On the chemo-thermo-hydro-mechanical behavior of geological and engineered barriers

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Abstract: An overview of the recent findings about the chemo-hydro-mechanical behaviour of materials used for both geological and engineered barriers in nuclear waste disposal is presented, through some examples about natural Boom Clay (BC) and compacted bentonite-based materials. For the natural BC, it was found that compression index identified from both oedometer and isotropic compression tests is similar and the compressibility of BC from the Mol site is higher than that of BC from the Essen site; the shear strength of Mol BC is also higher that the Essen BC, suggesting a significant effect of carbonates content; the thermal volume change is strongly overconsolidation ratio (OCR) dependent – low OCR values promote thermal contraction while high OCR values favour thermal dilation; the volume change behaviour is also strongly time dependent and this time dependent behaviour is governed by the stress level and temperature; the effect of pore-water salinity on the volume change behaviour can be significant when the smectite content is relatively high. For the bentonite-based materials, it was found that thermal contraction also occurs at low OCR values, but this is suction dependent – suction promotes thermal dilation. Under constant volume conditions, wetting results in a decrease of hydraulic conductivity, followed by an increase. This is found to be related to changes in macro-pores size – wetting induces a decrease of macro-pores size followed by an increase due to the aggregates fissuring. The presence of technological voids can increase the hydraulic conductivity but does not influence the swelling pressure.

Key words: Boom Clay (BC); bentonite-based materials; mechanical behavior; hydraulic conductivity; pore-water salinity; technological voids

1. Introduction

In the high-level radioactive waste geological disposal, multi-barrier concept with geological barrier and engineered barriers is usually considered. The geological barrier is the natural host formation as claystones (France and Switzerland), salts (Germany), granite (China and Sweden) and stiff clays (Belgium), etc. The main functions of geological barrier are its low hydraulic conductivity and its capacity of self healing. The engineered barrier is made up of compacted bentonite-based materials. It is usually used with granite geological barriers (China and Sweden) and constitutes a barrier before the geological one. Note that these materials are also used as filling materials for other geological barriers. Main functions of the bentonite-based materials are its sealing capacity related to its volume change behaviors, including its swelling properties and its retention capacity mainly related to its hydraulic behavior.

In the situation of nuclear waste disposal, the constitutive materials of both barriers are subjected to complex coupled chemo-thermo-hydro-mechanical (CTHM) loadings stemming from interaction between clay minerals and pore-water chemistry or other chemicals resulted from concrete alteration, from heat emission by the waste canisters, from water flow and from field stress/materials swelling. This justifies the wide studies conducted on their CTHM behaviors in both laboratory and field conditions.
In field, tests have been performed in different Underground Research Laboratories (URLs). For instance, the French agency (Andra) has carried out various tests in the Bure URL to investigate the involved Oxfordian claystone behavior during excavation, heating, etc. The Swiss agency (Nagra) has done the same in the Mont Terri URL. The Belgian agency (Ondraf/Euridice) has investigated the hydraulic behavior and mechanical behavior of Boom Clay (BC) in the Mol URL. Note also that the French institution IRSN (Institut de Radioprotection et de la Sûreté Nucléaire) has been conducting the infiltration tests aiming at identifying the key factors related to the long-term performance of bentonite-based sealing systems when considering the initial technological void. The thermal loading effect has been also investigated in different URLs, for instance, in the Mol URL, a 10-year heating project – Praclay project is underway, aiming at investigating the effect of heating on the hydro-mechanical of BC on one hand, and the sealing capacity of the compacted bentonite ring on the other hand.

2. Volume change behavior of Boom Clay

In the Belgian program for nuclear waste disposal, BC has been investigated for both Mol site (URL location) and Essen site (about 50 km east from Mol). In order to compare its volume change behavior for both sites, five cores (1 m long and 100 mm in diameter) were taken from the Essen site and one core was taken from the Mol site at depth of 223 m. The geotechnical properties of these cores are shown in Table 1.

For the BC at Essen, two cores were taken from the Putte member (Ess75 and Ess83) and three cores from the Terhagen member (Ess96, Ess104 and Ess112). The geotechnical properties of these cores are similar: specific gravity, \( G_s = 2.64–2.68 \); liquid limit, \( w_L = 62\%–78\% \); plastic index, \( I_p = 36–45 \). The void ratio \( (e_0) \) ranges from 0.700 to 0.785. The carbonate content of the core Ess104 (4.36\%) is significantly higher than those of other cores (lower than 1\%). Note that the carbonate content for Ess112 is also relatively high: 2.64\%.

After Francois et al. (2009), the main parameters of the BC at Mol, e.g. \( G_s, w_L \) and \( I_p \) are similar to those of the BC at Essen. The void ratio ranges from 0.49 to 0.67, significantly lower than that of BC at Essen. These differences suggest that the BC at Mol is denser and more carbonated.

Table 2 depicts the mineralogical compositions of different cores. It is observed that BC at Essen contains more active minerals such as smectite, but the total amount of smectite and interstratified illite/smectite is similar.

The particle size distribution curves are shown in Fig. 1. The curves of Ess75 and Ess96 are close to that of Mol, showing a clay content (< 2 \( \mu m \)) of 57\%–60\%. The curves of Ess75 and Ess104 are slightly below the curves of Ess83, Ess96 and Mol, showing a clay content of 43\%–50\%. The core Ess112 presents significantly larger particles and a lower clay content (about 40\%).

### Table 1. Geotechnical properties of the soil cores studied.

<table>
<thead>
<tr>
<th>Core</th>
<th>Depth (m)</th>
<th>Member</th>
<th>( G_s )</th>
<th>( w_L ) (%)</th>
<th>( I_p ) (%)</th>
<th>( e_0 )</th>
<th>Carbonate content (%)</th>
</tr>
</thead>
<tbody>
<tr>
<td>Ess75</td>
<td>218.91–219.91</td>
<td>Putte</td>
<td>2.65</td>
<td>78</td>
<td>45</td>
<td>0.785</td>
<td>0.91</td>
</tr>
<tr>
<td>Ess83</td>
<td>226.65–227.65</td>
<td>Putte</td>
<td>2.64</td>
<td>70</td>
<td>37</td>
<td>0.730</td>
<td>0.76</td>
</tr>
<tr>
<td>Ess96</td>
<td>239.62–240.62</td>
<td>Terhagen</td>
<td>2.68</td>
<td>69</td>
<td>36</td>
<td>0.715</td>
<td>0.24</td>
</tr>
<tr>
<td>Ess104</td>
<td>247.90–248.91</td>
<td>Terhagen</td>
<td>2.68</td>
<td>68</td>
<td>39</td>
<td>0.700</td>
<td>4.36</td>
</tr>
<tr>
<td>Ess112</td>
<td>255.92–256.93</td>
<td>Terhagen</td>
<td>2.67</td>
<td>62</td>
<td>37</td>
<td>0.755</td>
<td>2.64</td>
</tr>
<tr>
<td>Mol</td>
<td>223</td>
<td>Putte</td>
<td>2.67</td>
<td>59.83</td>
<td>0.49–0.67</td>
<td>5.9–8.3</td>
<td></td>
</tr>
</tbody>
</table>

![Particle size distribution curves](image-url)
Both oedometer and isotropic compression tests were carried out, allowing determination of the compression index \( C_c \). The obtained values of \( C_c \) are shown in Table 3. It appears clearly that both tests give similar results. Comparison between the two sites shows that BC at Mol is more compressible than BC at Essen.

3. Shear strength of Boom Clay

In Fig. 2, the results at failure of BC at Mol by various authors (Horseman et al., 1987; Sultan, 1997; Van Impe, 1993; Baldi et al., 1991; Coll, 2005) and the results on BC at Essen are gathered in \( p' - q \) plane (mean stress–deviator stress plane) for a direct comparison. It appears clearly that the slope of intact BC at Mol in the range of \( p' > 2 \) MPa and the slope of intact BC at Essen is very close (0.46 against 0.47), suggesting a similar internal friction angle (12°–13°). However, the \( q_0 \) value (intersection) for Mol is significantly larger than that for Essen (1.12 MPa against 0.39 MPa). This corresponds to a higher effective cohesion for Mol as compared with that for Essen: 0.53 MPa against 0.19 MPa. This difference in cohesion is likely due to the difference in carbonate contents: BC at Mol has a higher carbonate content (see Table 1).

The reconstituted Essen BC has an effective cohesion \( c' = 0.01 \) MPa and an internal friction angle \( \phi' = 20^\circ \). These values are close to those of Essen core Ess112.

4. Discussion

The volume change behavior of compacted MX80 bentonite was investigated using an isotropic cell that enables suction and temperature to be controlled (Tang et al., 2007; Tang and Cui, 2010a). The results of thermal volume change under a pressure \( p = 0.1 \) MPa are presented in Fig. 15. The results from tests T1 (suction \( s = 110 \) MPa) and T2 (\( s = 39 \) MPa) show that heating induced an expansion. Considering that this expansion is linear, a coefficient of thermal expansion \( \alpha = 2 \times 10^{-4} \) °C\(^{-1} \) can be deduced. On the contrary, the result from test T3 (\( s = 9 \) MPa) shows that heating induced a contraction. In addition, the subsequent cooling-reheating cycle undertaken shows a reversible behavior. Note that significant data scatter was observed in this test and it is thus difficult to quantify the volume change behavior during the cooling-reheating stage.

4.1. Effect of technological void on hydraulic conductivity

Compacted MX80/sand mixture was considered to study the effect of technological void on hydraulic conductivity. With a ring diameter of 38 mm, the annular technological void selected (14% of the total cell volume representing 17% of the initial sample volume) corresponds to a sample diameter of 35.13 mm. Four tests with the same technological void of 14% were conducted on samples with the same initial water content of 11% and various initial dry densities obtained by changing the compaction pressure (between 65 MPa and 85 MPa, giving rise to dry densities comprised between 1.93 Mg/m\(^3\) and 1.98 Mg/m\(^3\)).

The saturated hydraulic conductivity was determined after saturation of the samples, by both the constant head test and the indirect Casagrande’s method. Fig. 21 compares the results obtained with that obtained by Gatabin et al. (2008) by constant head test on homogeneous samples at similar densities. The data obtained for the heterogeneous samples with both methods are in good agreement. On the whole, the hydraulic conductivity decreases with density increase, following a slope comparable to that obtained by Gatabin et al. (2008). An in-depth examination shows that the samples with initial technological voids exhibit higher hydraulic conductivity than that by Gatabin et al. (2008), with a difference of one order of magnitude. This difference is suspected to be due to a preferential water flow in the looser zone (initial technological voids) around the samples.
4.2. Effect of technological void on swelling pressure

In order to analyze the effect of technological void on swelling pressure, various constitutive parameters of the compacted mixture are defined. It is supposed that in the mixture the volume of bentonite ($V_b$) in the mixture is equal to the difference between the total volume ($V_t$) and the volume of sand ($V_s$). $V_b$ is equal to the sum of the bentonite particle volume ($V_{bs}$) and the volume of void, namely intra-void volume ($V_i$). The bentonite void ratio ($e_b$) consists of two parts (Eq. (2)), the intra-bentonite void ratio inside the soil ($e_{bi}$) and the void ratio corresponding to the technological void ($e_{tech}$). Eqs. (3) and (4) define these two voids, respectively.

$$ e_b = e_{bi} + e_{tech} $$

$$ e_{bi} = \frac{V_i}{V_{bs}} $$

$$ e_{tech} = \frac{V_{tech}}{V_{bs}} $$

where $V_{tech}$ is the volume of technological voids. The value of $e_{bi}$ can be deduced from the initial dry unit mass of the mixture ($\rho_{dm}$) using the following equations:

$$ e_{bi} = \frac{G_{sb}\rho_w}{\rho_{db}} - 1 $$

$$ \rho_{db} = \frac{(B/100)\rho_s G_s \rho_w}{G_{ss}\rho_w(1 + W_{ms}/100) - \rho_m(1-B/100)} $$

where $\rho_w$ is the water unit mass, $G_{sb}$ is the specific gravity of bentonite, $\rho_{db}$ is the initial dry unit mass of bentonite in the mixture, which was calculated using Eq. (6) (Dixon et al., 1985; Lee et al., 1999; Agus and Schanz, 2008; Wang et al., 2012), $\rho_m$ is the unit mass of the mixture, $B$ (%) is the bentonite content (in dry mass) in the mixture, $G_{ss}$ is the specific gravity of sand, $W_{ms}$ is the water content of the mixture. In this study the decrease of water unit mass ($\rho_w$) during hydration (e.g. Skipper et al., 1991; Villar and Lloret, 2004) was not considered and the value was assumed to be constant (1.0 Mg/m$^3$), $B = 70\%$, $G_{ss} = 2.65$.

The values of vertical stress measured at the end of the hydration tests on samples with technological voids are presented in Fig. 22 with respect to the bentonite void ratio. The data of swelling pressure measured in homogeneous samples under the same conditions of constant volume by other authors are also plotted for comparison (MX80 70/30 bentonite/sand mixture from Karnland et al. (2008), and pure MX80 bentonite from Börgesson et al. (1996), Dixon et al. (1996), Karnland et al. (2008), and Komine et al. (2009)).

5. Conclusions

An overview of the recent findings about the CTHM behaviour of materials used for both geological and engineered barriers in nuclear waste disposal is presented, through some examples about the natural BC and the compacted bentonite-based materials.

For the natural BC, it was found that compression index identified from both oedometer and isotropic compression tests is similar and the compressibility of BC from the Mol site is higher than that of BC from the Essen site; the shear strength of Mol BC is also
higher than that of the Essen BC, suggesting a significant effect of carbonates content; the thermal volume change is strongly OCR dependent: low OCR values promote thermal contraction while high OCR values favour thermal dilation. The volume change behaviour is also strongly time dependent and this time dependent behaviour is governed by the stress level and temperature. The effect of pore-water salinity on the volume change behaviour can be significant when the smectite content is relatively high. However, the pore-water chemistry does not seem to affect the hydraulic conductivity.

For the bentonite-based materials, it was found that thermal contraction occurs also at low OCR values, but this is suction dependent – suction promotes thermal dilation. Under constant volume conditions, wetting results in a decrease of hydraulic conductivity followed by an increase. This is found to be related to changes in macro-pores size – wetting induces a decrease of macro-pores size followed by an increase due to the aggregates fissuring.

The presence of technological voids can form a loose zone with bentonite gel after hydration. This zone corresponds to the preferential path for water flow, with a relatively higher hydraulic conductivity. However, the swelling pressure depends only on the global bentonite void ratio, regardless of the technological void.

Conflict of interest

The authors wish to confirm that there are no known conflicts of interest associated with this publication and there has been no significant financial support for this work that could have influenced its outcome.

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