Effect of water salinity on the hydro-mechanical behaviour of granular bentonite as light backfill material

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ABSTRACT
Concepts for backfilling and sealing a deep geologic repository (DGR) for used nuclear fuel make use of remotely placed, bentonite-based “light backfill” (LBF). In some concepts LBF is used to fill construction voids in the DGR that cannot be practically backfilled using direct machine compaction or precompacted blocks. Other concepts use LBF as the primary backfill in the emplacement room. The compression, swelling, stiffness and hydraulic behaviour of granular bentonite LBF under one-dimensional oedometer testing conditions in the presence of distilled water and 100 g/l and 250 g/l CaCl$_2$ solutions is examined and compared.

RÉSUMÉ
Les concepts de remblayage et de scellement d’un dépôt géologique profond (DGP) pour les combustibles nucléaire usé utilisent de « remblai léger » (RL) à base de bentonite et qui est placé à distance. Dans certains concepts, RL est utilisé pour combler les vides de construction dans le DGP qui ne peuvent pas être pratiquement remblayés par l’utilisation directe d’une machine de compactage ou des blocs pré compactés. Autres concepts utilisent RL comme un remblai principal dans la salle d’emplacement. La compression, le gonflement, la raideur et le comportement hydraulique des granulés de bentonite RL sont examinés et comparés dans des conditions de test unidimensionnel à l’odomètre en présence d’eau distillée et des solutions de 100 g/l CaCl$_2$ et 250 g/l CaCl$_2$.

1 INTRODUCTION
Concepts for a deep geologic repository (DGR) for used nuclear fuel use swelling clay-based materials for backfilling and sealing the excavations associated with the repository. Backfill and buffer materials can be placed as highly compacted blocks, can be machine compacted in place or can be remotely placed by gravity pouring, pneumatic (shotcrete-like) placement and/or high-speed conveyance. Some DGR concepts use combinations of these methods. For example, Figure 1 illustrates a transverse cross-section of the emplacement room design put forward in Ontario Power Generation’s third case study for a deep geologic repository (DGR) for used nuclear fuel (Gierszewski et al. 2004). The design includes five different clay-based materials surrounding two used fuel containers aligned horizontally in the emplacement room. The Highly Compacted 100% Bentonite (HCB), the Compacted Buffer and Dense Backfill layers will be placed as large precompacted blocks. The space between the Dense Backfill and the emplacement room wall must be backfilled using a remote placement technique, such as illustrated in Figure 2. This space is inaccessible to direct machine compaction and too small and irregularly shaped for the practical placement of precompacted blocks of sealing materials. Backfill placed in this manner would have a lower in place density than direct machine compacted backfills and has been termed Light Backfill (LBF) in the Canadian program.

Figure 1. Third Case Study In-room Emplacement Geometry (after Gierszewski et al. 2004).

Figure 2. Remote Placement of Light Backfill Material.
Other DGR concepts (e.g. Spain, Switzerland and Germany) involve placement of the nuclear waste containers horizontally onto a bed of highly compacted bentonite blocks within emplacement rooms. The remaining space within the emplacement rooms is to be filled using remote methods, such as auger placement, with pelletized bentonite materials. Figure 3 shows the configuration of the large scale “Engineered Barrier” experiment performed at the Mont Terri Underground Laboratory in Switzerland. The granular bentonite pellet backfill was placed using an auger system. Advantages of this approach, as noted by Alonso and Hoffman (2007), are simpler and automated backfill emplacement, minimal voids and gaps in the sealing system after backfill emplacement and more accurate placement of the containers.

![Figure 3. Configuration of the Mont Terri Engineered Barrier Experiment (Fuentes-Cantillana and Huertas 2002).](image)

For the concept illustrated in Figure 1, the HCB layer adjacent to the container will have an as-placed dry density of 1.61 Mg/m$^3$ and Effective Montmorillonite Dry Density (EMDD) of 1.5 Mg/m$^3$. EMDD is defined as the mass of montmorillonite divided by the volume occupied by the montmorillonite and the volume of voids present in the system (Kjartanson et al. 2005 provide the detailed formulation for calculation of EMDD). EMDD is used to normalize the swelling and hydraulic behaviour of bentonite-aggregate mixtures (Dixon et al. 2002). Studies have shown that high clay-based sealing system dry densities and EMDD values will tend to minimize hydraulic conductivity, limit the viability and activity of microbes on and near the container (and thus limit the potential for microbially influenced corrosion) and maximize swelling pressures and self sealing and self healing potential.

Placement trials carried out on bentonite pellet materials and bentonite aggregate mixtures (Kjartanson et al. 2005; Martino and Dixon 2006) indicate that LBF materials can be pneumatically placed to EMDD values from about 0.8 to 1.0 Mg/m$^3$. Kjartanson et al. (2005) showed that post-emplacement compaction could increase the LBF EMDD up to about 1.2 Mg/m$^3$.

Immediately following container and sealing system emplacement, moisture will tend to be thermally driven from the sealing materials closest to the container towards the emplacement room wall and the outer sealing materials will be taking up moisture from the surrounding rock. The materials closest to the container will therefore tend to undergo thermal drying and shrinkage and the outer materials will tend to swell in the early stages after emplacement. In the longer term, on water uptake and saturation of the sealing system components, there will be a tendency for the components with high EMDD and high swelling pressure, such as the HCB blocks adjacent to the container, to expand and those with lower EMDD, such as the LBF, to compress. The stress-strain properties of the sealing materials play an important role in this “compliance effect”. A series of laboratory one-dimensional compression tests has been carried out on a granular bentonite LBF formulation to assess hydro-mechanical properties for compliance modelling.

The anticipated transient hydraulic and volume change paths followed by the sealing system components and the results of preliminary FLAC modelling carried out by Chandler (2005) were used to define testing protocols to make the tests representative of repository conditions. This assessment indicates that the LBF will first likely swell by up to about 20% and then be compressed by the swelling of the HCB in the longer term. Specifically, the preliminary modelling by Chandler (2005) suggests that the final strain in the LBF will be compressive by up to about 15%, while the HCB will be expanding by up to about 40%.

Crystalline rock of the Canadian Shield and sedimentary rock in southern Ontario are being considered as host rocks for a DGR (NWMO 2005). Gascoyne et al. (1987) and Mazurek (2004) have compiled data that show that groundwaters at proposed repository depths of 500 to 1000 m in crystalline rock of the Canadian Shield and the sedimentary rock in southern Ontario, respectively, can contain significant quantities of soluble salts. Salinities, in terms of Total Dissolved Solids (TDS) at proposed repository depths, can be >100 g/l in the Shield and >200 g/l in Ordovician-age sediments. Salt speciation is often Na-Ca-Cl at shallow depth trending to Ca-Na-Cl at greater depth (Baumgartner et al. 2008). The adverse effect that Ca-rich groundwater may have on the swelling, self-sealing and hydraulic performance of bentonite-based clay barriers is an important aspect of long term DGR performance. In this paper the one-dimensional compression, swelling and hydraulic behaviour and stiffness of granular bentonite LBF material in the presence of distilled water and 100 g/l CaCl$_2$ and 250 g/l CaCl$_2$ solutions is described and compared.

### 2 LBF MATERIAL AND TEST PROCEDURES

#### 2.1 LBF Material

National granular bentonite - industrial grade - LD-8, supplied by Bentonite Performance Minerals of Denver, Colorado was used as the LBF material in this study. The properties of the LD-8 granular bentonite are listed in Table 1. A specific gravity of soil solids of 2.75 was used in all weight-volume calculations for this LBF material.
Table 1. Properties of the LD-8 Granular Bentonite (after Kjartanson et al. 2005).

<table>
<thead>
<tr>
<th>Property</th>
<th>Value</th>
</tr>
</thead>
<tbody>
<tr>
<td>Montmorillonite Content, %</td>
<td>75</td>
</tr>
<tr>
<td>Nominal Granule Sizes, mm</td>
<td>2.36–0.85</td>
</tr>
<tr>
<td>Granule Dry Density, Mg/m$^3$</td>
<td>1.83</td>
</tr>
<tr>
<td>Granule EMDD, Mg/m$^3$</td>
<td>1.66</td>
</tr>
</tbody>
</table>

2.2 Test Procedures

Standard lever-arm consolidation frames with cells fitted with 50-mm-diameter by 19-mm-high consolidation rings were used for all one-dimensional compression tests carried out in this program. A total of nine tests were carried out. The average initial properties of the granular bentonite LBF samples, along with standard deviations of those properties, are shown in Table 2.

Table 2. Average Initial Properties of the Granular Bentonite LBF Samples.

<table>
<thead>
<tr>
<th>Property</th>
<th>Value ± Stnd Dev</th>
</tr>
</thead>
<tbody>
<tr>
<td>Water Content, %</td>
<td>11.20 ± 0.18</td>
</tr>
<tr>
<td>Height, cm</td>
<td>1.05 ± 0.03</td>
</tr>
<tr>
<td>Dry Density, Mg/m$^3$</td>
<td>0.93 ± 0.02</td>
</tr>
<tr>
<td>EMDD, Mg/m$^3$</td>
<td>0.76 ± 0.02</td>
</tr>
<tr>
<td>Void Ratio</td>
<td>1.97 ± 0.07</td>
</tr>
<tr>
<td>Degree of Saturation, %</td>
<td>15.7 ± 0.6</td>
</tr>
</tbody>
</table>

The target initial sample height was 1.0 cm which gave a target EMDD value of 0.8 Mg/m$^3$. This target initial EMDD value is at the lower range of the values achieved in the placement trials as described earlier. The granular bentonite LBF samples were poured air-dry into the consolidation ring and the surface was levelled using light finger pressure.

Rather than running ASTM standard-type consolidation tests, the tests were conducted in a manner to simulate the wetting and loading path of the LBF expected under in-situ, repository conditions as described in the Introduction. For the standard wetting/loading path, the LBF was allowed to swell to a target value of about 20% strain during initial fluid uptake from the reservoir. Sample swelling was closely monitored after the cell reservoir opened and the surface was levelled, and the applied stress was generally applied until the vertical deformation rate was less than about 0.02 mm/day.

Tests were conducted to compare the compression and swelling behaviour of the LBF with distilled water, and 100 g/l and 250 g/l CaCl$_2$ solutions in the consolidation cell reservoir. In addition, a test was carried out to examine the dependence of the one-dimensional compression and swelling behaviour of LBF on the wetting and loading path. Given this, the following series of one-dimensional compression tests was carried out:

- Samples tested with distilled water in the reservoir and allowed to swell to about 20% strain on water uptake (tests MN2 and MN3); this set of tests will hereafter be called "distilled water".
- Samples tested with 100 g/l CaCl$_2$ solution in the reservoir and allowed to swell to about 20% strain on solution uptake (tests MN5 and MN7); this set of tests will hereafter be called "100 g/l CaCl$_2".
- Samples tested with 250 g/l CaCl$_2$ solution in the reservoir and allowed to swell to up to about 20% strain on solution uptake (tests MN8, MN9, JPM1 and JPM5); this set of tests will hereafter be called "250 g/l CaCl$_2".
- A sample constrained during 250 g/l CaCl$_2$ solution uptake (test JPM4); this test will hereafter be called "250 g/l CaCl$_2$ constrained".

Once the samples came to equilibrium in the initial stage, the series of loading and unloading increments was applied.

3 RESULT AND ANALYSIS

3.1 Compression and Swelling Behaviour

The vertical strain versus time graphs for the loading and unloading increments for representative tests using distilled water (MN2) and 250 g/l CaCl$_2$ solution (JPM5) as the reservoir fluids are shown in Figure 4. Negative vertical strains correspond to swelling. The applied stress for each of the loading and unloading increments is indicated. These graphs show that there is significant hysteresis in the vertical strain when comparing the loading/compression with the unloading/swelling responses. This hysteresis is much more pronounced in the tests using CaCl$_2$ as the reservoir fluid than the tests using distilled water as the reservoir fluid.

Figure 5 illustrates the distinctly different initial swelling response of the LBF samples with distilled water in the reservoir versus either 100 g/l or 250 g/l CaCl$_2$ solution in the reservoir. For the sample with distilled water, increasing applied vertical stresses from 11 to 55 kN/m$^2$ needed to be applied to maintain the target swelling strain. The graph in Figure 5 shows that after the application of the 32 and 55 kN/m$^2$ stresses the sample began to swell slightly after some initial compression. This is likely due to time dependent hydration and swelling of the bentonite granules. The samples with 100 g/l and 250 g/l CaCl$_2$ solution achieved only about 16% to 7% swelling strain, respectively, under unloaded conditions. Tests MN8, MN9 and JPM5, also tested in the presence of 250 g/l CaCl$_2$ solution and
allowed to swell under unloaded conditions, swelled to about 8%, 7% and 10% strain, respectively.

Figure 4. Vertical strain versus time graphs for a) representative test using distilled water (MN2) and b) representative test using 250 g/l CaCl$_2$ solution (JPM5).

Figure 6 shows the first half hour of swelling response of the same tests shown in Figure 5. Note that for about the first half minute (0.008 hours) the rates of swelling are about the same for all three reservoir conditions. After this, the test with 250 g/l CaCl$_2$ solution (JPM1) equilibrates first, followed by the test with 100 g/l CaCl$_2$ solution (MN7) and then followed by the test with distilled water (MN2). This response is likely due to additional and prolonged hydration and swelling of the bentonite granules with lower reservoir salinities.

3.2 Void Ratio versus Applied Vertical Stress

Figure 7 shows the void ratio versus log applied vertical stress plots for the distilled water tests and the 100 g/l CaCl$_2$ tests. The two sets of tests show a high level of repeatability of response.

As illustrated in Figure 5 by test MN2, the tests with distilled water expanded significantly in the lower applied vertical stress range, giving a void ratio at the beginning of the loading phase higher than the initial, as placed void ratio of about two. Because of the limited initial swelling of the 100 g/l CaCl$_2$ tests under unloaded conditions (see Figure 5), the void ratio at the beginning of the loading phase for these samples is closer to the initial, as placed void ratio of about two. Figure 7 also shows that the distilled water tests have a steeper slope on the loading path (i.e. higher compression index) than the 100 g/l CaCl$_2$ tests. The rebound indices are similar in the higher stress range, but as the stress levels decrease the rebound index for the distilled water samples increase significantly. Also note that the rebound indices of MN5 and MN7 are virtually identical in spite of MN5 being unloaded in five increments and MN7 being unloaded in one increment. This indicates that the rebound index for these conditions is not stress path dependent.
Figure 8 compares the void ratio versus log applied vertical stress plots for a distilled water test (MN2), a 100 g/l CaCl₂ test (MN7), a 250 g/l CaCl₂ test (JPM5) and the 250 g/l CaCl₂ constrained test (JPM4).

Figure 8 shows that the void ratio versus log vertical stress plots for all of the tests in the presence of CaCl₂ are quite distinct from the distilled water test plot.

3.3 Constrained Modulus

The coefficient of volume compressibility ($m_v$) was calculated for the loading and unloading increments of the one-dimensional compression tests using the equation:

$$ m_v = \frac{1}{1+\varepsilon_0} \left( e_0 - e_1 \right) \frac{1}{\alpha_1 - \alpha_2} $$

[1]

where $\varepsilon_0$ is the void ratio corresponding to $\sigma_0^\prime$, the initial effective stress for the increment, and $\alpha_1$ is the void ratio corresponding to $\sigma_1^\prime$, the final effective stress for the increment (Bardet 1997). Furthermore, the constrained modulus ($M$) was calculated for the loading and unloading increments using the equation (Bardet 1997):

$$ M = \frac{1}{m_v} $$

[2]

The variation of constrained modulus ($M$) with the equilibrium EMDD for each loading increment of the tests within the four sample groups described in the Test Procedures section are shown in Figure 9. Data for the loading increments are used because the restraint provided by the LBF, in the compression mode, to the expansion of the HCB is the key consideration in the compliance modelling discussed earlier. Trendlines have been fit to the sample group data.

3.4 Hydraulic Conductivity

Hydraulic conductivity ($k$) values were calculated for the loading and unloading increments using the equation (Bardet 1997):

$$ k = c_v \left( m_v \right) \gamma_w $$

[3]

where $c_v$ is the coefficient of consolidation, $m_v$ is as defined previously and $\gamma_w$ is the unit weight of water. The $t_{90}$ values (time to 90% consolidation) used for calculating the $c_v$ values for the increments were determined using the square root of time graphical construction method (Bardet 1997).

The variation of $k$ values with the equilibrium EMDD for each test loading increment for the same four sample groups were calculated and are shown in Figure 10. The loading increment data are used because this is the likely path that the LBF will follow in the repository setting in the longer term.

It is evident from Figure 10 that $k$ generally increases with increasing CaCl₂ concentration and decreases with increasing EMDD. The trendlines for different salinities tend to be somewhat parallel, although there is some convergence of the data from the tests using CaCl₂ in the...
lower EMDD range. The change in k from distilled water to 100 g/l CaCl$_2$ is significantly larger than the change in k from 100 g/l to 250 g/l CaCl$_2$. The constrained test data are not significantly different from the other 250 g/l CaCl$_2$ data. For the tests with samples tested in the presence of 100 g/l CaCl$_2$, k values tend to be greater than $10^{-10}$ m/s with EMDD values less than about 1.0 Mg/m$^3$. For the tests with samples tested in the presence of 250 g/l CaCl$_2$, k values tend to be greater than $10^{-10}$ m/s with EMDD values less than about 1.3 Mg/m$^3$.

![Figure 10. Hydraulic conductivity versus EMDD for loading paths.](image)

4 SUMMARY

A series of one-dimensional compression tests was carried out on a LBF material composed of granular bentonite. The samples were prepared to an EMDD of about 0.8 Mg/m$^3$ at an air dry water content of about 11%. This relatively low EMDD, as compared with the higher EMDD of the HCB of 1.5 Mg/m$^3$, represents the lower bound of a practically achievable EMDD in a DGR using currently available remote placement techniques. Key aspects of DGR sealing system performance in the longer term are for the LBF to: maintain a low hydraulic conductivity; maintain a swelling and self-sealing ability; and maintain adequate stiffness to prevent excessive expansion of the HCB layer adjacent to the container. The last issue is particularly important as excessive expansion of the HCB layer could impact its ability to limit the viability and activity of microbes on and near the container, and microbially influenced corrosion of the used fuel container. Relatively high salinity, Ca-rich groundwaters are present in both the granitic and sedimentary host rock formations being considered for a DGR in Canada. A key objective of the one-dimensional compression tests carried out is to examine the potential effects that these groundwaters may have on the long term performance of LBF.

Key results of the one-dimensional compression tests are summarized as follows:

1. The swelling, and potentially the self-sealing ability of the granular bentonite LBF is adversely affected in the presence of 250 g/l CaCl$_2$ solution. In the initial swelling portion of the compression tests, the samples with distilled water achieved swelling strains of about 20% to 30% under applied vertical stresses of 32 kN/m$^2$ and 55 kN/m$^2$ while the samples with 250 g/l CaCl$_2$ solution achieved only about 7% to 10% swelling strain under unloaded conditions.

2. All of the tests showed significant hysteresis between the loading/compression and unloading/swelling paths. The hysteresis was more pronounced in the tests that used CaCl$_2$ in the consolidation cell reservoir. This means that future hydro-mechanical compliance modelling must use different constitutive parameters for the loading/compression and unloading/swelling paths.

3. The stiffness, in terms of constrained modulus (M) for the loading/compression path, increases exponentially with EMDD. The tests conducted in the presence of distilled water have higher M values than the tests conducted in the presence of CaCl$_2$ solutions up to an EMDD value of about 1.2 Mg/m$^3$. Of the samples tested in the presence of CaCl$_2$ solutions, the 100 g/l CaCl$_2$ tests tend to have the highest stiffness. The samples tested at 250 g/l CaCl$_2$ and allowed to swell initially had the lowest stiffness while initially constraining the granular bentonite sample in the presence of 250 g/l CaCl$_2$ solution tended to give a higher stiffness in compression.

4. Hydraulic conductivity (k) values calculated from the loading increments of the tests, using standard rate of consolidation theory, generally increase with increasing CaCl$_2$ concentration and decrease with increasing EMDD. The change in k from distilled water to 100 g/l CaCl$_2$ is significantly larger than the change in k from 100 g/l to 250 g/l CaCl$_2$. For the tests with samples tested in the presence of 100 g/l CaCl$_2$, k values tend to be greater than $10^{-10}$ m/s with EMDD values less than about 1.0 Mg/m$^3$. For the tests with samples tested in the presence of 250 g/l CaCl$_2$, k values tend to be greater than $10^{-10}$ m/s with EMDD values less than about 1.3 Mg/m$^3$.

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