Swelling deformation of compacted bentonite considering the effects of Na cations and OH anions

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Abstract. Compacted bentonite has become a viable candidate as suitable artificial barrier material for restricting the movement of high-level radionuclides released from the canister in the deep geological repository. However, this material itself is fragile mainly because it is sensitive to the complex chemical environment especially alkaline components. Based on the interaction mechanism of cations and anions, this paper is mainly focused on the analysis of swelling deformation characteristics of compacted bentonite. Laboratory test results are shown that the elevated Na cations concentration can inhibit both the primary and secondary expansion process in compacted bentonite. The elevated OH anions concentration can inhibit the primary expansion process of compacted bentonite and will promote the secondary expansion process. The effects of NaOH solution with different concentrations on the swelling strain present the interaction of Na cations and OH anions. The swelling deformation of compacted bentonite is not only affected by the concentration of solution but also may be related to ionic compositions.

1. Introduction
Despite these contemporary issues of the greenhouse effect and non-regeneration energy have relieved effectively, about how to deal with spent nuclear fuel is still a fundamental issue to solve during the utilization of atomic energy. Up to now, deep geological disposal system, especially the multiple barrier disposal system, as the most feasible disposal program is used for high-level radioactive waste storage. Because of its low hydraulic conductivity, high swelling potential, favourable radionuclide retardation capacity, good retention performance, self-sealing characteristics and adequate mechanical resistance (Castellanos et al., 2008; Siddiqua et al., 2011; Zhu et al., 2013; Ye et al., 2014), compacted bentonite has been considered as an alternative artificial barrier buffer material for the isolation of high-level radioactive waste in many counties (Komine and Ogata, 1996; Komine, 2004; Imbert and Villar, 2006; Lloret and Villar, 2007; Tang et al., 2008). At present, bentonites that have been studied abroad include MX-80 sodium bentonite from Wyoming, USA, IBECO bentonite from Sweden, Kunigel V1 sodium bentonite from Japan, FoCa7 calcium bentonite from France, Calcigel calcium bentonite from Germany, OPHELE artificial prefabricated bentonite from Belgium, and Cortijo de Archidona bentonite from Spain (Komine and Ogata, 1996; Lemaire et al., 2004; Komine et al., 2009). In fact, calcium bentonite has a slightly higher shear strength than sodium bentonite (Gleason et al., 1997). However, considering engineering barrier properties of bentonite, sodium bentonite is more
suitable than calcium bentonite as a buffer/backfill material for the high-level radioactive waste disposal repository.

A great dosage of concrete or cementitious material is almost 900,000 kg, which is often used as a structure, confining, backfill material or engineering barrier in the deep geological disposal of radioactive waste (Karland et al., 2007; Sun et al., 2018). In-situ groundwater with various chemical compositions would be altered and generates a high-pH alkaline solution due to the degradation of concrete or cementitious material over time. Fernandez et al. (2009) confirmed that alkaline fluids will be produced and the pH value of these fluids emanating from ordinary Portland cement (OPC) concrete could be 13-14. The main composition of early leaching yields Young Cement Water (YCW) is dominated by K+ (0.2-0.5M), Na+ (0.05-0.2M) and OH- (0.3-0.7M), and Evolved Cement Water (ECW) is characterized by Ca-OH (Faucon et al., 1998; Savage et al., 2002; Fernandez et al., 2009, 2010). These alkaline components infiltrated through the compacted bentonite and interact with montmorillonite in bentonite causing its dissolution which will weaken the buffer properties of bentonite (Baldi et al., 1988; Musso et al., 2003; Romero et al., 2005; Rao et al., 2006; Lloret and Villar, 2007; Zhu et al., 2015). Thus, it is meaningful to evaluate the reliability and long-term performance of bentonite by investigating the effect of chemical compositions on the swelling behaviour of compacted bentonite, especially based on cations and anions.

Many previous investigations mainly focus on determining the effect of saline or alkaline cement pore water on the bentonite mineralogy alteration (Ramirez et al., 2002; Fernandez et al., 2010; Chen et al., 2016). The reaction process between bentonite and hyperalkaline fluids begins with the cation exchange in the short time and dissolution-precipitation of minerals in the long period (Savage et al., 2002; Deneele et al., 2010). The pH value of the pore water can influence the cation exchange process at the interlayers of montmorillonite and the pore structure of bentonite. Only cation exchange process occurs at low pH, while both the cation exchange and surface complexation reactions may take place during the same time period as the pH value of pore water increases (Karland et al., 2007). Numerous studies have demonstrated that the swelling properties of compacted bentonite are seriously weakened because of saline or alkaline cement pore water infiltrating.

Generally, the swelling behaviour of bentonite decreases as the solution salinity or concentration increases (Castellanos et al., 2008; Komine et al., 2009; Siddiqua et al., 2011). Rao and Thyagaraj (2007) showed that the inflow of NaCl solution reduced the swelling magnitude and pressure and even caused the compacted clay specimens to experience compressive strains. Rao et al. (2006) pointed that the salinity difference between soil pore water and infiltration solution induces an osmotic suction gradient when water saturated bentonite was exposed to salt solution, which can be dissipated by the inward diffusion of ions and the outflow of soil pore water. This phenomenon is called osmotically induced consolidation, that is, the inward diffusion of cation decreases the thickness of diffusion double layer (DDL) and thus restricts the swelling capacity; and meanwhile, the outflow of soil pore water induces negative porewater pressure, which increases the effective stress and consequently compresses the soil (Chen et al., 2017).

The influence of the permeant alkaline solution on the swelling behaviour of bentonite or its mixture was also highlighted by numerous researchers (Herbert et al., 2008; Komine et al., 2009; Lee et al., 2012; Ye et al., 2014; Anh et al., 2017). To investigate the effects of alkaline solutions on the long-term swelling behaviour, it is necessary in order to study the mineralogy evolution of bentonite under the geochemical conditions (Bauer and Velde, 1999; Fernandez et al., 2013). Fernandez et al. (2009) immersed FEBEX bentonite into the synthetic alkaline solutions (K-Na-OH and Ca-(OH)2) with a pH of 8-8.5 for 60 days, and found that some new minerals, such as brucite and magnesium-rich clay generated. Ramirez et al. (2002) studied mineral composition changes of bentonite under the permeation of alkaline solution found that some zeolite crystallization was detected and much more Mg2+ was found in non-exchangeable form in the smectite mineral. Savage and Benbow (2007) investigated the cement/bentonite interface behaviour and found that the clay minerals were dissolved and the calcium (aluminium) silicate hydrates (C-(A)-S-H) and zeolites were the most likely byproduct minerals, and that feldspars, hydroxides, carbonates, polymorphs of silica and some sheet
silicates may also precipitate. The formation of secondary minerals as a result of the alkaline alteration of bentonite will change the pore size distribution (PSD) of bentonite and lead to the change the swelling behaviour, which is in agreement with the observation by Cuisinier et al. (2008) and Sarkar and Siddiqua (2016).

Thanks to its favorable physical properties and large volume in deposit, a local sodium bentonite named Gaomiaozi (GMZ) has been identified as a potential buffer/backfill material for the Chinese HLW disposal (Liu et al., 2001; Ye et al., 2010). Contributions have been received more and more attentions for these investigations relating to the effects of saline (NaCl, CaCl2 and KCl, etc.) or pH (NaOH and KOH, etc.) on swelling properties of GMZ bentonite (Ye et al., 2009; Cui et al., 2012; Zhu et al., 2013; Zhang et al., 2014; Sun et al., 2018). The swelling potential present a decrease tendency as the concentration of NaOH, KOH or cement solutions increases. But examination of the aforementioned studies shows that the effect of swelling capacity of bentonite by alkaline solutions, especially based on cations and anions, is scarce.

Based on this, the swelling behaviour of compacted bentonite specimens considering the effects of Na cations and OH anions will be investigated. In the present work, compacted GMZ bentonite with an initial dry density of 1.70 Mg/m³ was performed on one-dimensional swelling deformation test. During the test, a vertical stress of 0.1 MPa was applied along with the penetration of NaCl and NaOH solutions. The study will discuss the NaOH concentrations and ionic compositions effect on swelling strain of compacted bentonite. It is divided into threefold, that is, firstly, to study the effect of NaOH concentration on swelling strain; secondly, to study the effect of Na cations on swelling strain; thirdly, to study the effect of OH anions on swelling strain.

2. Materials and methods

2.1 Materials

GMZ bentonite deposited from Gaomiaozi in the Inner Mongolia Autonomous Region, 300 km northwest from Beijing, China, will be used as a barrier or backfill material of China’s nuclear waste repository. It presents a grain size of no more than 200 μm. Its basic properties of the GMZ bentonite are listed in Table 1.

Table 1. Basic properties of the GMZ bentonite (Wen, 2006; Ye et al., 2014).

| Property                              | Description       |
|---------------------------------------|-------------------|
| Main minerals                         | Montmorillonite: 75.4%  |
|                                       | Quartz: 11.7%     |
|                                       | Cristobalite: 7.3% |
|                                       | Feldspar: 4.3%    |
| Main exchanged cation (cmol/kg)       | Na⁺: 43.36        |
|                                       | 1/2Ca²⁺: 14.57    |
|                                       | 1/2Mg²⁺: 6.17     |
|                                       | K⁺: 2.51          |
| Cation exchange capacity (cmol/kg)    | 77.30             |
| Liquid limit (%)                      | 276               |
| Plastic limit (%)                     | 37                |

The chemical reagents of NaCl and NaOH used throughout this laboratory test are of analytical grade, corresponding to the purity of more than 99%, respectively. The conductivity of distilled water used in this test was EC ≤ 4 um/cm, corresponding to a pH of 6.5. Saline solutions were configured by
using NaCl reagent and de-ionized water, and alkaline solutions were configured by using NaOH reagent and de-ionized water.

2.2 Specimens preparation
A given quantity of GMZ bentonite power with an initial water content of 11.92% was first compacted in a stainless-steel ring mold to obtain the cylindrical specimen of 50 mm diameters and 10 mm height at a target dry density of 1.70 Mg/m$^3$. Compaction was done at a constant speed of 0.1 mm/min according to the displacement-controlled compaction method described by Ye et al. (2009). It should be noted that the full height of the specimens was achieved in three layers to maintain a uniform compacted dry density. The applied load and displacements of the piston were recorded by a load cell and a digital gage, respectively. In order to achieve homogenization of the compacted specimens, the current load was maintained for 1h to minimize the possible rebound during the follow-up unloading once the target displacement was reached. Then, the vertical load was released and the compacted specimen was pushed out from the compaction mold at a rate of 0.2 mm/min. The compaction process of the bentonite specimen is shown in Figure 1.

2.3 Swelling strain test
The specimen placed in the stainless-steel ring will be covered with a piece of oven-dried filter paper and porous stone at each end, respectively; meanwhile, these sandwiched specimens should be immediately placed in the rigid specimen ring (3 in Figure 2) for the swelling capacity test. The constant vertical stress was set to 0.1 MPa through the entire test process. Compacted bentonite specimens were infiltrated with de-ionized water, NaCl solution or NaOH solution. The one-dimensional vertical swelling deformation occurred when absorbing de-ionized water or alkaline NaOH solutions. Consequently, the vertical deformation was recorded by the dial indicator. The test was considered to be completed when the vertical deformation recorded was stable for 72 h.

3. Definition of swelling deformation parameters
In this study, the swelling strain was calculated as follows, that is

$$\varepsilon = \frac{\Delta H}{H_0} \times 100\% \quad (1)$$

**Figure 1.** The compaction process of the bentonite specimen.

**Figure 2.** Schematic one-dimensional vertical swelling deformation apparatus (1: Dial indicator; 2: Flange; 3: Ring; 4: Piston; 5: Specimen; 6: Porous stone; 7: Weights; 8: Reagent bottle; 9: Peristaltic pump; 10: solutions).
where ε was defined as the swelling strain; ΔH was defined as the deformation over the current infiltration step, mm; and H0 was defined as the initial height of bentonite specimen, mm.

Sridharan and Gurtug (2004) and Rao et al. (2007) divided the swelling dynamics process into three stages, namely initial swell, primary swell and secondary swell. Coefficients were often used to denote the time change rate of primary swell or secondary swell evolution process (Rao et al., 2007). The calculation formulas of the coefficients of the primary and secondary swell were expressed as (2) and (3), respectively, namely

\[
C_{ps} = \frac{\Delta H_p / H_0}{\Delta \log_{10} t_p}
\]

\[
C_{ss} = \frac{\Delta H_s / H_0}{\Delta \log_{10} t_s}
\]

Where ΔH_p and ΔH_s were the primary swelling strain and the secondary swelling strain, respectively, mm; in addition, Δlog_{10} t_p = log_{10} t_{p2} - log_{10} t_{p1}, Δlog_{10} t_s = log_{10} t_{s2} - log_{10} t_{s1}. t_{p1} and t_{p2} were the time at which the process of primary swell began and ended in turn, h; t_{s1} and t_{s2} were the time at which the process of secondary swell began and ended in turn, h.

4. Results and discussion

4.1 Effect of NaOH concentration on swelling strain

The evolutionary process of the swelling strain was divided into three stages, namely the initial swell, the primary swell and the secondary swell (Sridharan et al., 2004; Rao and Thyagaraj, 2007). Generally speaking, the swelling process of bentonite after absorbing water or electrolyte solution mainly depends on the two mechanisms of crystal layer expansion and double layer expansion (Madsen et al., 1989; Savage, 2002). Crystal layer expansion and double layer expansion control the primary and secondary swelling strain respectively. Schematic diagram of evolutionary mechanism of the swelling strain was shown in Figure 3. Stage I - the adsorbed water occupied the interlayer space between smectite layers and made bound water membrane thickness thicker, which caused the thickness of quasicrystals to increase; thicker quasicrystals split into several thinner quasicrystals with fewer layers and the micropores in the aggregates are filled. Stage II - under the one-dimensional vertical deformation condition, thinner quasicrystals further expansion, split-up and then the aggregates expansion. The aggregates expansion will cause the disintegration of all aggregates so that soil skeleton collapsed. Stage III - the double layer expansion began with the end of the crystal layer expansion once bound water membrane thickness of the exchangeable cations reached the maximum; the diffusion double layer repulsion would make the quasicrystals expansion again to split into several single crystal layers filled with macropores. The swelling deformation was stable at this stage with the end of the double layer expansion.

![Figure 3. Schematic diagram of swelling deformation mechanism.](image)

Deformation curves were presented in Figure 4 with the infiltration of de-ionized water or
different concentrations of NaOH alkaline solutions. It can be observed that the swelling strain of compacted bentonite was weakened along with increasing NaOH solution concentration. The greater the concentration of NaOH solution was, the shorter time was required for the swelling strain to stabilize. Furthermore, the difference between swelling strains was significantly reduced as the concentration of NaOH solution increases.

Figure 5 showed the calculation results of coefficients of the primary and secondary swell. The coefficient of primary swell decreased with NaOH alkaline solution concentration. Whereas the coefficient of secondary swell showed a general increasing tendency. The evolutionary process of the coefficient of primary swell and the total swelling strain were consistent, which indicated the dominance throughout the evolutionary process of the swelling strain. What’s more, the coefficient of secondary swell increasing suggests that the swelling deformation of compacted bentonite is not only affected by the concentration of solution but also may be related to ionic compositions.

4.2 Effect of Na cations on swelling strain

Figure 6 presents the effect of Na cations concentration on the swelling strain of compacted bentonite. The primary swelling strain and the secondary swelling strain show a decrease tendency as Na cations concentration increasing. Compared with de-ionized water, the primary swelling strain and the secondary swelling strain of compacted bentonite decrease by 29.45% and 4.83% for the infiltration of 0.1 M Na cations, respectively. When the Na cations concentration increase from 0.1 M to 1.0 M, the reductions of the primary and secondary swelling strain of compacted bentonite are 23.39% and 13.28%, respectively. It suggests that Na cations can inhibit both the primary and secondary expansion processes in compacted bentonite.

For the infiltration of Na cations, the high Na cations concentration difference between montmorillonite often affects the adsorption of water molecules (Malusis and Shackelford, 2002; Castellanos et al., 2008; Lee et al., 2012; Bohnhoff et al., 2013). The elevated Na cations concentration makes the suction potential of water molecules outside the montmorillonite crystal layer higher than that inside the montmorillonite layer. Water molecules are transferred from the internal crystal layer to the external crystal layer in order to shrink down the ionic concentration gradient of inside and outside of the crystal layer. Thus, the absorption of water molecules inside the montmorillonite layer is inhibited, which leads to the decrease of the expansion ability of the crystal layer and the inhibition of the primary swell process.

The inhibition effect of elevated Na cations concentration on the secondary swelling strain is related to the compression of the diffusion double layer (DDL) structure (Xu et al., 2003; Zhu et al., 2013, 2015). Tripathy et al. (2004) showed that the thickness of the diffusion double layer (DDL) was
inversely proportional to the square root of hydration cations (Na\(^+\)) concentrations. The thickness of the diffusion double layer decreases with the infiltration of elevated Na cations concentration. The elevated Na cations concentration inhibits the expansion of the diffusion double layer and causes it to compress (Shirazi et al., 2011). The reduction in the repulsion force between the bentonite particles occurs and the secondary swell process of compacted bentonite is weakened. The calculation formulas for the diffusion double layer thickness and ion valence and ion concentration are presented in Tripathy et al. (2004).

The calculation results of coefficients of the primary and secondary swell are shown in Table 2. Compared with the infiltration of de-ionized water, the primary swelling coefficient decreases by 22.21% and 29.47% when bentonite specimens are infiltrated with 0.1 M and 1.0 M Na cations, respectively. However, the change in the coefficient of secondary swell is not fixed, that is, the coefficient of secondary swell increases by 37.86% for 0.1 M Na cations infiltration, and the coefficient of secondary swell decreases by 10.31% for 1.0 M Na cations infiltration. When the concentration of Na cations increases from 0.1 M to 1.0 M, the coefficients of primary and secondary swell decrease by 9.33% and 34.94%, respectively.

With the increase of Na cations concentration, the coefficient of primary swell shows a tendency of decreasing, while the secondary swelling coefficient shows a trend of first increasing and then decreasing. Considering the evolutionary tendency of swelling strain of compacted bentonite, it is indicated that the primary swell rather than the secondary swell is controlled by Na cations. Even so, the analysis of the coefficients of the primary and secondary swell still shows that Na cations dominate the primary expansion process of compacted bentonite.

| Specimen labeled | Infiltration solution | Concentration | \(C_p\) | \(C_s\) |
|------------------|----------------------|--------------|--------|--------|
| L1               | De-ionized water     | 0            | 14.255 | 4.105  |
| L2               | NaCl                 | 0.1          | 11.089 | 5.660  |
| L3               | NaCl                 | 1.0          | 10.054 | 3.682  |
| L4               | NaOH                 | 0.1          | 10.630 | 5.425  |
| L5               | NaOH                 | 1.0          | 5.742  | 9.064  |

**Table 2.** Partial coefficients of primary and secondary swell.

**4.3 Effect of OH anions on swelling strain**

The effect of OH anions concentration on the swelling strain of compacted bentonite is showed in Figure 7. When the concentrations of OH anions increase from 0.1 M to 1.0 M, the primary swelling strain decreases by 9.71% and the secondary swelling strain increases by 7.55%. The primary swelling
strain that 0.1 M and 1.0 M OH anions can restrain is 9.66% and 54.89%, and the promotion rate of 0.1 M and 1.0 M OH anion to secondary swelling strain is 57.42% and 406.31%, respectively. It indicates that OH anions will inhibit the primary expansion process of compacted bentonite and will promote the secondary expansion process.

For the infiltration of OH anions, the elevated concentration of OH anions can promote the dissolution of montmorillonite and the increase of the tetrahedral layer charge (Malla and Douglas, 1987; Laird, 2006), which increases the stability of the quasicrystals to moisture or solution absorption to a certain extent (Laird, 2006). The swelling capacity of the crystal layer is also reduced as the stability of the quasicrystals increases. The electric potential will decrease with the increase of total negative charge between the quasicrystals, which will promote secondary swell process.

With the infiltration of OH anions from 0.1 M and 1.0 M, the coefficient of primary swell decreases by 3.85, and the coefficient of secondary swell increases by 5.62. In other words, the concentration of OH anions can weaken the coefficient of primary swell and can promote the coefficient of secondary swell. Considering the evolutionary tendency of swelling strains of compacted bentonite, it can be seen that the primary and secondary expansion processes are all controlled by OH anions concentration. The effects of NaOH solution with different concentrations on the swelling strain present the interaction of Na cations and OH anions.

Generally speaking, the swelling strain of compacted bentonite shows a decrease tendency as NaOH concentration increasing. Meanwhile, the effects of NaOH solution with different concentrations on the swelling strain present the interaction of Na cations and OH anions. The swelling deformation of compacted bentonite is not only affected by the concentration of solution but also may be related to ionic compositions. The dissolution of montmorillonite occurs during the infiltration of alkaline solutions. It means that the high swelling properties of compacted bentonite cannot be regenerated even after de-salination or de-alkalization process.

5. Conclusion
In this study, a series of swelling strain tests were performed on compacted GMZ bentonite with a dry density of 1.70 Mg/m³. Three types solutions of de-ionized water, NaCl solution and NaOH solution were used to investigate swelling deformation characteristics of compacted bentonite considering the effects of cations and anions. The main observations of this study are shown as following:

1) The swelling strain of compacted bentonite was weakened along with increasing NaOH solution concentration. The greater the concentration of NaOH solution was, the shorter time was required for the swelling strain to stabilize.

2) The elevated Na cations concentration can inhibit both the primary and secondary expansion process of compacted bentonite, which are related to the osmotic suction gradient and the compression of the diffusion double layer (DDL), respectively.

3) The elevated OH anions concentration will inhibit the primary expansion of compacted bentonite and promote the secondary expansion process, which are related to the dissolution of montmorillonite.

4) The swelling strain of compacted bentonite shows a decrease tendency as NaOH concentration increasing. Meanwhile, the effects of NaOH solution with different concentrations on the swelling strain present the interaction of Na cations and OH anions. The swelling deformation of compacted bentonite is not only affected by the concentration of solution but also may be related to ionic compositions.

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