Gas breakthrough tests on saturated GMZ01 bentonite using RCP technique with consideration of dry
density effect

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ABSTRACT

In this study, gas breakthrough tests were conducted on GMZ01 bentonite specimens with dry densities 1.3, 1.5 and 1.7 Mg/m^3 under rigid boundary conditions using the residual capillary pressure (RCP) technique. Prior to the gas breakthrough tests, water permeability tests were performed for determining intrinsic water permeability and establishing full water saturation for the follow-up gas breakthrough tests. The intrinsic water permeability measured ranges between 11 and 150 nDarcy (1.10×10^{-20} and 1.50×10^{-19} m^2). As dry density increases, the residual capillary pressure differences of the initially water-saturated bentonite specimens recorded increase from 0.12 to 0.61 MPa. Meanwhile, the time to breakthrough was recorded to increase sharply as dry density increases. In addition, the maximum effective gas permeability decreases from 8.91×10^{-18} to 8.92×10^{-19} m^2 as dry density increases from 1.3 to 1.7 Mg/m^3. These results indicate that the capillary drainage in the bentonite specimen is highly related to the capillarity effects in the largest pores. The higher dry density, the smaller flow channels, resulting in more difficult for gas phase to dispel water from the interconnected flow pathways and a better sealing capacity with a higher residual capillary pressure.

Keywords: GMZ bentonite, RCP technique, gas breakthrough, sealing capacity, dry density effect

1 INTRODUCTION

In a conceptual multi-barrier deep geologic repository for disposal of high-level radioactive waste (HLW), bentonite has been selected as a potential buffer/backfill material, due to its low permeability, high swelling and favorable radionuclide retardation capacities (Villar et al., 2010). Literatures show that, during the long-term operation of a repository, gas could be generated and accumulated in the artificial barrier system, due to the corrosion of the metallic waste canister, micro-biological degradation and radiolysis of water (Horseman et al., 1999; Birgersson et al., 2008; Ye et al., 2009). The gas accumulated in the repository could induce increases of gas pressure to a very high level that could eventually damage the sealing system, due to the low permeability of bentonite. In this regard, determination of gas transport parameters and evaluation of the sealing capacity of bentonite system are of great importance to assess the long-term operation safety of a deep geological disposal repository.

Gas breakthrough pressure is an important parameter to characterize the ability of a saturated porous medium with a non-wetting phase to dispel the wetting phase from its pore system. Generally, magnitude of gas breakthrough pressure is determined by the capillary pressure at which the wetting phase is displaced from the pore network and continuous flow pathways for the non-wetting phase formed in the specimen. Therefore, the capillary entry pressure in a cylindrical tube with radius \( r \) can be given by the Laplace’s law,

\[
P_c = P_g - P_w = \frac{2\sigma \cos(\theta)}{r}
\]

Where, \( P_g \) and \( P_w \) are the gas pressure (non-wetting phase) and the water pressure (wetting phase), respectively.
respectively, $\sigma_w$ is the surface tension ($N\cdot m^{-1}$) and $\theta$ is contact angle which highly depends on the basic properties of the two phases contacted.

When the pressure difference between the gas phase and the water phase surpasses the capillary entry pressure ($P_c$), the gas phase starts to displace the water phase from the pore space, leading to reduction of water saturation. After gas breakthrough phenomenon happens, the difference between the pressures at two ends of the specimen decreases and ultimately stabilizes at a definite value, which is denoted as the residual capillary pressure (Hildenbrand et al., 2002; Thomas et al., 1968).

For measurement of the gas breakthrough pressure, the step-by-step approach was commonly used, by which the gas pressure at the upstream surface of a fully saturated specimen was increased step-by-step and the gas production at the downstream of the specimen was recorded (Li et al., 2005). However, this approach has problems in result accuracy and test duration. In this regard, the residual pressure approach was proposed and performed by instantaneously imposing a high gas-pressure gradient across the bentonite specimen and monitoring the variations of gas pressures at both ends of the specimen (Hildenbrand et al. 2002).

Dry density is one of the important factors influencing gas migration behavior in compacted bentonite. Contributions have been made to investigate the dry density effects on threshold capillary pressure of bentonite specimens (Liu et al., 2014; Xu et al., 2017). Graham et al. (2002) performed gas breakthrough tests on illite and bentonite specimens and reported that the gas breakthrough pressures increased as clay density increased. Push et al. (1985) also reported that for specimens with lower dry densities, the capillary drainage effect induced lower gas breakthrough pressure. However, for specimens with higher dry densities, the mechanical effect became significant, accompanied by higher gas breakthrough pressures. Generally, for specimens with lower dry density, larger pores would facilitate gas migration. In contrary, for specimens with higher density, it became more difficult for gas to pass through the narrow pore networks due to high capillary resistances. Moreover, the geometrical arrangements of clay particles and organization of water molecules could be influenced by dry density.

In this study, water permeability and gas breakthrough tests were successively conducted on bentonite specimens with dry densities of 1.3, 1.5, and 1.7 Mg/m$^3$ using the RCP (residual capillary pressure) approach. Intrinsic water permeability was measured and influence of dry density on the residual capillary pressure was analyzed.

2  MATERIALS AND SPECIMEN PREPARATION

The material tested in the present work was GMZ01 bentonite, which was taken from Gaomiaozi in the Inner Mongolia Autonomous Region, China, 300 km northwest from Beijing (Ye et al. 2009). Bentonite has been considered as buffer/backfill materials in the Chinese deep geological disposal program for HLW due to its favorable physical and mineralogical properties. The fundamental properties of GMZ01 bentonite have already been reported elsewhere (Table 1) (Wen, 2006). A high cation exchange capacity and adsorption ability can be identified.

Table 1. Basic properties of GMZ bentonite (Wen 2006).

| Property                        | Description       |
|--------------------------------|-------------------|
| Specific gravity of soil grain | 2.66              |
| pH                             | 8.68–9.86        |
| Liquid limit (%)               | 276               |
| Plastic limit (%)              | 37                |
| Total specific surface area    | 597               |
| (m2/g)                         |                   |
| Cation exchange capacity       | 77.3              |
| (mmol/100 g)                   |                   |
| Na+ (43.36), Ca2+ (29.14), Mg2+ (12.33), K+ (2.51) | |
| Main minerals                  | Montmorillonite (75.4%), quartz (11.7%), feldspar (4.3%), cristobalite (7.3%) |

For specimen preparation, the GMZ01 bentonite powder was equilibrated to an initial water content 10.7%. Then, according to the target dry density and dimensions of a cylindrical specimen (with a height of 10.79 mm and a diameter of 50.15 mm), the powder was weighed and put into a custom-designed mold. Statically compaction was conducted with a piston at a constant rate of 0.3 mm/min. When the designed dimension of the specimen was reached, compaction was stopped and the vertical load was kept for 1 hour for improving the homogeneity. Procedures mentioned above were repeated until specimens with dry densities of 1.3, 1.5 and 1.7 Mg/cm$^3$ were compacted.

3  EXPERIMENTAL INVESTIGATIONS

3.1 Apparatus

The conceptual sketch of the testing equipment was presented in Fig. 1. This equipment includes four components: a rigid boundary cell, a helium injection system and a receiving compartment, as well as a data logger. The gas injection system includes a helium tank, a gas compartment, and a digital pressure transducer etc. The receiving apparatus at the downstream is a compartment with a digital pressure transducer. The volume of the two compartments both at the upstream and the downstream was fixed at 100 cm$^3$. The data logger was designed for monitoring the gas pressure at
the two compartments through the digital pressure transducers.

Fig. 1. Equipment for gas injection tests using the residual capillary pressure approach.

3.2 Water permeability test

Prior to the gas breakthrough tests, the steady state water permeability tests were performed for determining the intrinsic water permeability of the specimens. With the volume-pressure controller (Fig. 1), distilled water was injected with 1 MPa from the inlet in the top end of the specimen and the water injected was recorded simultaneously. When the water injected stabilized, the steady state water permeability test was finished and the bentonite specimen was considered as saturated. Then, the specimen was submitted for conducting the follow up gas breakthrough test.

The intrinsic water permeability of the bentonite specimen can be calculated using the Darcy’s law:

\[ k_{iw} = -\frac{\eta Q}{A} \frac{dx}{dp} \]  

(2)

Where, \( \eta \) is the viscosity of water (\( P_a \cdot s \)), \( Q \) is the flux (\( m/s \)), \( A \) is the cross-section area of the bentonite specimen and \( dx/dp \) represents the reciprocal of the pressure gradient.

3.3 Gas breakthrough tests

In this study, the residual capillary pressure approach firstly proposed by Hildenbrand et al. (2002) was employed to measure the residual capillary pressure using helium in an initially saturated bentonite specimen.

After the water permeability test, an instantaneous pressure difference was imposed on the saturated specimen and changes of gas pressures both in the upstream and downstream compartments were monitored. Based on these, the gas flux through the specimen was calculated by means of changes of the pressure in the upstream and downstream compartments. During the test, the injection pressure applied at the upstream compartment decreased and that in the downstream compartment increased. Finally, a residual capillary pressure difference between the two compartments reached a stabilized value, which was also defined as the ‘snap-off’ pressure. Meanwhile, the effective gas permeability can be calculated through the pressure curves of the two compartments using Darcy’s law (Hildenbrand et al., 2002).

\[ k_{ef} = \frac{V_2 \cdot \mu_2 \cdot 2 \cdot L}{A \cdot (P_2^2 - P_1^2)} \frac{dP_2}{dt} \]  

(3)

Where, \( k_{ef} \) is the effective gas permeability, \( V_2 \) is the volume of the downstream compartment, \( \mu_2 \) is the viscosity of water, \( L \) is the length of the bentonite specimen, \( A \) is the cross-section area of the specimen, \( P_1 \) and \( P_2 \) represent the gas pressures in the upstream and downstream compartments, respectively.

All the water permeability tests and gas breakthrough tests were performed at an ambient temperature about 20 ± 0.1 °C.

4 RESULTS AND DISCUSSION

Evolution curves of gas pressures with time measured in both upstream and downstream compartments for bentonite specimens with different dry densities were shown in Fig. 2.

Results of water permeability tests and gas injection tests conducted on compacted bentonite specimens with different dry densities were presented in Table 2.
Initial pressure difference = 8.41 MPa

Fig. 2. Evolutions of gas pressures in both upstream and downstream compartments for bentonite specimens with different dry densities.

Table 2. Specifications of water permeability tests and gas breakthrough tests

| Specimen | Dry density (Mg/cm³) | Water permeability tests | Gas breakthrough test |
|----------|----------------------|-------------------------|----------------------|
|          |                      | $k_{im}$ (m²)           | $k_{eff(max)}$ (m²)  | $P_{snap-off}$ (MPa) |
| 1        | 1.3                  | 1.50×10⁻¹⁹              | 8.91×10⁻¹⁸           | 0.12                 |
| 2        | 1.5                  | 3.41×10⁻²⁰              | 4.40×10⁻¹⁸           | 0.37                 |
| 3        | 1.7                  | 1.10×10⁻²⁰              | 8.52×10⁻¹⁹           | 0.67                 |

Where, $k_{im}$ represent the intrinsic permeability; $k_{eff(max)}$ represents the maximum gas permeability observed at the gas breakthrough; $P_{snap-off}$ represents the residual capillary pressure difference at the end of the gas breakthrough test.

4.1 Water permeability tests

The intrinsic water permeability, measured with deionized water at room temperature (20 ± 0.1°C) and fluid pressure at 1MPa, ranges between 1.10 × 10⁻²⁰ to 1.50 × 10⁻¹⁹ m² (Table 2). The intrinsic water permeability increases sharply as dry density increases, which is consistent with the observation given by Xu et al. (2017).

4.2 Gas breakthrough tests

4.2.1 Residual capillary pressure difference

The residual capillary pressure differences (‘snap-off’ pressures), which were observed at the final stage of the gas breakthrough tests conducted on specimens with different dry densities, were shown in Fig. 2 and Table 2. It could be observed that during the gas breakthrough test conducted on the specimen with dry density of 1.3 Mg/cm³, the upstream and downstream pressures converged and stabilized at a residual capillary pressure difference of 0.12 MPa. Similarly, curves in Fig. 2b and 2c also provided residual capillary pressure differences with 0.37 and 0.61 MPa for specimens with dry densities of 1.5 and 1.7 Mg/cm³, respectively. Results showed that the residual capillary pressure difference increased as the dry density increased. Literature point out that, the residual capillary pressure corresponds to the endpoint of the spontaneous imbibition process that the wetting phase shut off the last interconnected pore channels, which consist of the larger pores in compacted bentonite specimens (Hildenbrand et al., 2013). In addition, the residual capillary pressure highly depends on the intrinsic water permeability or pore throat size (Bush and Hildenbrand, 2013). This deduction was confirmed by Hildenbrand et al. (2013), who observed a decrease of the residual capillary pressure from 2.7 to 0.6 MPa for specimens with intrinsic permeability increasing from 1.5×10⁻²⁰ m² (Boom clay) to 3.8×10⁻¹⁸ m² (limestone). In this regard, the increase of the dry density with lower intrinsic water permeability and smaller macro-pores in the distribution induce a high residual capillary pressure.

It could also be observed that the breakthrough phenomenon occurred after 216, 335 and 394 hours of the gas breakthrough tests conducted on specimens with dry densities 1.3, 1.5 and 1.7 Mg/cm³ (Fig. 2), respectively. These observations indicated that the time to breakthrough increased as the dry density increased. Graham et al. (2002) reported that, the gas breakthrough will occur when the flow pathways were fully occupied by the gas phase. Generally, the flow pathways consist of the largest interconnected pores, which induce the least capillary resistance (Amann-Hildenbrand et al., 2013). In this regard, the time to breakthrough is highly related to the rate at which the water phase was dispelled from the flow channels by advective flow, that is, to the pressure gradient and the hydraulic conductivity.

4.2.2 The maximum effective gas permeability

The maximum effective gas permeability obtained at the occurrence of the breakthrough are presented in Fig. 2 and Table 2. Results showed that the range of the effective gas permeability extended from 8.9×10⁻¹⁸ m² to 8.5×10⁻¹⁵ m². It can be concluded that the maximum effective gas permeability decreased sharply as the dry density increased, which was consistent with the observation by Gallé. (1998) who noted a decrease in gas permeability values with three orders of magnitude for Fo-Ca clay specimens with dry density increasing from 1.6 to 1.9 Mg/m³. Explanation to this phenomenon could be that, the effective gas permeability was highly related to the porosity (Gallé, 1998). With a specific density of 2.66, the porosities of bentonite material compacted at dry densities of 1.3, 1.5 and 1.7 are 36.09, 43.61 and 51.29%, respectively. In addition, higher density will produce smaller flow channels and a higher gas entry pressure, which was difficult for gas to dispel water and migrate in the water-saturated pathways.
5 CONCLUSIONS

In this study, a series of water permeability and gas breakthrough tests were conducted and test results were analyzed.

Intrinsic water permeability measured for specimens with dry densities 1.3, 1.5 and 1.7 Mg/cm³ ranges from $1.10 \times 10^{-20}$ to $1.50 \times 10^{-19}$ m². The residual capillary pressure difference observed at the final stage of the gas breakthrough tests increases as the dry density increases. Explanations to these observations could be that higher dry density induces smaller pores resulting in higher residual capillary pressure. In the meantime, the time to breakthrough also showed an increase trend as the dry density increased, which highly depended on the hydraulic conductivity and the pressure gradient.

The maximum effective gas permeability recorded at the occurrence of the breakthrough during the gas breakthrough tests decreased with increasing dry density. This observation could be resulted from the higher dry density accompanied by smaller flow channels, which was difficult to disel water from the flow channels.

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