THE PREDICTION MODEL OF THERMAL CONDUCTIVITY OF SAND-BENTONITE BASED BUFFER MATERIAL

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ABSTRACT
The thermal conductivity of sand-bentonite based buffer materials is a key factor for the design of HLW depository. In the Thermal-Hydraulic-Mechanical environment, the thermal conductivity varies due to the change in clay density, the water content, and the volumetric fraction of sand or crushed granite. In this article, an improved thermal probe method for the measurement of thermal conductivity is proposed. The probe is placed within the sand-bentonite powder inside the specially designed mold which the volume can be controlled by the position of the compacting piston. While the clay density reaches to a designated level, the measurement is executed to evaluate the thermal conductivity. With repeating the procedure, the relationship of clay dry density and the thermal conductivity can be established in one specimen. The weight water content of the bentonite is adjusted by placing in a humid chamber or in an oven for different periods. The relationship of thermal conductivity with clay dry density, water content, and sand or crushed granite is well established in this article.

The prediction model for buffer material is evaluated in two parts. The bentonite-water fraction is evaluated with empirical models. By modifying the DeVries and Campbell model (1985) for thermal conductivity of soils, the results fit well with the experiment data under different clay dry density and water content (Figure 1). The sand or crushed granite fraction of the buffer material is dealt with micro mechanical theory. By assigning the sand or crushed granite fraction as the discrete part and the bentonite-water fraction as the continuum part, the differential scheme or self-consistent scheme can be applied and the results agree with the experiment date well (Figure 2).

![Figure 1: The experimental data of Black Hill Bentonite at varied dry density and water content with the modified DeVries and Campbell model.](image-url)
The prediction model of thermal conductivity of sand-bentonite based buffer material

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Abstract

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Keywords: Buffer material, bentonite, thermal conductivity, micromechanics

1. Introduction

Buffer materials are used as engineered barriers in high level nuclear waste depository systems. The thermal conductivity of buffer material embedding the waste canisters has the impact on the maximum thermal load that can be applied in the canisters. In order to keep the maximum temperature in the buffer material below a certain value, the batch design of buffer material should be concerned (Börgesson, 1994).

The thermal properties of porous media are affected by a variety of parameters. According to Farouki(1986), heat conduction is mainly influenced by the composition, structure, packing, density, porosity, grain- and pore-size structure, as well as by contacts and binding effects. Previous researches
reveal that there are three major parameters that affect the thermal conductivity of bentonite-sand (crushed granite) buffer materials: the proportion of bentonite/sand (crushed granite), the dry density (or void ratio) of the bentonite, and the water content of bentonite. For many researches on thermal conductivity prediction models, pure empirical regression models, numerical models or semi-empirical models with theoretical basis are widely used for calculating and predicting for soils or rocks of various conditions. Due to the complexity of soils, these models are either complicate or with too many restrictions.

The development of thermal conductivity predicting model depends on the accuracy of original data from carefully conducted laboratory experiments. The methods for thermal conductivity measurement can be categorized, by applied theory, as steady state methods and transient methods. The steady state methods need to apply a stationary heat flow through the sample to create a constant thermal gradient in the specimen for a long time. This is a problem in partially water saturated materials since the gradient will cause moisture redistribution in the sample. Therefore, transient state methods are fairly common in soil testing for the short time of measurement. For bentonite-sand (crushed granite) buffer materials, thermal probe method is used in many researches. But the preparation of sample and the probe installation may cause too much uncertainty in experiment result. In this article, a modified thermal probe method based on ASTM D5334(2000) is developed, in order to investigate the change of thermal conductivity in a single buffer material due to the change in clay density. Thermal conductivity of samples with different water content and proportion of bentonite/sand (crushed granite) are measured with this method, and a model is proposed with the acquired data

2. Measuring Techniques

The method proposed in this article is called the continuous line source method. With this method, the thermal conductivity of bentonite-based buffer material can be measured with different clay dry density in a single sample. The concept is to put the thermal probe inside the buffer material sample while compaction. When a dry density is reached, the thermal conductivity is measured. With repeating the process, the relationship between thermal conductivity and clay dry density can be established. This method includes two systems – the compaction mold and the line-source measurement system.

2.1 The compaction mold

The schematic layout of the compaction mold is in Figure 1. The mold is composed by the following components.

The inner split mold The four-piece steel split mold forms a cubic space of 18 cm × 7 cm and 19 cm in height inside. Thermal hardening process is applied over the surface of the steel mold to archive a higher stiffness to resist the scratching from the compaction process. The outside shape of the split mold when assembled is a cylindrical cone with transition in diameter from 25.3 cm at bottom and 24.3 cm at the top.

The confining ring The ring is made of steel without thermal process. The outer diameter is 31.5 cm. The inner shape of the ring is fit with the split inner mold. The reason for the design is that it will be easier to remove the inner mold out the ring after the sample is compacted, and the mold would not slip out when the compaction force exerts a lateral force through the sample. The height of the ring is 18 cm, less then that of the inner split mold. When the ring is put on the inner split mold assembly, there is a space between the ring and the base plate. This is to ensure the confining of the ring on the inner split mold.

The compaction piston The cubic piston is made with steel and thermal hardening process is also applied on it. The dimension of the piston is 17.9 cm × 6.9 cm and 19 cm in height. A threaded rod is installed on top of it to connect to the loading frame.

The base plate The base plate is also made with thermal hardening processed steel. The base plate has a upper deck and a lower deck. Figure 2 shows the detail of the base plate. A hole is drilled on the upper deck, while a groove is made at the same place in the lower deck. The wire of the thermal probe goes through the hole to connect to the logging system and the power supply.

The loading system A servo-controlled loading system was used to apply the compaction loading.
During the compaction procedure, the applied loading and the displacement of piston were continuously recorded by a load cell and LVDT. The capacity of loading system is 1 MN. The maximum compaction pressure is around 100 MPa. The control method of loading can be force-controlled or displacement-controlled. The compaction method used in this research is uniaxial static compaction. By controlling the distance from the base plate and the piston, the volume of the sample is determined, and the clay dry density can be calculated.

The line-source method

The thermal probe or “needle” method is a rapid and convenient method for measuring thermal conductivity of soils in the laboratory or in-situ. The probe is inserted into the soil to be tested and being thin, should cause little disturbance. It consists of a heater producing thermal energy at a constant rate and a temperature sensing element (thermocouple or thermistor). The rate of rise in the temperature of the probe depends on the thermal conductivity of the surrounding medium.

The theory of the line-source method is based on the theory of the line heat source placed in a semi-finite, homogeneous and isotropic medium (Carslaw and Jaeger 1959). The testing equipment used in this research is based on ASTM D53340 - Standard test method for determination of thermal conductivity of soil and soft rock by thermal needle probe procedure. The apparatus is listed below:

**Thermal Needle Probe** The thermal probe, which is 15 cm long and 0.3 cm in diameter, consists of a nichrome heater wire and a T-type thermocouple (made of copper and constantan wire). The thermocouple is suitable for its durability and the temperature range at -200°C ~ 400°C. The heater and the thermocouple is placed in a stainless tube and magnesium oxide (MgO) powder is filled in the gaps. A hydrostatic pressure at 30000 PSI is applied on the probe to compress the tube and the fill to form the probe. Therefore, the strength of the thermal probe is suitable for the experiment configuration in this research. To protect the wires during compaction, a metal mesh tube with TEFLOm liner covers the connecting wire all the way through the hole on the base plate. The position of the probe in the sample is kept in the central by first putting half amount of the sample in the mold, and bending the flexible wire to lay the probe on the surface, and then cover with the other half of the sample. The resistance of the nichrome heater wire is checked with Ohm meter before and after each experiment.

![Fig. 1 The compaction mold](image1)

**Thermal Readout Unit** Agilent 34970A data acquisition/switch unit with HP 34901A 20-channel armature multiplexer is used for temperature logging with time. While using the T-type thermocouple with this system, an ice-bath should be used to create a known reference temperature (0°C) in order to prevent the internal junction error. The resistance of the nichrome heater and the voltage of the power supply is also calibrated and recorded with this system to ensure the quality of the experiment.
The calculation of thermal conductivity is described in the standard suggested by ASTM D5334.

2.3 The procedure

This method consists of the following assumptions.
1. The water inside the sample is incompressible.
2. The sand (crushed granite) particles in the sample are incompressible for the sake of its high stiffness comparing to clay.
3. The volume change of the sample is contributed by the compaction on clay only.

Tien and Wu (2003) described the relationship between clay dry density, volumetric ratio and weight ratio in the compaction process of the bentonite-sand (or crushed granite) buffer material.

With these assumptions, the clay dry density can be determined by the distance from the piston to the base plate.

The procedure for the continuous line-source method is described below.

Determine the clay dry density at final stage
The clay dry density is determined by the dry weight of the clay proportion in the sample and the volume formed by the inner split mold and the piston. The final stage of experiment means that the volume of the sample equals to 18cm × 7cm × 7cm. This prism shape of sample is valid for the geometry specification by ASTM D5334. The gross weight remains constant all over the experiment. Once the clay density at the final stage is determined, the gross weight of bentonite-sand (or crushed granite) powder that will spur into the mold is determined.

Determine the Levels of clay dry density that the measurement will be executed
After the final (maximum) clay dry density is determined, the other clay dry density, at which the thermal conductivity measurement will be conducted, can be decided. Then the positions of the piston can be also decided.

Pour the mixtures into the mold
Half of the powdery mixture of bentonite and sand (crushed granite) is put into the mold and tamped to form a flat surface. The flexible wire of the thermal probe is immersed in the sample near the mold (as shown in Figure 3). After tamping the first layer, the thermal probe is bended to lay on the surface and temporarily fixed on it. The other half of the powder is then poured onto it and tamped.

Apply the initial contact load
The piston is lowered to the surface of the sample, and then a contact load of 0.5kN is applied. The settlement of the sample at this sequence is large due to the loose powdery state of sample. The force applying rate is slow to allow the particles to be rearranged. The readings of load cell will be stable when the settlement of the sample stops.

The follow sequence can be repeated to perform thermal conductivity measurement at different clay dry density that is determined previously.

Compact to the designated clay dry density
Displacement-controlled loading is applied at a displacement rate of 0.1mm/min. After the piston reaches the designated position, the position is fixed for 30 minutes for the sample to be stabilized.

Thermal conductivity measurement
The measurement for thermal conductivity is now performed. First the data logging system start to record the temperature by the thermocouple at a period of 0.5 sec. Then the power supply is turn on to input the heat and the time is recorded as the start of measurement. The heating time for the measurement is 1200 sec. After the test, the sample needs to be cool down to room temperature for the next stage of measurement. The cooling time for the materials we used is 40 minutes, and this time interval can overlap with the compacting and stabilizing time at next stage. Therefore, once the heating is stopped, the piston starts to compact to the next stage of dry density, and followed by stabilization time.
These two sequences are repeated till the final stage of dry density is reached and the thermal conductivity measurement is conducted. After the test, the compaction force is released at a slow rate of 1kN/s. The sample is then taken out cut into slices to measure the water content for comparison with that before experiment. The electrical resistance of the nichrome heater wire is also examined to see if the heater is corrupted by the compaction procedure.

3. Materials

The materials for the bentonite-sand (crushed granite) buffer material in this research are discussed below.

3.1 Bentonite

MX-80 (Na-bentonite), from Wyoming, USA, is used in this study. It is also known as Black Hill bentonite (B.H. bentonite). The chemical composition and basic physical properties are shown in Table 1 and Table 2.

Table 1 Typical compositions of MX-80 bentonite (all entries are in the unit of %)

<table>
<thead>
<tr>
<th></th>
<th>SiO₂</th>
<th>Al₂O₃</th>
<th>Fe₂O₃</th>
<th>K₂O</th>
<th>CaO</th>
<th>Na₂O</th>
<th>MgO</th>
<th>TiO₂</th>
</tr>
</thead>
<tbody>
<tr>
<td>MX-80 bentonite</td>
<td>64.5</td>
<td>18.5</td>
<td>3.45</td>
<td>0.39</td>
<td>1.18</td>
<td>1.94</td>
<td>2.48</td>
<td>0.29</td>
</tr>
</tbody>
</table>

Table 2 Physical Index properties of MX-80 bentonite

<table>
<thead>
<tr>
<th></th>
<th>Gₛ (g/cm³)</th>
<th>w₀ (%)</th>
<th>Aₜ (%)</th>
<th>LL (%)</th>
<th>PL (%)</th>
<th>PI (%)</th>
</tr>
</thead>
<tbody>
<tr>
<td>MX-80 bentonite</td>
<td>2.65</td>
<td>9.69</td>
<td>6.2</td>
<td>497</td>
<td>33</td>
<td>344</td>
</tr>
</tbody>
</table>

Since we are trying to investigate the relationship between the thermal conductivity and the water content of clay fraction, the water content is adjust with following procedure:

**Moisture chamber curing** To acquire the bentonite with water content more then nature water content, the bentonite is poured over a 30cm × 40cm flat punched pan with non-woven textile liner. The thickness of the bentonite layer is 1 cm. The pan is covered with another sheet of non-woven textile and put into a moisture chamber with relative humidity (RH) at 100% and temperature at 38°C. The change of water content over time is shown in Figure 4. After this relationship is established, the bentonite sample with different water content can be prepared with putting it in the moisture chamber within corresponding time interval.

![Fig. 4](image_url) The change of water content of B.H. Bentonite over time in a moisture chamber

**Oven dry method** To acquire the bentonite with water content less then nature water content, the bentonite is poured over a 65cm × 100cm flat stainless steel pan. The thickness of the bentonite layer is 1 cm. The pan is put into an oven with constant temperature at 105°C. The change of water content over time is

![Fig. 5](image_url) The change of water content of B.H. Bentonite over time in an oven
shown in Figure 5. After this relationship is established, the bentonite sample with different water content can be also prepared with putting it in the oven within corresponding time interval.

The bentonite prepared with both methods is first sieved with a #100 sieve to remove the clotted blocks, and then stored in re-sealable plastic buckets for further use. The reason to use a re-sealable bucket is to retain the water content more efficiently. The water content should be checked before each test.

3.2 Sand (crushed granite)

The sand we used was obtained from crushed granite from Kinmen Island, Taiwan. The gradation of crushed granite is passing through #10 and retaining on #20. The specified gravity (Gs) and void ratio (eg) of crushed granite are 2.67g/cm$^3$ and 0.01, respectively. Table 3 shows the mechanical properties of Kinmen granite before crushing.

Table 3 Typical mechanical properties of Kinmen granite

<table>
<thead>
<tr>
<th>Mechanical properties</th>
<th>Unconfined strength (MPa)</th>
<th>Bulk modulus (GPa)</th>
<th>Poisson’s ratio</th>
</tr>
</thead>
<tbody>
<tr>
<td></td>
<td>147</td>
<td>21</td>
<td>0.25</td>
</tr>
</tbody>
</table>

3.3 The Mixtures for sample preparation

Since the thermal conductivity affected by three major factors: dry density of clay, water content of clay, and the bentonite/sand (crushed granite) proportion, this research focused on these parameters to investigate the relationship and establish the model for prediction.

Dry density The dry density for buffer material suggested by various researches is at a range of 16 kN/m$^3$ ~ 17 kN/m$^3$, while the backfill material is at 14 kN/m$^3$ ~15 kN/m$^3$ (JNC,1999)(Wardrop,1985). In this research, we take the benefit of the continuous line-source method to deal with the dry density issue. The designated dry density at which thermal conductivity is to be measured are 14 kN/m$^3$, 15 kN/m$^3$, 16 kN/m$^3$, 17 kN/m$^3$, and 18 kN/m$^3$. Since these measurements are conducted in one sample, the combination of the other two factor dominates the amount of samples.

Water content The water content of bentonite that prepared in this research are 0%, 3.84%, 8.13%, 12.73%, 16.12%, and 20.04%. These different water contents are measured right before executing the experiments.

Bentonite/sand (crushed granite) proportion The weight fraction of the crushed granite, $G_g = 0\%$, 12.5%, 25%, 37.5% and 50% is used.

For the traditional experiments, the amount of tests should be conducted is determined simply by multiplexing all the parameters, that is, $5 \times 6 \times 5 = 150$ samples must be prepared, compacted, and measured. For such amount of samples, the quality of experiment should be extremely careful to avoid the errors induced by the individuality of samples. With continuous line-source method, the amount of samples is reduced to $1 \times 6 \times 5 = 30$ samples. The parameters can be easily controlled and retained. Therefore, the data quality is much better.

4. Results of experiments

4.1 Pure Bentonite

The thermal conductivity of pure bentonite with different dry density and different water content are shown in Figure 6 and figure 7 with respect to each parameter.

Fig. 6 The thermal conductivity of B.H. Bentonite at varied dry density
thermal conductivity and water content, a sudden raise of thermal conductivity occurs at the water content range of 8% ~ 12%. This shows that the water not only contributes it high thermal conductivity to the bulk thermal conductivity, but also interacts with particles in clay to form a water bridge for smaller contact thermal resistance.

4.2 Bentonite/sand (crushed granite) mixture

The result of buffer material with different crushed granite fraction is shown in figure 8. Note that the weight fraction of granite has been changed to volumetric fraction of granite in order to meet the need for applying micromechanical models. The relationship of weight fraction and volumetric fraction of crushed granite in buffer material with different clay dry densities is listed in Table 4.

From the result, the thermal conductivity of mixture with low water content in bentonite increases significantly when the volumetric fracture of granite increases. With higher water content in bentonite, the difference of thermal conductivity between the bentonite and the granite become smaller, therefore, the increasing of volumetric fraction of crushed granite is not that effective.

5. Prediction Models

Lots of efforts on prediction models for thermal conductivity in soils had been done by many researchers. Most of the models rely on the fraction analysis of constituents in soils. A discussion of the validity of the models has been reviewed by Farouki (1986).

The prediction model for buffer material is more simple then ordinary soil for the simplicity in the constituents – clay, water, granite, and void. In this approach, we try to reduce the number of the constituents again. Buffer material is considered to be a statistically isotropic composite material. One phase is the bentonite with water and void, while the other is granite only. With this definition, two-phase micromechanical models can be applied. The approach is to establish the models for bentonite first. When the thermal conductivity of bentonite with certain condition is determined and the fraction of granite is set, the over all thermal conductivity is known by composite material theory.

5.1 Pure Bentonite

Campbell (1985) fitted the relationship between water content and thermal conductivity of sand, silt loam, and forest litter by the following equation (McInnes, 1981)
Fig. 8 The thermal conductivity of buffer material with different water content, clay dry density, and volumetric fraction of granite.
where $\theta_v$ is the volumetric water content, and $A$, $B$, $C$, $D$, and $E$ are coefficients that can related to soil properties. By regressions and curve fitting with data from DeVries (1963), the coefficients are listed below:

\[
A = 0.65 - 0.78\gamma_d + 0.60\gamma_d^2 \tag{2}
\]

\[
B = 1.06\gamma_d \tag{3}
\]

\[
C = 1 + 2.6/(mc^{0.5}) \tag{4}
\]

\[
D = 0.03 + 0.1\gamma_d^2 \tag{5}
\]

\[
E = 4 \tag{6}
\]

where $mc$ is the clay content.

By using same procedure proposed by Campbell with the experimental data from this research, we established the suitable coefficients for bentonite with following coefficients:

\[
A = 0.55 - 0.78\gamma_d + 0.60\gamma_d^2 \tag{7}
\]

\[
B = 0.259\gamma_d \tag{8}
\]

\[
C = 14.5 \tag{9}
\]

\[
D = 0.067 + 0.2\gamma_d^2 \tag{10}
\]

\[
E = 4.1 \tag{11}
\]

The prediction curve of the modified DeVries and Campbell model for bentonite is shown in Figure 9 with experimental data.

The comparison of the prediction value and the experimental data is shown in figure 10. The $R^2$ equals to 0.9847, and shows a good correlation.

5.2 Micromechanics models

A composite is statistically isotropic when its effective stress strain relation is independent of the choice of coordinate system. Examples include random mixture of two phases, matrix containing spherical type particles, random oriented elongated particles, and porous media (Hashin, 1983).

From the viewpoint of micromechanics, the bentonite-sand mixture can be regarded as a kind of composite materials. Bentonite can be treated as a matrix and the sand can be treated as an inclusion. The mixtures of bentonite-sand have near equal-dimension shape of inclusions and mixed randomly, thus can be regarded as a two-phase statistically isotropic composite material. By using the volumetric fraction and the thermal conductivity of each phase, the over all thermal conductivity can be calculated with micromechanics models.
The Self-Consistent-scheme and Differential scheme are used for thermal conductivity prediction (Nemat-Nasser, 1993) (Mclaughlin, 1977). The first step is to calculate the thermal conductivity of the phase of bentonite at a specified clay dry density and water content with modified DeVries and Campbell model. The thermal conductivity of phase of granite is decided with the data from Touloukian (1970) to be 1.69 W/mK. The next is to determine the volumetric fraction of each constituent. Finally the prediction is calculated with the two models. The results are listed in table 5. The correlations of the prediction with both methods are shown in figure 11.

With the combination of these two kinds of...
models, a new model concerning clay dry density, water content, and fraction of sand (crushed granite) simultaneously is prepared.

6. Conclusions

The method for measuring the thermal conductivity of buffer materials with different density, water content, and volumetric proportion of sand (crush granite) is established. The method is also suitable for powdery materials in which the relationship between thermal conductivity and the density need to be established. With this method, the amount of experiments due to different densities can be reduced, and the results of measurement keep a fair consistency.

Since the measurement of thermal conductivity is performed on a specimen under a compaction loading, a further study on the unloaded specimen with rebounded deformation (change in dry density) will be conducted.

From the data acquired, a model concerning about the three major factors affecting the thermal conductivity of buffer materials is prepared. The combination of a semi-empirical model and micromechanics models gives a good result. For other kind of candidate clay minerals or ballast, the procedure of establishing a suitable thermal conductivity prediction model can be done.

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