diffusion coefficient for oxygen in free air ($D_0$) at 20°C was taken as 0.20 cm$^2$/s (Currie, 1960; Glinski and Stepniewski, 1985). The $k_a$ was measured using an air permeameter by flowing air through the packed GBM core at three flow rates (each flow rate falling within 0.2–2.3, 1.7–10.3, and 5.7–60 dm$^3$/min, respectively). Darcy’s equation was applied to calculate $k_a$ based on the pressure difference across the core and the viscosity of the air (1.86 x 10$^{-5}$ Pa s) (Iversen et al., 2001). The $\lambda$ and C were measured using a portable dual-needle probe analyzer (KD2-Pro, Decagon Devices Inc., Pullman, WA, USA).

3 RESULTS AND DISCUSSION

3.1 Microstructural analyses

The typical scanned MRP images for both varying dry density-controlled (DD-controlled) samples and relative humidity-controlled (RH=90%) samples are shown in Fig. 2 and Fig. 3, respectively. The measured CT brightness was plotted as a function of dry density for DD-controlled samples (Fig. 4) and time for RH-controlled samples (RH= 90%) (Fig. 5). Figure 4 shows that as density increases, CT brightness increases. However, no significant change has been noted in the CT brightness values for RH-controlled samples over a period of 12 weeks (Fig.5). In the histograms of CT brightness (Fig. 6), two peaks were observed for the lower dry density samples; the peak at low brightness indicates that fine particles are loosely distributed whereas the other peak at high brightness corresponds to the larger pellets with a higher dry density. Additionally, the peaks merged into a single peak with increasing dry density. On the other hand, single peak was observed in the histograms of the CT brightness in all the RH-controlled samples (Fig. 7).
Fig. 2. Typical scanned MPR images of DD-controlled samples.

(a) DD = 1.25g/cm$^3$  
(b) DD = 1.75g/cm$^3$

Fig. 3. MPR images of A.D. (RH= 60%) and 8 weeks sample at RH= 90%.

- Fig. 4. Measured CT brightness for DD-controlled samples. The average and standard deviations are measured from three Regions of Interest (ROIs) inside each scanned sample.

- Fig. 5. Measured CT brightness for RH-controlled samples (RH=90%).

- Fig. 6. Histograms of CT brightness for DD-controlled samples ranging from 1.25g/cm$^3$ to 1.75g/cm$^3$.

- Fig. 7. Histograms of CT brightness for RH-controlled samples (RH=90%)

### 3.2 Mass transport parameters

**Gas diffusivity and air permeability**

The measured gas transport parameters, $D_p/D_0$ and $k_a$, were plotted as a function of air content ($\epsilon$) and shown in Figs. 8 and 9. Higher $D_p/D_0$ values were observed at higher $\epsilon$, and it was found that there was not significant difference between DD- and RH-controlled samples. Similar to $D_p/D_0$, the measured $k_a$ increased with increasing in $\epsilon$, and a small difference between DD- and RH-controlled samples.

- Fig. 8. Measured gas diffusivity ($D_p/D_0$) as a function of air content ($\epsilon$).
Fig. 9: Measured air permeability ($k_a$) as a function of air content ($\varepsilon$).

**Thermal properties**

The measured thermal properties such as thermal conductivity ($\lambda$) and volumetric heat capacity ($C$) were plotted as a function of volumetric water content ($\theta$) and air content ($\varepsilon$). Figs. 10 and 11 showed that both $\lambda$ and $C$ increased with increasing in $\theta$. Conversely, with increasing $\varepsilon$, $\lambda$ and $C$ decreased (Figs. 12 and 13). The results indicated that the both compaction density (i.e., DD) and RH conditions affected the $\lambda$ and $C$ values, resulting in higher $\lambda$ and $C$ values were observed for RH-controlled samples.

**CONCLUSIONS**

This study examined the effect of DD and RH on the microstructure and mass transport characteristics of Granulated Bentonite Mixtures (GBM). The MFXCT observations indicated that DD was an important factor that governed the microstructure of GBM. The histograms of CT brightness yielded a productive insight to understand the internal distribution of density of GBM. Both $D_p/D_0$ and $k_a$ increased with the increase in air content, and there was not significant difference between DD- and RH-controlled samples. On the other hand, the measured $\lambda$ and $C$ varied depending on both DD and RH, and the measured $\lambda$ and $C$ increased with the increase in volumetric water content (the decrease in air content). Further investigations and analyses are needed to understand the pore network characteristics of the GBM that would lead to refine the mass transport parameters.
of GBM and to establish an effective link between mass transport parameters and micro-structural parameters.

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