GEOQuébec 2015
Challenges from North to South
Des défis du Nord au Sud

Swelling characteristics of bentonite based barriers for deep geological repository for nuclear wastes: Impact of underground water chemistry and temperature

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ABSTRACT
The presence of bentonite-sand in deep geological repositories surrounded by aggressive environments of highly saline underground water, high temperature and under confining stress may cause mineralogical changes for bentonite-based materials and impact their physical and physiochemical properties. This could affect the bentonite performance as an engineered barrier system and the overall safety of the deep geological repositories. The objective of this research is to investigate the impact of Ontario's underground water salinity and heat generated by the nuclear wastes on the swelling capacity of bentonite-sand engineered barrier materials. Free-swelling and swelling pressure tests have been performed on bentonite-sand mixtures subjected to various chemical (groundwater chemistry) and thermal (heat generated) loading conditions. The obtained results have shown that the groundwater chemistry can significantly deteriorate the swelling properties of the tested bentonite-sand barrier material, whereas the temperature has little effect on the bentonite-sand swelling potential.

RÉSUMÉ
La présence de bentonite-sable dans les dépôts en couches géologiques profondes situés dans des environnements agressifs marqués par des eaux souterraines très salées, hautes températures et des contraintes de confinement, peut causer des changements minéralogiques dans ces matériaux à base de bentonite. Ceci peut avoir un impact sur leurs propriétés physiques et physico-chimiques et ainsi affecter la performance de la bentonite comme une barrière technique et aussi la sécurité globale des dépôts en couches géologiques profondes. L'objectif de cette recherche est d'étudier l'impact de la salinité de l'eau souterraine de l'Ontario et de la chaleur générée par les déchets nucléaires sur la capacité de gonflement de ces matériaux de barrière technique. Des essais de gonflement libre et de gonflement sous pression ont été réalisés sur des mélanges de bentonite-sable soumis à diverses conditions chimiques (chimie des eaux souterraines) et thermiques (chaleur produite). Les résultats obtenus ont montré que la chimie des eaux souterraines peut détériorer considérablement les propriétés de gonflement des matériaux de barrière (bentonite-sable) testés, alors que la température a peu d'effet sur le potentiel de gonflement du matériau, bentonite-sable.

1 INTRODUCTION

More than 30 countries around the world, including Canada, have proposed construction of very deep geological repositories (DGRs) to permanently store their radioactive wastes for thousands years. DGR would be located at a depth of 400 to 1000 m under the ground surface. The main task of DGRs is to isolate nuclear waste inside a multi-barrier system to prevent radionuclide contamination (RWM, 2003), as illustrated in Figure 1. This multi-barrier system consists of a natural geological barrier (host rock) and an engineered barrier system (EBS) to provide a safe and secure place for nuclear waste for thousands of years in order to protect humans and their environment (RWM, 2003; NWMO, 2010).

The host rock, commonly made of sedimentary or crystalline rock formation, plays a very important role in the selection of a DGR location. The properties of the host rock and underground water are among the major factors that control the site specifications. The host rock should have very low permeability to reduce underground water movement. Long-term stability and high strength are needed to sustain a stable geological environment for the safe and long-term storage of nuclear waste (Quintessa, 2011; NWMO, 2012).

Compacted bentonite-sand has been proposed as an appropriate material that would be a component of the EBS in a DGR (Cho et al; 1999; Karnland; 2006, Quintessa; 2011). This is due to its contents of clay minerals of the smectite group which has excellent swelling ability and adsorption capacity, high cation exchange capacity (CEC), low permeability and thermal conductivity, high plasticity, and self-sealing properties (Pusch, 1982; Mitchell, 1993; Melamed and Petteri, 1996; Quintessa, 2011). The initially unsaturated engineered bentonite-based barrier material will be placed in the DGR, and once it comes into contact with the host rocks, it will adsorb the groundwater at a very slow rate due to the low permeability and very small pore size from the developing swelling pressure and sufficient confinement (Delage et al., 2010; Man et al., 2010; Quintessa, 2011).

The Nuclear Waste Management Organization (NWMO) is currently investigating sixteen locations in Ontario and three locations in Northern Saskatchewan to construct a DGR for burying nuclear wastes (NWMO, 2012). For example, the construction of a DGR for low and intermediate level radioactive waste (L&ILW) in the Municipality of Kincardine at a depth of 680 m is currently being investigated. However, In Ontario at the depth of the proposed DGR, the groundwater is highly saline. It contains Na-Ca-Cl or Ca-Na-Cl type of water, and the total dissolved solids (TDS) range from 200,000 to 400,000 mg/L (Dollar, 1988; NWMO, 2011; 2012). The presence of bentonite-sand in DGRs, surrounded by an aggressive environment of high salinity with high underground temperature and under confining stress may cause mineralogical changes in bentonite-based materials and alter their favorable engineering (swelling) properties. This would affect the bentonite performance in an EBS as well as the overall safety of the DGR. Thus, there is crucial need to understand the effect of groundwater salinity and heat generated by radioactive decay on the swelling properties of bentonite-sand barrier materials. The objective of the present study is therefore to experimentally assess the effect of the groundwater water salinity found in Ontario and heat generated by nuclear waste on the swelling properties and mineralogical composition of bentonite-sand based materials.

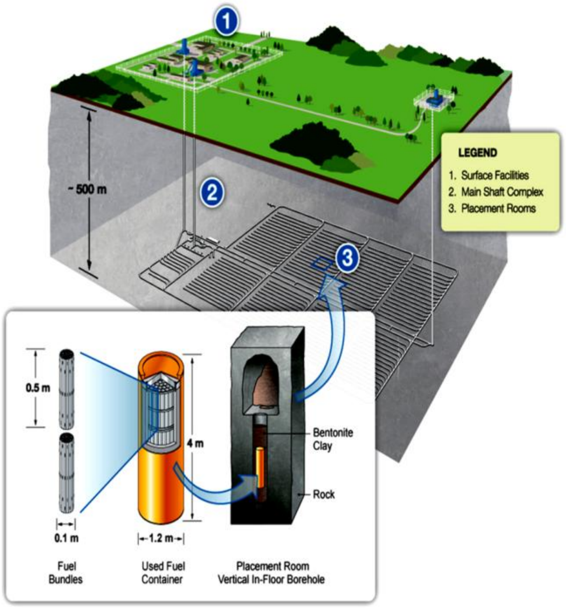

Figure 1 Deep geological repository design in crystalline rock (Villagran, 2012).

2 EXPERIMENTAL PROGRAM

2.1 Materials

All of the experiments have been conducted by using a mixture of MX-80 bentonite-sand (30% and 70% by mass, respectively) mixed with one of three water solutions, which simulate the different groundwater salinity or chemistry found in Ontario.

The results of the X-ray diffraction (XRD) analysis have shown that the MX-80 bentonite contains 92% montmorillonite and variable quantities of feldspar, quartz, calcite and kaolinite. The CEC is 105 meq/100 g and Na^+ is the predominant exchangeable cation. The grain size distribution of the bentonite-sand mixture (30/70) is shown by Figure 2, whereas it's chemical composition is given in Table 1. The MX-80 bentonite used has a liquid limit of 530%, a plastic limit of 48.5% and an average specific gravity of 2.66.

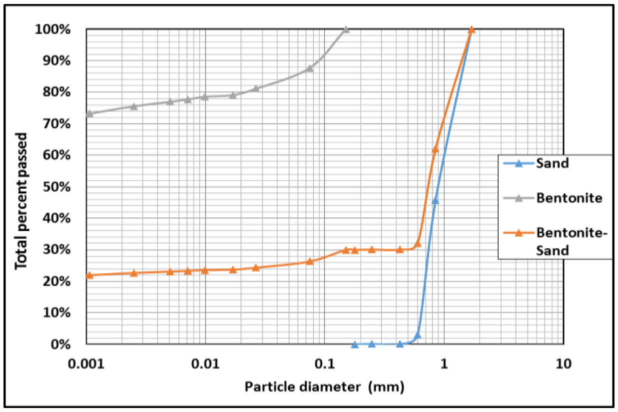

Figure 2. Sand, bentonite and bentonite- sand (30/70) grain size distributions

Table 1. Chemical composition of MX-80 bentonite

Element	Composition	Percentage (%)
Silicon Dioxide (total)	SiO_2	63.59
Aluminum Oxide	Al_2O_3	21.43
Iron Oxide	Fe_2O_3	3.78
Calcium Oxide	CaO	0.66
Magnesium Oxide	MgO	2.03
Sodium Oxide	Na_2O	2.70
Potassium Oxide	K_2O	0.31
Bound Water	H_2O	5.50

Three different water solutions are used in all of the experiments; distilled water (DW) and two water solutions (Table 2). The two water solutions were prepared in the laboratory to obtain the same chemical composition as that of the underground water located at more than a depth of 700 m in southwestern Ontario (Dollar 1988; NWMO 2011).

Table 2. Chemical composition of Water Solutions G and T (g/l) used in this study (sources: Dollar, 1988; NWMO 2011).

Element (g/l)	Distilled water (DW)	Guelph water (G)	Trenton water (T)
$CACL_2$	0	159.0	64.0
NACL	0	100.0	102.0
$MGCL_2$	0	34.3	21.6
KCL	0	6.3	3.2
K_2SO_4	0	0.4	1.2
TDS	0	300.0	192.0
PH	7	5.7	6.5

2.2 Specimen preparation and mix proportions

Bentonite-sand mixture specimens were mixed with either DW, or Water Solution T or G, and all of the specimens were compacted to an initial dry density of 1.7 g/cm³ and water content of 15%. The last step of the preparation for the specimens was the treatment for 15 days at three different temperatures of 23°C, 40°C and 80°C, respectively. The different groups of samples prepared are given in Table 3.

Table 3. Mixing solutions and treatment temperatures of the samples prepared.

	Samples	**Mixing and saturating solution**	**Treatment temperature**
Group (1)	DW23	Distilled water	23^0 C
	DW40		40^0 C
	DW80		80^0 C
Group (2)	T23	Water solution T	23^0 C
	T40		40^0 C
	T80		80^0 C
Group (3)	G23	Water solution G	23^0 C
	G40		40^0 C
	G80		80^0 C

2.3 Swelling tests

One-dimensional free swell, swelling deformation, and swelling pressure tests were performed on the compacted specimens with an initial dry density of 1.7 g/cm³ and water content of 15%, by following ASTM standard procedures. All of the tests were conducted at the laboratory temperature (23°C). Each sample was soaked during the swelling tests in the same mixing water solution.

The one-dimensional free swell test was conducted on compacted and treated specimens of 63 mm in diameter and 13 mm in height, by following ASTM standard D-4546-08. The specimens were inserted into 250 ml beakers with a 63 mm diameter (the same diameter as the specimen diameter) to prevent lateral swelling. The specimens were then saturated with 150 ml of the appropriate water solution. Groups (1), (2) and (3) specimens (see Table 3) were saturated with DW, and Water Solutions T and G, respectively. The vertical free swell strain for each specimen was measured with time by using dial gauges until the specimens reached full saturation and stopped swelling between 7 to 39 days.

The swelling pressure measurements were conducted by inserting the compacted and treated samples into a consolidometer ring (63 mm in diameter x 13 mm in height) between two filter papers and porous stones under a vertical seating load of 8.25 kPa. The samples were saturated by using the desired water solution from the top. Group (1), (2) and (3) specimens were floated with DW, and Water Solutions T and G, respectively, until reaching full saturation from about 7 to 36 days. The specimens were allowed to swell up to 0.01 mm and then small pressure increments were added to bring the sample back to its initial volume. The same procedure was repeated until the specimens stopped swelling at their fully saturated state. The swelling strain was measured with time by using a dial gauge. The specimen swelling capacity was obtained by recording the pressure needed to keep the sample volume constant with time.

2.4 Mineralogical analysis

XRD analysis was conducted on the pure bentonite and treated bentonite-sand samples to determine the mineralogical changes due to the interactions with the highly saline water solutions and heat. X-ray powder diffraction analyses were performed by using a Philips X'Pert diffractometer with a Cu source and High Score Plus analysis software. The XRD patterns were measured at room temperature with an instrument power of 45 kV, 40 mA and an automatic divergent slit and 0.2 mm receiving slit. The scans were between 2-60 degrees 2Theta with a step size of 0.02 degree 2theta and 0.6 s continuous scan time per step.

3 RESULTS AND DISCUSSIONS

3.1 Effect of temperature on the swelling characteristics of the studied bentonite-sand material

The obtained results have shown that the temperature does not have a significant impact on the swelling capacity of the studied bentonite-sand based barrier material. Typical results of the effect of temperature on the swelling strain and pressure of the studied bentonite based material are presented and discussed below.

Figure 3 depicts typical results of the effect of temperature on the swelling strain of the compacted bentonite-sand. It can be observed that the sample mixed with DW and kept at room temperature (DW23) records a maximum swelling strain of 198% within 32 days. The sample treated at 80°C (DW80) records a swelling strain of 194.5% within 30 days. The difference between these two values of swelling strain is minor. The sample treated at 80°C shows less than a 2% decrease in the swelling strain.

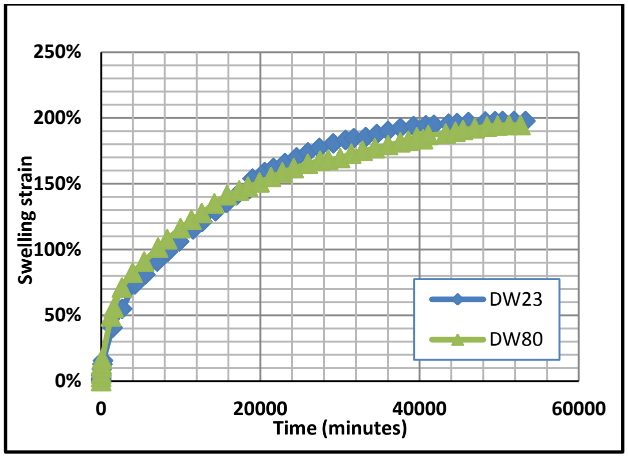

Figure 3. Effect of treatment temperature on the swelling strain of the studied compacted bentonite-sand

Figure 4 provides an example of results with regards to the effect of temperature on the swelling pressure of the studied engineered barrier material. The swelling pressure vs time curve of the sample treated at 80°C (DW80) is compared with that of the sample treated at room temperature (23°C, DW23). The sample treated at 80°C shows less than 12% decrease in the swelling pressure compared to the samples treated at 23°C. DW23 records a pressure of 228 kPa, which is in good agreement with the test results of Pusch (1998, 2001) and Quintessa (2011) on 30/70 MX-80 bentonite/ballast mixtures. DW80 shows swelling pressures of 201 kPa. These results fall in good agreement with those in an experimental study by Pusch (2000) on bentonite powder steam treated at 90°C and 110°C, which showed no significant changes in the swelling pressure with increases in the temperature.

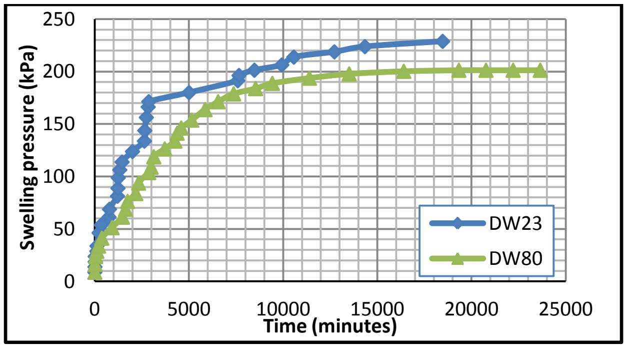

Figure 4 Impact of the treatment temperature on the swelling pressure of the compacted bentonite-sand.

3.2 Effects of groundwater chemistry on the swelling characteristics of the studied bentonite-sand material

The results obtained have shown that the groundwater salinity has a significant impact on the swelling strain and pressure, i.e. on the swelling properties of the studied engineered barrier materials.

For example, Figures 5 and 6 show the results of the effect of the salinity of the groundwater on the swelling strain and pressure of the studied materials, respectively. From Figure 5, it can be observed that the sample mixed and saturated with DW records a swelling strain of 198%, while the samples mixed and saturated with Water Solutions T and G record a swelling strain of only 8% and 4.7%, respectively. The swelling pressure drops from a pressure of 228 kPa for the sample mixed and treated with DW to 16.35 kPa and 13.35 kPa for the samples mixed and treated with Water Solutions T and G, respectively (Figure 6). The porewater chemistry reduces the swelling pressure by more than 90% and the swelling strain by 95%. These results are in good agreement with those obtained by Pusch (1998, 2001) and Bradbury and Baeyens (2002), Dhawan and Rao (2012).

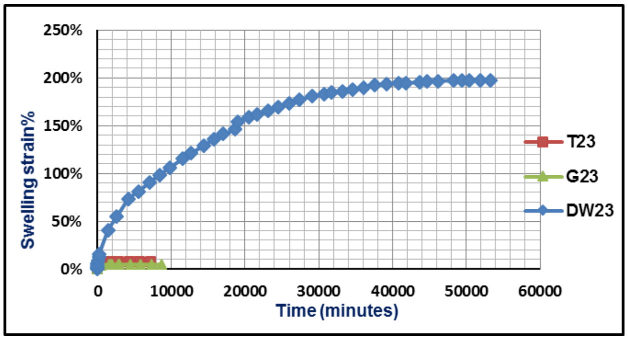

Figure 5. Groundwater chemistry impacts on the swelling strain of bentonite- sand based barrier material.

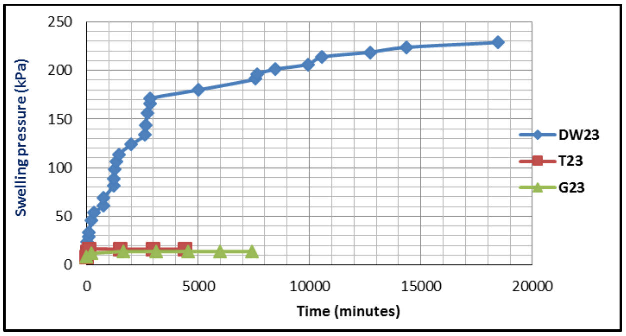

Figure 6. Groundwater chemistry impacts on the swelling pressure of bentonite- sand based barrier material.

This observed severe degradation of the swelling capacity or properties of the bentonite-sand swelling exposed to highly saline water results from the mineralogical and chemical changes that occur in the barrier material because of the chemical attacks from the salinity. Ca replaced the amounts of the initial Na of the Na-montmorillonite and produced Ca-montmorillonite with less swelling ability, whereas K and Al replaced the Na and Si and converted most of the montmorillonite to illite, which is characterized by a much lower swelling ability. These arguments are supported by the results of the XRD analyses performed on the samples DW23, G23, G40 and G80, presented in Figure 7. This figure shows that the mineralogical composition (particularly the montmorillonite content) of the samples mixed with mildly acidic (pH < 6) water solutions, that is, Water Solutions G or T, treated at temperatures of 23°C, 40°C and 80°C for 15 days, has changed. The montmorillonite-to-illite conversion, and Na-montmorillonite conversion to Ca-montmorillonite in specimens G23, G40 and G80 can be identified. The XRD spectra confirm the smectite -to- illite conversion and cation exchange processes. Indeed, the XRD spectrum of DW23 shows that the first peak for Na-montmorillonite is at (2ɵ) 8.92^0 and the basal spacing is d (001) = 9.90 Å, while G23, G40 and G80 show their first peaks at (2ɵ) 6.288^0, 6.200^0 and 6.632^0 with lower reflection intensities and d (001) = 14.04, 14.24 and 13.14 Å, respectively. This observed shift to the left of the first peaks and increase of the interlamellar spacing are due to the ion diffusion and cation exchange processes. The peak shifting and lower reflection intensity indicate a reduction in the swelling component (Moore and Reynolds, 1989). Na^+ was substituted by Ca^{2+} which resulted in the change of montmorillonite from a sodium-rich to a calcium- rich form with lower swelling capacity. In addition, K^+ replaced Na^+ and Al^{3+} replaced Si^{4+} and montmorillonite converted to a low-swelling component, illite, in the presence of the mildly acidic water solutions, that is, Water Solutions T and G, (Table 2), silica sand, and accessory minerals such as cristobalite, albite (plagioclase feldspar) and quartz. Therefore, the swelling potential of the samples exposed to the aforementioned solutions sharply decreases.

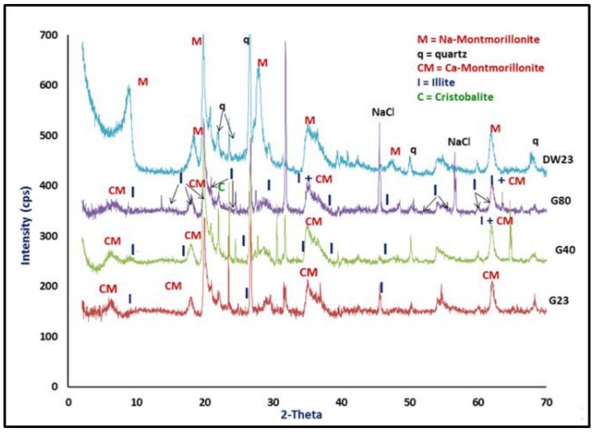

Figure 7. XRD patterns of DW23, G23, G40 and G80

4 CONCLUSIONS

The following conclusions can be drawn:

- The temperature does not have a significant impact on the swelling capacity of the studied bentonite-sand based barrier material.
- The porewater chemistry or the groundwater chemistry found in Ontario has a significant impact on the swelling capacity of bentonite-sand barrier material. The swelling ability of the barrier material is significantly reduced when exposed to saline groundwater.
- The XRD analyses demonstrate that the bentonite mineralogical compositions have changed. Most of the Na-montmorillonite has converted to Mg, Ca-montmorillonite and illite when treated with highly saline water solutions in the presence of silica sand and accessory minerals. The swelling component or the montmorillonite has become Ca-montmorillonite which has a lower swelling capacity as well as low-swelling component, or illite.
- The results of this research provide valuable information that will contribute to a better understanding of the impacts of the groundwater chemistry found in Ontario and the heat generated by nuclear waste on the long term performance of a DGR in Ontario.

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