Gas breakthrough in saturated compacted GMZ bentonite under rigid boundary conditions

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Gas breakthrough in saturated compacted GMZ bentonite
under rigid boundary conditions

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Abstract: In a geological repository for disposal of high-level radioactive waste, gas breakthrough is an important phenomenon during gas migration process in the saturated engineered barrier. In this paper, gas injection, swelling pressure, water permeability and water retention tests were conducted on saturated compacted GMZ bentonite to investigate the gas breakthrough mechanism. Results show that, for saturated GMZ bentonite tested under rigid boundary conditions, the gas breakthrough pressure is significantly larger than the swelling pressure and slightly lower than the gas entry pressure obtained from the water retention characteristic and the van Genuchten model. Gas breakthrough pressure deviates from the swelling pressure and approaches the calculated gas entry pressure as the dry density increases. Mechanical and capillary effects are both important to gas migration process for specimens with lower dry densities, and the capillary effect becomes more important with the increase of dry density. The desaturation and shrinkage of the specimen will result in unexpectedly high and disordered interfacial gas flux. For specimens with higher dry densities, gas will only flow through interconnected larger pores, then results in minor desaturation/shrinkage of the specimen. Finally, a new model with consideration of both mechanical and capillary effects is proposed, which can accurately predict gas breakthrough pressure for GMZ bentonite specimen.

Key words: gas breakthrough; GMZ bentonite; mechanical effect; capillary effect; interfacial gas flux

Introduction

A promising option in managing the high-level radioactive waste is resort to the deep geological disposal technique, which commonly adopts the multi-barrier system including the vitrification, waste canister, buffer/backfill material and geological host
rock to effectively isolate the radioactive substance from the biosphere (NEA 2001; Komine 2004; Alonso 2006; Davy et al. 2009). As an important component of the multi-barrier system, bentonite is preferentially adopted in most countries for using as the buffer/backfill materials due to its proper swelling property, low permeability and high absorptive capacity (Komine et al. 2009; Villar et al. 2004; Ye et al. 2009; Wang et al. 2012). One of the key issues in safety assessment of deep geological disposal repository is that the corrosion of metallic waste canister as well as the radiolysis of water and degradation of organic materials, will lead to the generation of gases within the repositories. Due to the high capillary sealing efficiency of bentonite, gas may be accumulated leading to increases of the gas pressure. This pressure increase could deteriorate the sealing efficiency of the barrier system, thus the repository. Therefore, investigations on gas flow behaviors in saturated compacted bentonite have attracted intensive attentions during the past decades (Bonin et al. 2000; NEA 2001; Ortiz et al. 2002; Senger et al. 2008; Yu and Weetjens 2009).

Pusch et al. (1985) conducted systemic gas migration tests on MX80 bentonite and firstly reported the occurrence of gas breakthrough, i.e., gas will not flow through the saturated bentonite until gas injection pressure reached a threshold value (gas breakthrough pressure), after that, gas flux will sharply increase. This flow characteristic was confirmed by subsequent researches conducted on various ultra-low or low permeability materials (Tanai et al. 1997; Gallé 1998; Horseman et al. 1999; Gerard et al. 2014). In further analysis, Horseman et al. (1999) and Gerard et al. (2014) reported that, owing to the extremely large capillary resistance of the ultra-low permeability materials, gas breakthrough was possibly caused by the mechanical-induced dilatancy of flow pathways. For examination of this mechanism, Harrington et al. (2012) designed a tracer experiment by injecting helium gas with mixed gold
and TiO$_2$ nanoparticles into a Boom clay specimen. Test results confirmed the localized distribution of the pressure-induced dilatant pathways (Fig. 1). From the mechanical point of view, gas breakthrough pressure was usually regarded to be equivalent to the swelling pressure (plus the value of the interstitial water pressure if it was not zero) (Tanai et al. 1997; Horseman et al. 1999; Gallé 1998; Navarro 2009).

Unlike the mechanism described in the pathway-dilatancy gas flow, Ortiz et al. (1996), Graham et al. (2002), Hildenbrand et al. (2002, 2012, 2013) and Busch et al. (2013) emphasized the capillary effect on gas flow process. Busch et al. (2013) and Hildenbrand et al. (2013) stated that the occurrence of gas breakthrough referred to the moment at which the liquid phase was displaced by the gas phase to form a continuous flow pathway across the specimens (Fig. 2). Graham et al. (2002) additionally pointed out that, although the capillary resistance of the entire specimen was extremely large, gas migration may act in discrete channels consisting of interconnected larger pores. Liu et al. (2014, 2015) observed expulsion of mixed water/gas bubbles in gas migration tests and concluded that, besides the pathway-dilatancy, capillary effects still controlled gas migration through ultra-low permeability materials, especially for tests under quasi-rigid boundary conditions.

In further investigation of gas breakthrough, Liu et al. (2014) designed gas migration tests using tubes with smooth and grooved inner surface, respectively. Results showed that, the swelling pressures (with an average value of 7.32MPa) of the specimens were closed to the gas breakthrough pressures determined on tests using smooth inner surface (between 7 MPa and 8MPa) and extremely lower than those determined on tests using grooved inner surface (>10MPa). The final conclusion of
the high dependence of gas breakthrough pressure on the swelling pressure was possibly attributed to the preferential gas flow along the interface between the testing cell and the specimen other than that through the ultra-low permeability material itself (Fig. 3). Davy et al. (2009) also designed special tests using obturators to allow gas flow through the specimen/tube interface rather than the specimen itself. Results showed that gas breakthrough pressure determined with obturators was 3.6MPa, which was almost equaled to the gas breakthrough pressure determined without the obturators (4-4.5 MPa). This observation confirmed the interfacial leakage during gas migration process. Xu et al. (2015) performed gas injection tests on low permeability materials and reported the similar results. According to these findings, this interfacial gas flow as gas injection pressure approaching/overcoming the swelling pressure may not be ignored.

To sum up, there are three possible gas breakthrough mechanisms, including the mechanical effect induced pathway-dilatant flow, the capillary effect induced continuous flow, as well as the interfacial gas flow, raised by different researchers (Figs. 1, 2 and 3). The different roles they play during the gas migration process should be investigated in further details. In this paper, stepwise gas injection tests, as well as swelling pressure, water permeability and water retention tests were conducted on saturated compacted GMZ bentonite specimens under rigid boundary conditions, in order to comprehensively investigate the gas breakthrough mechanism.

Experimental investigation

Materials and specimen preparation
The GMZ bentonite, which is extracted from the GaoMiaoZi county in the Inner Mongolia Autonomous Region, 300 km northwest from Beijing, China (Ye et al. 2009), is selected preferentially as the component of the buffer/backfill materials for the repository in China. It is a light gray powder, dominated by montmorillonite (75.4% in mass). The fundamental properties of the GMZ bentonite have already been reported (Table 1) (Wen 2006). A high cation exchange capacity and adsorption ability can be identified.

The GMZ bentonite powder with initial water content of 10.70% is prepared by homogenizing it in sealed containers in which saturated saline solutions have been used to control the relative humidity (Tang and Cui 2005). Then, weighed mixture powder is put into a mould ring and compacted statically with a piston at a rate of 0.4 kN/min. Target cylindrical specimens with a height of 10.79 mm, a diameter of 50.15 mm and corresponding dry densities of 1.3, 1.4, 1.5, 1.6 and 1.7 Mg/m$^3$ are compacted for the swelling pressure, water permeability and gas injection tests. Specimens with a height of 2 mm, a diameter of 6 mm and corresponding dry densities of 1.3, 1.5, 1.7 and 1.9 Mg/m$^3$ are compacted for water retention tests.

**Test apparatus and procedures**

**Swelling pressure and water permeability tests**

The experimental apparatus for swelling pressure and water permeability tests is schematically presented in Fig. 4. It is composed of three parts: a stainless steel testing cell assembled to guarantee the specimens tested under rigid boundary conditions; a liquid volume/pressure controller (with accuracy of ±1 mm$^3$ in volume and ±1 kPa in pressure) connected to the top of the specimen for applying constant water injection pressure and recording the volume of water injected; a load sensor
along with the data logger for recording the evolution of swelling pressure of the specimen during hydration process.

The prepared specimen is assembled into the test setup shown in Fig. 4. Deionized water is infiltrated into the specimen under a pressure, which is gradually increased to and maintains at 1MPa throughout the test using a volume/pressure controller. The swelling pressure is recorded by the load sensor and the volume of water injected is recorded by the controller simultaneously. The swelling pressure test is stopped until the swelling pressure reaches stable. Then, the hydraulic conductivity test is conducted using the constant head method. When the water flux recorded by the volume/pressure controller reaches constant values, the water permeability test is finished. Then, the intrinsic permeability is determined using Darcy’s law.

Gas injection test

After the swelling pressure and water permeability tests, the inlet at the bottom of the cell is connected to the gas injection unit that consisted of a helium cylinder, a helium booster pump, a regulator, the inlet buffer tank and digital pressure gauge sequentially, while the conduit connected to the top of the specimen is replaced by the multi-gas mass flowmeters with a reservoir in advanced for separating the mixed water/gas phases. The assembled setup is schematically presented in Fig. 5(a).

Step-by-step gas injection method is adopted in this test, because it could reproduce more closely the in situ case by increasing gas pressure gradually and allow the observation of water repulsion and gas breakthrough (Liu et al. 2014). High-purity helium gas is injected into the specimen with increasing step of 500-1000 kPa at
intervals of 24h. The test on current specimen is finished until the “gas breakthrough” is observed.

With reference to the design of Davy et al. (2009) and Liu et al. (2014), the stainless steel porous discs are designed to be embedded into the basement and the piston respectively, in order to lead gas flow through the specimen itself rather than the annular interface between the specimen and the inner wall of the testing cell (Fig. 5(b)). Theoretically, as the surface area of the porous disc is somewhat smaller than that of the specimen, water flow in the specimen is not linear. However, due to the difference of the surface area between the specimen and porous disc is too small, a linear flow assumption is adopted in the present work. The hydraulic conductivity is calculated using the surface area of the porous disc.

Water retention test

According to the previous works, drainage/imbibition controlled saturation hysteresis behavior of compacted GMZ bentonite is insignificant (Ye et al. 2009). The difference of water retention properties between the saturation and drying processes could be ignored. Water distribution within the pore space is assumed to be independent of the drying/drainage technique. Therefore, the relationship between water saturation and capillary pressure (suction) can be used to describe the gas migration process through bentonite (Senger et al. 2008; Navarro 2009; Gerard et al. 2014; Ye et al. 2014). In this study, water retention tests on confined compacted GMZ bentonite specimens are conducted (Fig. 6).

For this purpose, two well-known suction control techniques, the osmotic technique (for suctions lower than 1.5 MPa) and the vapor phase technique (for suctions higher than 1.5 MPa), were adopted for determination of the water retention characteristics of GMZ bentonite specimens with different dry densities.
The osmotic technique was employed for low suction control. The prepared specimen was wrapped with semi-permeable membrane (Spectra/Por 4 MWCO 14000) and assembled in a rigid testing cell. After that, the assembled specimen cell was completely immersed in the PEG solution (PEG 20000). When the suction equilibrium between the specimen and the PEG solution was reached, the water content of the specimen was tested and the corresponding suction was determined through measurement of the Brix of the PEG solution (Table 2).

The vapor phase technique was employed for high suction control. The confined GMZ specimen was placed in a desiccator and the water vapour above a saturated salt solution was circulated for applying a given relative humidity on the specimen. After the suction equilibrium between the specimen and the water vapor was achieved, the water content of the specimen was measured and the corresponding suction was calculated by the Kelvin-Laplace equation (Eq. (1)) with the relative humidity generated by the saturated salt solution. Saturated salt solutions and their corresponding suction imposed are given in Table 3 (Tang and Cui 2005).

\[
s = -\frac{\rho_w R T}{M_w} \ln \frac{R H \%}{100}
\]  

Where \( s \) is suction (kPa); \( R \) is the universal (molar) gas constant (8.314 J.mol\(^{-1}\)K\(^{-1}\)); \( T \) is absolute temperature (K); \( M_w \) is the molecular mass of water vapour (18.016g/mol); \( \rho_w \) is the volumetric mass of water (kg/m\(^3\)); \( RH \) is relative humidity.

In fact, both the osmotic technique and the vapour equilibrium method are commonly employed for controlling total suction in unsaturated soil tests, detailed descriptions of the two techniques can be found elsewhere (Delage et al. 1998; Blatz

All the tests are conducted in an ambient temperature of 20±0.1℃.

Results and discussion

Gas breakthrough tests

The measured evolutions of gas flow rate with gas injection pressure for specimens with different dry densities are presented in Fig. 7.

In Fig. 7, gas breakthrough phenomena are universally observed for saturated GMZ bentonite specimens with different dry densities. During each test, almost no gas outflow could be detected until the gas breakthrough pressure (denoted in Fig. 7) is reached. After that, gas flow rate will increase sharply and the gas injection system cannot hold the defined pressure any longer, which results in the decline of gas injection pressure after gas breakthrough. In the meantime, smaller impulses of gas outflow could be observed for specimens with lower dry densities (Fig. 7(a) and (b)), while it is disappeared for those with higher dry densities (Fig. 7(c), (d) and (e)).

Similarly, investigations of liquid saturation effect on gas migration process (Gallé 1998, 2000) showed that, two critical pressures were observed for unsaturated specimen and faded to a single one as the specimen reached fully saturated. Without revealing experimental evidence, Gallé (2000) regarded that the two critical pressures were resulted from the influences of capillary and mechanical effects on gas flow, respectively. Liu et al. (2014, 2015) stated that the physical origin of the gas breakthrough was hardly determined and may be attributed to capillary or mechanical effect or both of them. The terminologies of discontinuous and continuous breakthrough were more suitable to describe the flow behavior. Generally speaking, although gas breakthroughs are widely observed in a lot of experimental researches,
the essential mechanism is still confused. In literatures, gas breakthrough mechanism is explained based on different processes: the purely capillary-controlled flow (Graham et al. 2002; Skoczylas and Davy 2011; Busch et al. 2013; Hildenbrand et al. 2015), the dilatancy-controlled flow (Horseman et al. 1999; Popp et al. 2007; Harrington et al. 2012; Gerard et al. 2014; Ye et al. 2014) and the failure of sealing efficiency induced flow (Davy et al. 2009; Liu et al. 2015; Xu et al. 2015). In different mechanisms, the gas breakthrough pressure can be related to different soil properties. Harrington et al. (1999) reported that if mechanical effect dominated the gas flow behavior, gas breakthrough will occur at a threshold gas pressure almost equal to the effective stress on the clay, which can be directly correlated to the swelling pressure for a high-swelling clay such as bentonite. This conclusion is widely accepted by previous researches (Pusch et al. 1985; Tanai et al. 1997; Horseman et al. 1999; Harrington and Horseman 2003). Meanwhile, if the gas flow is controlled by the capillary effect, gas breakthrough will occur at the critical capillary pressure corresponding to the formation of a continuous flow pathway across the entire specimen (Hildenbrand et al. 2002; Busch et al. 2013; Liu et al. 2014; Hildenbrand et al. 2015). For high-plastic saturated clayey material with high homogeneity and short height, like the scenario presented in this paper, the value of the critical capillary pressure is approximated to that of the gas entry pressure due to the small specimen size (Gallé 2000; Skoczylas and Davy 2011; Farokhpoor et al. 2013; Hildenbrand et al. 2015). Therefore, swelling pressure and gas entry pressure should be determined to analyze gas breakthrough mechanism in details, as outlined in the sections below.

Swelling pressure tests

Swelling pressure tests are conducted on specimens with different dry densities and the results are presented in Fig. 8.
Curves in Fig. 8 show that typical “double-peak” shape of the swelling pressure evolution curves can be observed for specimens with different dry densities. This phenomenon is consistent with observations reported by Zhu et al. (2013) and Imbert et al. (2006). This observation could be interpreted as that, in the initial stage of hydration, clay aggregates swell inducing collapse of the soil skeleton on suction reduction. After that, the swelling pressure increases again and reaches its final steady-state (the second peak), which results from the diffuse double layer swelling of the bentonite (Villar and Lloret 2008; Ye et al. 2013).

**Water retention tests**

For determination of the critical capillary pressure, water retention characteristics obtained using the osmotic technique and the vapor phase technique were analyzed and summarized in Fig. 9.

Curves in Fig. 9 show that, the water saturation of the specimen decreases slowly at the initial stage and drops notably once the gas entry pressure (the critical capillary pressure) is reached, which is corresponding to the formation of continuous flow pathways throughout the entire specimen (Gallé 2000; Skoczylas and Davy 2011; Farokhpoor et al. 2013; Hildenbrand et al. 2015). In addition, the drainage process will be accelerated with the decrease of dry density. For such a small size specimen, the critical capillary pressure would be roughly equivalent to the calculated gas entry pressure determined from the water retention curves using a mathematical model, such as the van Genuchten model. Therefore, the van Genuchten model (Eq. (2)) (van Genuchten 1980) is adopted for obtaining the calculated gas entry pressures, $P_{entry}$, of the specimen.
\[
S_e = \left\{ 1 + \left[ \frac{P_g - P_w}{P_{entry}} \right]^m \right\}^{\frac{1-m}{m}}
\]

Here, \( S_e \) is the effective degree of saturation, which is calculated by \( S_e = \frac{S_{sw} - S_{gr}}{1 - S_w - S_{sw}} \), \( S_{sw} \) and \( S_{gr} \) are the residual saturation degree of the water and gas phases, respectively; \( m \) is the shape factor of van Genuchten model; \( P_w \) is water pressure and \( P_{entry} \) is the calculated gas entry pressure.

The fitted results of the water retention properties are presented in Fig. 9. Related parameters for the van Genuchten model are listed in Table 4. According to the calculated gas entry pressures determined for specimens with dry densities 1.3, 1.5, 1.7 and 1.9 Mg/m\(^3\), a fitting curve can be developed and presented in Fig. 10. The calculated gas entry pressures for specimens with dry densities 1.4 and 1.6 Mg/m\(^3\) are obtained to be 3.181 MPa and 6.024 MPa, respectively.

**Comparison of critical pressures**

Gas breakthrough normally indicates the dynamic gas flow and drainage process, the corresponding pressure is usually determined as the critical pressure related to the occurrence of sharp gas outflow and pressure drop in step-by-step gas injection test. Swelling pressure is obtained as the final stable pressure after water flooding is stopped in swelling pressure test. The calculated gas entry pressure is defined as the critical capillary pressure corresponding to the formation of a continuous flow pathway across the entire specimen (without dilatancy), and is derived from the water retention tests (in static conditions, ignoring drainage/imbibition hysteresis) and the van Genuchten model in present study due to the small specimen size. A comparison between these three critical pressures is given in Figure 11.
Results in Fig. 11 clearly show that the gas breakthrough pressure is larger than the swelling pressure at a difference of 0.5~2.5MPa and lower than the calculated gas entry pressure with a difference of 0.5~1.2MPa. As the dry density increases, gas breakthrough pressure deviates from the swelling pressure and approaches the calculated gas entry pressure. The calculated gas entry pressure is higher than the breakthrough pressure may be due to the overestimation of the van Genuchten approach or the mixed mechanical and capillary effects. It is clear that, if no changes occur in the microstructure (without dilatancy of the flow pathway), gas breakthrough pressure should be roughly equal to the calculated gas entry pressure. However, the mechanically induced dilatancy of flow pathways (reflected by the swelling pressure) will reduce the gas breakthrough pressure (Gerard et al., 2014). Results in Fig. 11 indicate that, for specimens with lower dry densities (with larger pore spaces and good pore connectivity), the gas flow behavior will be controlled by both the capillary effect and the mechanical effect. With increasing dry density, formation of the localized dilatancy of flow pathways becomes more and more difficult due to the strong interaction between the pore liquid and clay minerals, as well as the limited deformation space restricted by the rigid boundary conditions (Birgersson et al. 2008). Under this circumstance, the capillary effect is dominant in controlling gas flow behavior.

This conclusion is inconsistent with some previous conclusions, which speculated that the capillary effect was more important for specimens with lower dry densities, while the mechanical effect was extremely significant for specimens with higher densities (Pusch et al. 1985; Harrington and Horseman 2003). However, it should be noted that, for highly compacted bentonite specimen, the capillary effect is
still important although the critical capillary pressure of the entire specimen is extremely large. The capillary induced drainage process may not extend over the entire specimen, but can only act in regions with relatively large-size pores, then gas breakthrough pressure is a little lower than the calculated gas entry pressure for specimen with highest dry density as shown in Fig. 11. Experimental researches conducted by Graham et al. (2002) also supported this result that gas breakthrough mechanism strongly depends on the test boundary conditions. Capillary flow is appeared to be the most likely process for specimens tested under rigid boundary conditions. Based on the observation of water-gas mixtures expelled from the specimens, Liu et al. (2014; 2015) also emphasized the importance of capillary effect on gas migration process.

**Confirmation of interfacial gas flow**

Curves in Fig. 7 also show that, after gas breakthrough, gas flow rates increase to unexpectedly high values (over 2000 ml/min) and present disorder relationships with dry densities. Similar phenomena were observed by Gallé (1998, 2000). This observation may be attributed to the specimen shrinkage on drying due to the significant capillary effect on gas migration process and the dry gas adopted for the test. This shrinkage will result in generation of flow pathways in the interface between the specimen and the inner wall of the testing cell (Fig. 12). Consequently, the gas flow rate can increase sharply to unexpected values. For verification, after the gas injection tests, the diameter and water content of the specimens are cautiously measured. At the same time, water permeability tests on additional parallel specimens are conducted. The diameter and water content of the specimen are carefully measured at the end of each test. Then, based on the diameters and water contents of the specimens measured, the evolutions of the water saturation...
degree and the diameter (during gas injection test) for specimens with different dry densities are obtained and presented in Figs. 13 and 14, respectively.

Curve in Fig. 13 shows that, after gas breakthrough, gas enters the specimen and dispels the pore water, results in desiccation of the specimen, which confirms the important role of capillary effect plays during gas migration process. With the dry density increasing, discharge of pore water becomes more difficult due to the formation of extremely narrow and tortuous flow pathways. Results in Fig. 14 confirm the specimen shrinkage after the gas breakthrough and show that, for specimen with lower dry densities, the shrinkage is more significant. This is also consistent with previous conclusions that, for specimens with lower dry densities, the capillary pressure can take significant effects on the entire specimen and result in notable shrinkage/desaturation of the specimen. While for specimens with higher dry densities, gas flow pathways will be concentrated in local interconnected larger pores, which will result in lower level of desaturation and shrinkage.

**Gas breakthrough mechanism**

Analyses show that the mechanical and capillary effects, as well as the interfacial effect, all play important roles in gas breakthrough. The occurrence of gas breakthrough highly depends on the soil properties. The diagrammatic sketch of the gas breakthrough mechanism in saturated GMZ bentonite specimens under rigid boundary conditions is shown in Fig. 15.

For specimens with relatively low dry densities (Fig. 15 (a)), the existence of larger pores and good pore connectivity will easily induce the formation of flow pathways through dispelling of free water (capillary effect) and local dilatancy of flow
channels (mechanical effect) with the increase of gas injection pressure. Therefore, the capillary and mechanical effects both play significant roles on gas migration process (Fig. 11 and Fig. 15 (b, c)). With further increase of capillary pressure, the pore water will be dispelled in the entire specimen resulting in notable desaturation as shown in Fig. 13. Due to the adoption of dry gas in the test and the capillary effect on gas migration process, the specimen shrinks (Fig. 14) with altering of the hydraulic property of the porous medium, which results in gas flow along the interface between the specimen and the rigid testing cell (Fig. 15 (d)) at an unexpectedly high and disordered gas flow rate.

For specimens with higher dry densities (Fig. 15 (e)), which own larger capillary sealing capacity, gas can only flow through the interconnected largest pores in the specimen (Fig. 15 (f, g)). In the meantime, it is hard for the increasingly applied gas injection pressure to dilate the localized flow pathways and compress other pore space due to the narrow channels and restricted boundary conditions. Therefore, gas is mainly controlled by the discrete capillary effect (Fig. 11) and the desaturation of the specimen decreases with increasing dry density (Fig. 13). Similarly, unexpectedly high and disordered gas flow rate will be observed due to the shrinkage of the specimen induced by the dry gas and capillary effect (Fig. 15 (h)). In addition, owing to the localized distribution of capillary flow pathways, the shrinkage is slighter for specimens with higher dry densities (Fig. 14).

**Relationships between gas breakthrough pressure and soil properties**

Gas breakthrough is highly relevant to the stabilization of the repository structure and the possible leakage of radionuclides to the biosphere. Evaluation of the corresponding pressures is important in safety assessment of a geological repository. However, gas breakthrough pressure is extraordinarily difficult to be measured in...
laboratory due to the extremely time-consuming of the test (Busch et al. 2013). Therefore, predictions of gas breakthrough pressure using other properties of the soil tested are widely investigated.

Intrinsic permeability is strongly related to the soil properties (e.g., pore size, tortuosity of flow pathways, etc.) and is widely used to predict the gas breakthrough pressure due to its easy accessibility. According to Hildenbrand et al. (2002) and Busch et al. (2013), linear relationships can be identified between the gas breakthrough pressure and the intrinsic permeability in a log-log space. In the present work, the intrinsic permeability measured on compacted GMZ bentonite specimens with different dry densities are listed in Table 5.

The relationship between the gas breakthrough pressure and intrinsic permeability can be fitted in Eq. (3), where \( P_{b} \) denotes the gas breakthrough pressure and \( k_{in} \) denotes the intrinsic permeability. This is consistent with the general trend given by literatures (Hildenbrand et al. 2002; and Busch et al. 2013).

\[
\log_{10}(P_{b}) = -0.7272 \times \log_{10}(k_{in}) - 13.58 \quad R^2=0.9759
\]  

(3)

In the meantime, based on the conclusions obtained in the present work, a new model with consideration of both mechanical effect (denoted by the swelling pressure, \( P_{swell} \)) and capillary effect (denoted by the calculated gas entry pressure, \( P_{entry} \)) also can be given in Eq. (4).

\[
P_{b} = a \cdot P_{swell} + (1 - a) \cdot P_{entry}
\]  

(4)

Here, parameter \( a \) is a weight coefficient that depends on the dry density of the tested specimen. According to the test results on specimens with dry density of 1.3, 1.5, 1.7 Mg/m\(^3\) (Fig. 11), the relationship between parameter \( a \) and dry density can be presented in Fig. 16. Therefore, Eq. (4) can be rearranged as Eq. (5).
\[ P_b = 1.3679 \cdot (\rho_d)^{-3.975} \cdot P_{swell} + \{1 - 1.3679 \cdot (\rho_d)^{-3.975}\} \cdot P_{entry} \quad (5) \]

Using the swelling pressure/the water retention curves obtained (Figs. 8 and 9) and Eq. (5), the gas breakthrough pressures for GMZ bentonite specimens with dry densities of 1.4 and 1.6 Mg/m\(^3\) are predicted to be 2.18 MPa and 5.34 MPa, respectively, which are approximately to the measured values 2.16 MPa and 5.29 MPa correspondingly. This confirms the validation of the model proposed in this work.

Conclusions

For investigation of the gas breakthrough mechanism in saturated compacted GMZ bentonite, a series of gas injection, swelling pressure, water permeability and water retention tests were conducted on specimens under rigid boundary conditions.

Gas breakthrough phenomena, where gas flow rate increased sharply accompanied by declining gas injection pressure, were universally confirmed for saturated compacted GMZ bentonite specimens with different dry densities.

For saturated GMZ bentonite tested under rigid boundary conditions, gas breakthrough pressure was significantly larger than the swelling pressure and slightly lower than the calculated gas entry pressure. This indicates that gas flow is controlled by both dilatancy and capillary processes. For lower dry densities, the capillary and mechanical effects were both important to gas migration process, while at the higher dry density range, the capillary effect became more significant.

Due to the desaturation and shrinkage of the specimen caused by the adoption of dry gas and the capillary effect on gas migration process, gas can flow along the interface between the specimen and the inner wall of the rigid testing cell resulting in an unexpectedly high and disordered gas flow rate. With the increase of dry density,
gas flow pathways will be concentrated in interconnected larger pores, then result in
discrete capillary gas flow and slighter desaturation/shrinkage of the specimen.

A linear relationship between the gas breakthrough pressures and the intrinsic
permeability on a log-log space can be confirmed for compacted GMZ bentonite
tested under rigid boundary conditions. In addition, a new model with consideration
of both mechanical effect and capillary effect was proposed and verified for
prediction of gas breakthrough pressure accurately using the swelling pressure and the
calculated gas entry pressure.

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Figure captions

Fig. 1 A trail of aggregated gold nanoparticles trapped within the traced flow pathway (Harrington et al. 2012)

Fig. 2 Conceptual sketch of capillary flow process in low permeability materials (after Busch et al. 2013; Hildenbrand et al. 2013)

Fig. 3 Preferential gas flow pathways along the interface (Liu et al. 2015)

Fig. 4 Setup for swelling pressure and water permeability tests

Fig. 5 Setup for gas injection test, (a) the assembled view; (b) the embed stainless steel porous disc

Fig. 6 Setup for water retention tests (Ye et al. 2012)

Fig. 7 Evolutions of gas flow rate with time

Fig. 8 Evolutions of swelling pressure of compacted GMZ bentonite specimens

Fig. 9 Water retention curves for compacted GMZ bentonite specimens

Fig. 10 Evolution of calculated gas entry pressure with dry density for compacted GMZ bentonite specimens

Fig. 11 Comparisons among the gas breakthrough pressure, the swelling pressure and the calculated gas entry pressure

Fig. 12 Diagrammatic sketch of interfacial gas flow upon drying and shrinkage of the specimen

Fig. 13 Reduction magnitude of water saturation before/after gas injection test for compacted GMZ bentonite specimen

Fig. 14 The specimen diameter measured before and after gas injection test

Fig. 15 Gas breakthrough mechanism in saturated GMZ bentonite specimen under rigid boundary conditions

Fig. 16 Relationship between parameter a and dry density
Fig. 1 A trail of aggregated gold nanoparticles trapped within the traced flow pathway

(Harrington et al. 2012)
Fig. 2 Conceptual sketch of capillary flow process in low permeability materials (after Busch et al. 2013; Hildenbrand et al. 2013)
Fig. 3 Preferential gas flow pathways along the interface (Liu et al. 2015)
Fig. 4 Setup for swelling pressure and water permeability tests
Fig. 5 Setup for gas injection test, (a) the assembled view; (b) the embed stainless steel porous disc
(a) Setup for the osmotic technique

(b) Setup for the vapor phase technique

Fig. 6 Setup for water retention tests (Ye et al. 2012)
(a) Dry density 1.3 Mg/m$^3$

(b) Dry density 1.4 Mg/m$^3$

(c) Dry density 1.5 Mg/m$^3$
(d) Dry density 1.6 Mg/m$^3$

(e) Dry density 1.7 Mg/m$^3$

Fig. 7 Evolutions of gas flow rate with time
Fig. 8 Evolutions of swelling pressure of compacted GMZ bentonite specimens
Fig. 9 Water retention curves for compacted GMZ bentonite specimens
Fig. 10 Evolution of calculated gas entry pressure with dry density for compacted GMZ bentonite specimens

\[ P_{\text{entry}} = 0.6364 \cdot \rho_d^{4.7824} \]

\[ R^2 = 0.9984 \]
Fig. 11 Comparisons among the gas breakthrough pressure, the swelling pressure and the calculated gas entry pressure.
Fig. 12 Diagrammatic sketch of interfacial gas flow upon drying and shrinkage of the specimen
Fig. 13 Reduction magnitude of water saturation before/after gas injection test for compacted GMZ bentonite specimen
Fig. 14 The specimen diameter measured before and after gas injection test
Fig. 15 Gas breakthrough mechanism in saturated GMZ bentonite specimen under rigid boundary conditions
Fig. 16 Relationship between parameter $a$ and dry density

$$a = 1.3679 \times (\rho_d)^{-0.875} \quad R^2 = 0.9744$$
<table>
<thead>
<tr>
<th>Property</th>
<th>Description</th>
</tr>
</thead>
<tbody>
<tr>
<td>Specific gravity of soil grain</td>
<td>2.66</td>
</tr>
<tr>
<td>pH</td>
<td>8.68-9.86</td>
</tr>
<tr>
<td>Liquid limit (%)</td>
<td>276</td>
</tr>
<tr>
<td>Plastic limit (%)</td>
<td>37</td>
</tr>
<tr>
<td>Total specific surface area (m²/g)</td>
<td>597</td>
</tr>
<tr>
<td>Cation exchange capacity (mmol/100g)</td>
<td>77.3</td>
</tr>
<tr>
<td>Main exchanged cation (mmol/100g)</td>
<td>Na⁺ (43.36), Ca²⁺ (29.14), Mg²⁺ (12.33), K⁺ (2.51)</td>
</tr>
<tr>
<td>Montmorillonite (75.4%)</td>
<td></td>
</tr>
<tr>
<td>Quartz (11.7%)</td>
<td></td>
</tr>
<tr>
<td>Fekdspar (4.3%)</td>
<td></td>
</tr>
<tr>
<td>Cristobalite (7.3%)</td>
<td></td>
</tr>
</tbody>
</table>
Table 2 The brix and concentration of the PEG solution and corresponding suction

<table>
<thead>
<tr>
<th>Suction (MPa)</th>
<th>Concentration of PEG 20000 (%)</th>
<th>Brix index</th>
</tr>
</thead>
<tbody>
<tr>
<td>0.01</td>
<td>3.015</td>
<td>2.634</td>
</tr>
<tr>
<td>0.1</td>
<td>9.535</td>
<td>7.834</td>
</tr>
</tbody>
</table>
Table 3 Salt solutions and corresponding suctions (Tang and Cui 2005)

<table>
<thead>
<tr>
<th>Salt solution</th>
<th>Suction (MPa) (20°C)</th>
</tr>
</thead>
<tbody>
<tr>
<td>LiCl₂</td>
<td>309</td>
</tr>
<tr>
<td>MgCl₂</td>
<td>150</td>
</tr>
<tr>
<td>K₂CO₃</td>
<td>113</td>
</tr>
<tr>
<td>Mg(NO₃)₂</td>
<td>82</td>
</tr>
<tr>
<td>NaNO₂</td>
<td>57</td>
</tr>
<tr>
<td>NaNO₃</td>
<td>39</td>
</tr>
<tr>
<td>NaCl</td>
<td>38</td>
</tr>
<tr>
<td>(NH₄)₂SO₄</td>
<td>24.9</td>
</tr>
<tr>
<td>KCl</td>
<td>21</td>
</tr>
<tr>
<td>ZnSO₄</td>
<td>12.6</td>
</tr>
<tr>
<td>KNO₃</td>
<td>9</td>
</tr>
<tr>
<td>K₂SO₄</td>
<td>4.2</td>
</tr>
</tbody>
</table>
Table 4 Parameters in van Genuchten model for specimens with different dry densities

<table>
<thead>
<tr>
<th>Dry density (Mg/m$^3$)</th>
<th>1.3</th>
<th>1.5</th>
<th>1.7</th>
<th>1.9</th>
</tr>
</thead>
<tbody>
<tr>
<td>Calculated gas entry pressure $P_{entry}$ (MPa)</td>
<td>2.284</td>
<td>4.229</td>
<td>8.192</td>
<td>13.77</td>
</tr>
<tr>
<td>Shape factor of van Genuchten model $m$</td>
<td>1.298</td>
<td>1.263</td>
<td>1.298</td>
<td>1.322</td>
</tr>
<tr>
<td>Water residual saturation $S_{wr}$ (%)</td>
<td>0</td>
<td></td>
<td></td>
<td></td>
</tr>
<tr>
<td>Gas residual saturation $S_{gr}$ (%)</td>
<td>0</td>
<td></td>
<td></td>
<td></td>
</tr>
<tr>
<td>Goodness of fit $R^2$</td>
<td>0.9929</td>
<td>0.9880</td>
<td>0.9858</td>
<td>0.9766</td>
</tr>
</tbody>
</table>
Table 5 Intrinsic permeability of GMZ bentonite

<table>
<thead>
<tr>
<th>Dry density (Mg/m³)</th>
<th>1.3</th>
<th>1.4</th>
<th>1.5</th>
<th>1.6</th>
<th>1.7</th>
</tr>
</thead>
<tbody>
<tr>
<td>Intrinsic permeability (m²)</td>
<td>1.37e-19</td>
<td>8.82e-20</td>
<td>3.87e-20</td>
<td>2.38e-20</td>
<td>1.14e-20</td>
</tr>
</tbody>
</table>