Thermal property testing of an engineered barrier for use in a deep geological repository

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ABSTRACT
There are over 2.5 million bundles of used nuclear fuel in Canada and upwards of an additional 2 million bundles anticipated to be generated in the future. Canada, and all countries around the world that have taken a decision on long-term storage of spent nuclear fuel, has decided on central storage within a deep geological repository. A deep geological repository provides centralized containment and isolation of the used nuclear fuel hundreds of meters below ground. Spent nuclear fuel bundles are to be placed at depth within containers and surrounded by engineered barriers. A key component of the performance of a repository is the capability of the engineered barriers to transfer the thermal energy to the surrounding geosphere. In this paper thermal property testing of a high density bentonite material is presented. The challenges associated with high density specimen preparation are highlighted and preliminary results are reported.

RÉSUMÉ
Il y a plus de 2,5 millions de grappes de combustible nucléaire utilisé au Canada et plus de 2 millions de grappes additionnelles à être générées sont anticipées pour le futur. Le Canada et les autres pays autour du monde, qui ont pris la décision d'un stockage à long terme des déchets de combustible nucléaire, se sont décidés sur un stockage central dans un dépôt géologique profond. Un dépôt géologique profond fournit un confinement centralisé et une isolation du combustible nucléaire utilisé à des centaines de mètres sous le sol. Les déchets de combustible nucléaire seront placés en profondeur dans des conteneurs entourés de barrières techniques. Un élément clé de la performance d'un dépôt, est la capacité des barrières techniques à transférer l'énergie thermique à la roche environnante. Dans cet article, des essais de qualité thermique de matériel à grande masse volumique de bentonite sont présentés. Les défis associés avec la préparation d'échantillons à grande masse volumique sont mis en évidence et les résultats sont rapportés.

1 INTRODUCTION

Within Canada there are more than 2.5 million bundles of spent nuclear fuel with another approximately 2 million bundles to be generated in the future. Canada, and every country around the world that has taken a decision on management of spent nuclear fuel, has decided to on long-term containment and isolation of the fuel within a deep geological repository. At depth, a deep geological repository (Figure 1) consists of a network of placement rooms where the bundles will be located within containers and surrounded by an engineered barrier system.

Amongst other design aspects, the engineered barriers will transfer the thermal energy from the spent nuclear fuel to the surrounding geosphere. The barriers will be placed in a complex thermal-hydraulic-mechanical-chemical environment. The environment will include competing gradients of groundwater pressure driving moisture into the repository and thermal gradients driving moisture out. A current design criterion of the repository is to keep temperatures below $100^{o}C$ (Maak 2006). Therefore the thermal properties of the engineered barriers are a critical component of the system. Barrier materials will be at variable saturation levels and temperatures over their design life. An experimental program was initiated to measure the thermal properties of a number of potential barrier materials under variable moisture and temperature conditions. In this paper the experimental methodology for thermal property measurement is presented along with preliminary results. High density specimen preparation methods are also highlighted.

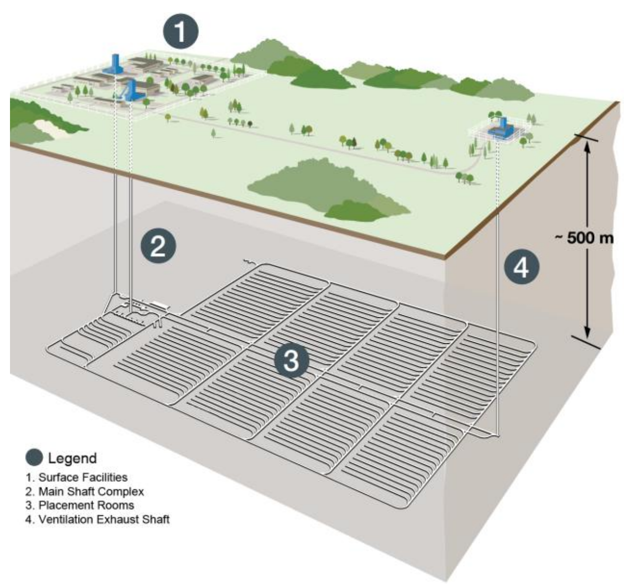

Figure 1. Concept of a deep geological repository (after NWMO 2015)

2 MATERIALS AND METHODS

2.1 Material Description and Preparation

A number of engineered barriers are proposed for use within the DGR. Thermal property testing, reported here, is performed on Highly Compacted Bentonite (HCB), which is composed of 100% bentonite clay to a dry density of 1.7 Mg/m^3 (Russell and Simmons 2003). The test specimens were prepared using National Standard Bentonite acquired from Bentonite Performance Minerals LLC.

2.2 High Density Specimen Preparation

HCB specimens are prepared using a process based on Martino et al. (2010) to achieve reliable moisture content and density. The constituent bentonite is placed within an oven at 105^{o}C for at least 24 hours. The material is removed from the oven, sealed, and allowed to come to thermal equilibrium with the laboratory. Then the quantitiy of bentonite required for specimen preparation is removed and placed in a mixing bowl. Water is added to the bentonite via misting with a spray bottle to achieve the target moisture content. Following mixing, the soil is placed within two sealed bags in a fridge for at least 48 hours for moisture equilibrium. Each weekday the soil is mixed inside the bag to encourage moisture equilibrium.

The high target dry density values required for HCB (ρ_d= 1.7 Mg/m^3) necessitates a specialized compaction method. In addition, drier specimens are damaged during removal from a compaction mould. Therefore two types of compaction moulds are manufactured to compact high-density specimens (Figure 2a and Figure 3a). Wetter specimens are prepared by compacting two adjacent cylindrical pucks in a 50 mm-diameter mould (Figure 2a). A piece of plastic wrap was placed between the pucks to allow them to be separated after compaction. This type of test is termed a ‘two-sided test’ as the thermal conductivity sensor (Figure 2b) is placed between two soil specimens (Figure 2c). Two 50 mm-diameter by 20-mm pucks are used in each two-sided test.

A second mould was constructed to allow for compaction and testing of drier specimens without removal from the compaction mould. Photographs of the compaction and thermal property testing are given in Figure 3a-c. In Figure 3a the compaction mould is shown with a slot cut into the side of the mould to allow for placement of the thermal conductivity sensor on the top face of the compacted specimen. During compaction the slot is covered to avoid soil spillage and loss of confinement. Figure 3b shows the compacted specimen with the thermal conductivity sensor in place. A block of insulation is placed above the specimen to limit thermal energy loss (Figure 3c) and a normal stress encourages contact between the sensor and the specimen. This type of test is termed a ‘one-sided test’ as the thermal conductivity sensor is only in contact with the test specimen on one side. One-sided test specimens are compacted to 20 mm-height by 75 mm-diameter.

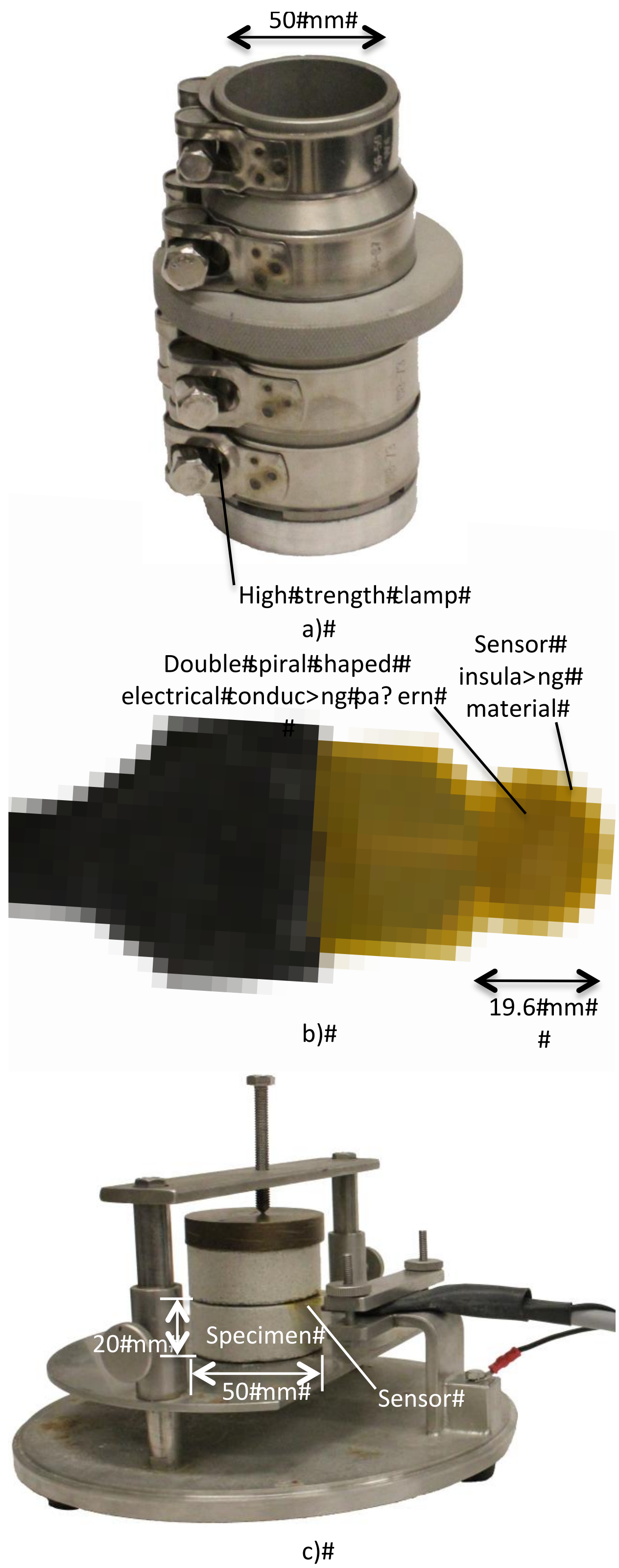

Figure 2. Two-sided thermal property test: a) 50 mm-diameter compaction mold and b) thermal conductivity sensor and c) two-sided test with sensor placed between two pucks.

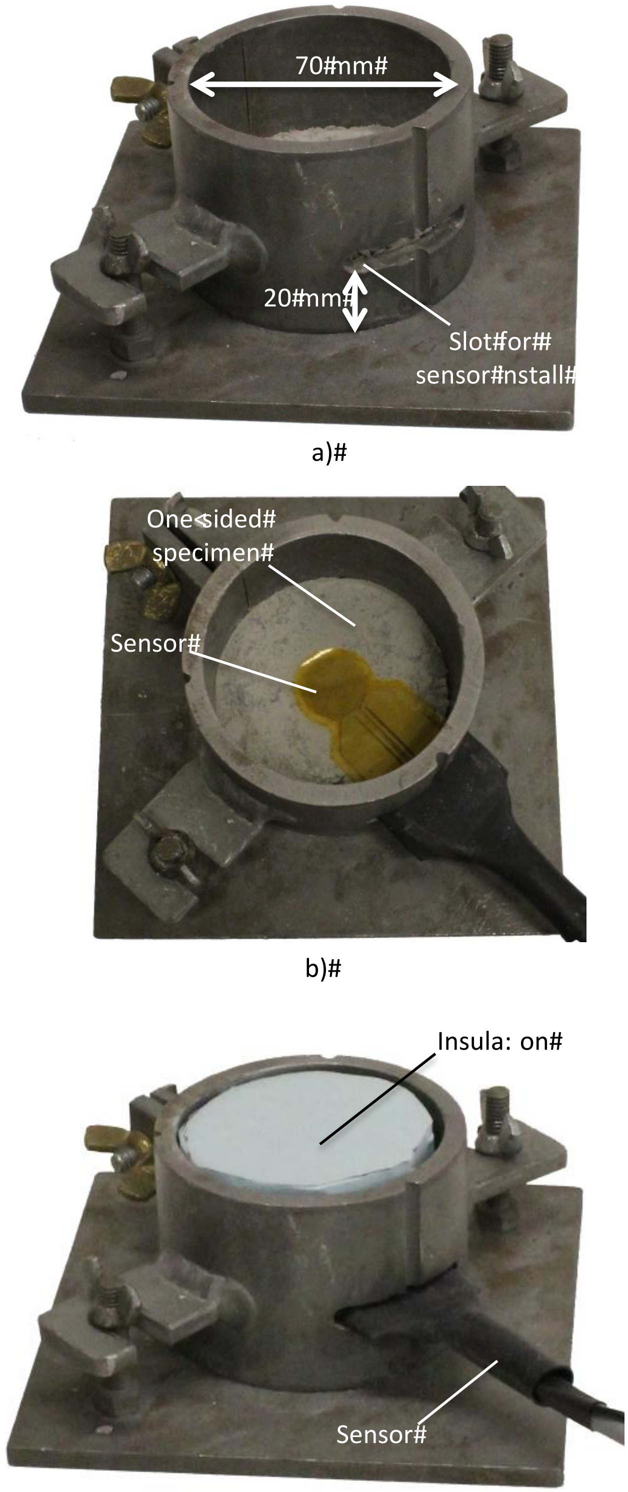

Figure 3. One-sided thermal conductivity test: a) compacted soil in 70 mm-diameter mold, b) thermal conductivity sensor placed on soil, and c) insulation placed on top of sensor.

Static compaction was selected for specimen preparation as it results in a specimen with uniform density over a wide range of moisture content (Radhakrishna et al. 1989). A strain-based criterion was developed to the compaction procedure using a compaction frame. Each compression stage consisted of a 10-second interval in which the specimen was over compressed by 0.8 mm to allow for elastic rebound to the target height.

Large compression loads were required to achieve the high target dry density (ρ_d= 1.7 Mg/m^3) at the cross-sectional area necessitated for thermal conductivity testing. Compaction was performed using a 30,000 lb (130 kN) electromatic universal testing machine by GE (General Electric). Typical results in terms of compaction pressure and force versus degree of saturation and gravimetric water content are plotted in Figure 4. Compaction force is a minimum at 100% saturation and increases for lower moisture contents. Compaction force increases two-fold from 40 kN to over 90 kN at Sr=10%.

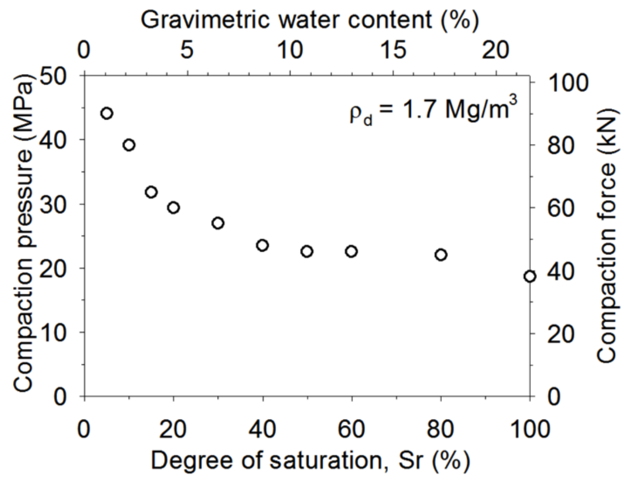

Figure 4. Compaction results to achieve dry density ρ_d=1.7 Mg/m^3 in terms of compaction pressure and force versus degree of saturation and gravimetric water content.

2.3 Thermal Conductivity Test Interpretation

The device used in the thermal property testing is a Hot Disk Thermal Constants Analyser (Hot Disk 2014). This test method is based on a Transient Plane Source Technology, which allows for measurement of thermal conductivity, thermal diffusivity as well as heat per unit volume of the material during a single test. The method uses a transiently heated planar sensor (Figure 2b), which consists of an electrical conducting pattern in a shape of double spiral etched out of the thin sheet of nickel. Covering both sides of the sensor are thin sheets of an insulating material (Kapton). When carrying out a measurement, the sensor is both a heat source and a dynamic temperature sensor. The sensor is assumed to be located within an infinite medium. In the two-sided test the sensor is sandwiched between two-pieces of soil (Figure 2c). In the one-sided test the sensor is located on one side with the specimen to be tested and the other side with an insulation material (Figure 3b and Figure 3c). During a test, the thermal wave generated by the sensor must not reach the outer boundary of the specimen. The thermal wave penetration distance is defined as the probing depth (Δp) and calculated as:

$$\Delta p = 2\sqrt{\kappa t} \qquad [1]$$

where κ is a thermal diffusivity of soil specimen and t is the measurement time. In order to measure thermal properties accurately, any distance between sensor edge and any outer of the specimen must exceed the probing depth (Δp).

The sensor applies a constant power to the specimen and also measures the incurred temperature change. Tests are interpreted by solving the thermal conductivity equation assuming that hot disk consists of a certain number of concentric ring heat sources located in an infinitely large sample. For the configuration specific to the sensor used in the tests, temperature increase of the specimen surface is given as:

$$\Delta T_i + \Delta T_{avg}(\tau) = \left(\frac{1}{\alpha}\right)\left\{\left[\frac{R(t)}{R_0}\right] - 1\right\} \quad [2]$$

where $\Delta T_{avg}(\tau)$ is the average temperature increase of the specimen surface in contact with the sensor surface, ΔT_i is the constant temperature difference that develops over the insulation on the sensor, R_0 is the resistance of the disk prior to heating at time (t) =0 and α is the temperature coefficient of resistivity of the insulating layer.

The temperature difference across the sensor's insulating layer, ΔT_i, becomes constant over a short period of time Δt, which can be estimated as:

$$\Delta t = \left(\frac{\delta^2}{\kappa_j}\right) \qquad [3]$$

where δ is the thickness of the sensor's insulating layer and κ_j is the thermal diffusivity of the insulating layer. Generally Δt is less than a 10 s in the experiments.

Solving the thermal conductivity equation gives the time dependent temperature increase of the specimen surface as:

$$\Delta T_{avg}(\tau) = \left(\frac{P_0}{\pi^{1.5} r \lambda}\right) D(\tau) \qquad [4]$$

where P_0 is power output from the sensor, r is the radius of the sensor disk, λ is the thermal conductivity of the soil specimen and $D(\tau)$ is a dimensionless time function with τ defined as:

$$\tau = \sqrt{\frac{t}{\theta}} \qquad [5]$$

where t is the time measured from the start of transient recording and θ is characteristic time calculated as:

$$\theta = \frac{r^2}{\kappa} \qquad [6]$$

where κ is thermal diffusivity of specimen.

By constructing a graph of increase in temperature versus $D(\tau)$, a straight line is obtained. The y-intercept of the line is ΔT_i. The slope of the line is a function of both the specimen's thermal conductivity, λ, and specimen's thermal diffusivity, κ. Therefore an iterative process is performed to solve for both thermal conductivity and thermal diffusivity.

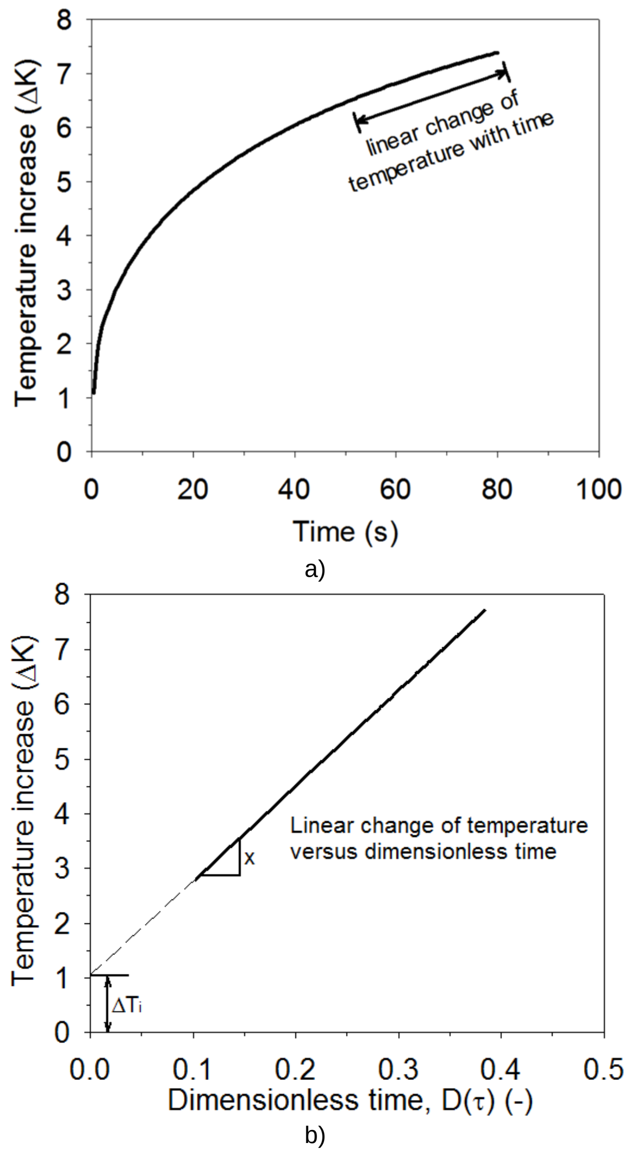

Figure 5. Typical thermal conductivity test result: change in temperature versus: a) time and b) normalized time, $D(\tau)$.

3 EXPERIMENTAL RESULTS

A thermal property test consists of applying a constant power to the specimen and measuring the temperature change. Sensor 8563 is used in tests reported here (Hot Disk 2014). From guidelines given by the manufacturer, a heating power of 0.5 to 1 W and measurement time of 80-160 s has been used for high density, clay-based materials. A typical test result plotted as change in temperature versus time is plotted in Figure 5a. Initially, a rapid increase in temperature is measured followed by a linear increase in temperature with time. Using the procedure described above the time data is normalized to calculate $D(\tau)$ and replotted in Figure 5b. In this version a clear linear relationship is observed, which allows for interpretation of the thermal properties.

Selected thermal conductivity results are plotted in Figure 6. In general thermal conductivity increases with degree of saturation. The thermal conductivity of water is greater than the clay particles and air. Therefore filling of the air voids with water causes an increase on the bulk thermal conductivity of the multi-phase system. Generally a linear relationship between thermal conductivity versus degree of saturation is interpreted from the data.

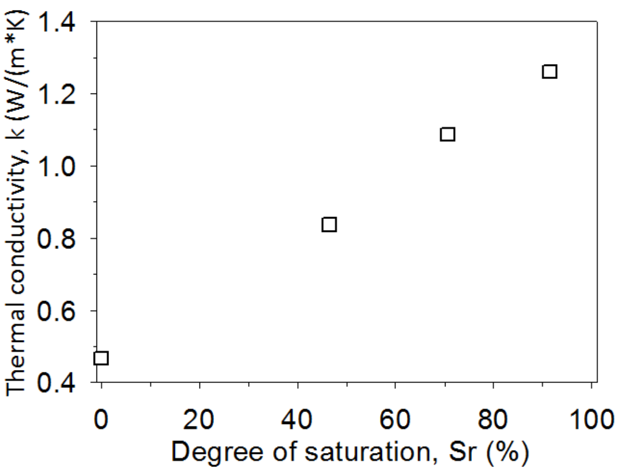

Figure 6. Thermal conductivity measurements as a function of degree of saturation.

4 SUMMARY

The current concept for Canada's millions of bundles of spent nuclear fuel is a deep geological repository. During the transient phase of the deep geological repository a number of thermal, hydraulic, chemical and mechanical aspects of the engineered barriers must be considered. The engineered barriers within the reopsitory will be under a wide range of physical phenomena including temperature gradients as the containers release their thermal energy. A current design criterion of the repository is for temperatures to remain less than 100°C. Therefore the characterization of the thermal properties of the engineered barriers as a function of moisture content and temperature is a key component of the analysis. In this paper the thermal property test methodology and select results on a highly compacted bentonite are presented. The elevated compaction effort required to prepare high density specimens is highlighted. In general, thermal conductivity generally increases with degree of saturation. Future experimental programs will examine the range of anticipated moisture contents within the repository.

ACKNOWLEDGEMENTS

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