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EFFECT OF COMPACTION PRESSURE AND WATER CONTENT ON THE THERMAL CONDUCTIVITY OF SOME NATURAL CLAYS

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Abstract—This paper presents thermal conductivity data for highly compacted Ca-smectite, Na-smectite, illite, and palygorskite as a function of density (i.e., compaction pressure), water content, and temperature. All the clays behaved similarly: thermal conductivity increased directly with density and water content. Specifically, the thermal conductivity increased from 0.63 to 1.32 W/m·K as the dry density increased from 1.2 to 1.8 g/cm^3 (for a water content of 17%). An increase of water content from 6 to 17% resulted in an increase in thermal conductivity from 0.63 to 1.22 W/m·K (for a dry density of 1.6 g/cm^3). Differences from one clay to the other were less important. The thermal conductivity (in W/m·K) for constant conditions of 12% of water and a dry density of 1.6 g/cm^3 were: Ca-smectite 0.80, Na-smectite 0.74, palygorskite 0.71, and illite 0.69. Heating to 188°C produced only a 10% increase in the thermal conductivity.

Key Words—Compaction pressure, Illite, Palygorskite, Smectite, Thermal conductivity, Water content.

Résumé—Cet article présente des mesures de conductivité thermique effectuées sur des argiles hautement compactées—smectite-Ca, smectite-Na, illite, palygorskite—en fonction de la densité (c'est à dire de la pression de compaction), de la teneur en eau et de la temperature. Toutes les argiles étudiées ont le même comportement: la conductivité thermique augmente avec la densité et la teneur en eau. La conductivité thermique croit de 0,63 à 1,32 W/m·K quand la densité augmente de 1,2 à 1,8 g/cm^3 (pour une teneur en eau de 17%). Une variation de teneur en eau de 6 à 17% produit une augmentation de conductivité de 0,6 à 1,22 W/m·K (pour une densité sèche de 1,6 g/cm^3).

Les différences d'une argile à l'autre sont moins importantes: la conductivité en W/m·K pour une teneur en eau de 12% et une densité sèche de 1,6 g/cm^3 sont: 0,80 pour la smectite Ca, 0,74 pour la smectite Na, 0,71 pour la palygorskite et 0,69 pour l'illite. La température, jusqu'à 188°C, n'a qu'une faible influence sur la conductivité thermique (elle ne provoque qu'une augmentation de l'ordre de 10%).

INTRODUCTION

Efficient heat transfer is one of the main properties required for engineered barriers between vitrified nuclear wastes and the surrounding rocks. A study was therefore undertaken to determine the thermal conductivity of highly compacted clay materials that have been proposed for such barriers.

Kahr and Müller-Vonmoos (1982) determined the thermal conductivity of a Na-smectite from Wyoming and a Ca-smectite (trade name = Montigel) to range from 0.45 W/m·K (for a water content of 0 wt. % and a bulk density of 1.7 g/cm^3) to 1.34 W/m·K (for a water content of 14 wt. % and a density of 2.21 g/cm^3). They reported only a slight increase in conductivity with temperature. Pusch (1983) found that the thermal conductivity of the same Na-smectite increased from 0.8 to 1.5 W/m·K as the water content increased from 5 to 20%, at a bulk density of 2.1 g/cm^3. Radhakrishna (1984) pointed out the importance of water content and density on thermal conductivity and the nonsignificance of temperature. For dry bentonites he reported values of about 0.6 W/m·K at a bulk density of 1.6 g/cm^3. According to Tassoni (1980), the *in situ* thermal conductivity of Italian smectites was about 1.5 W/m·K. Lappin and Olsson (1979) noticed a decrease in thermal conductivity during shrinkage as clay dries.

The aim of this paper is to examine in detail the parameters that affect the thermal conductivity of highly compacted clays (i.e., water content, density, temperature, mineralogical type) and to estimate their relative importance. These data are of interest in the domains of radioactive waste disposal, soil science, and, more generally, heat transfer in porous media.

THERMAL CONDUCTIVITY MEASUREMENTS

Thermal conductivity of materials may be measured by static or dynamic methods (Touloukian, 1975). In the present investigation, static methods were not used because of the long time required to attain thermal equilibrium of the sample during which water may migrate within the clay. In contrast, dynamic methods generally require less than 5 min and are not compromised by water migration.

An instrument based on dynamic methods was used: the Quick Thermal Meter (QTM) developed by Showa

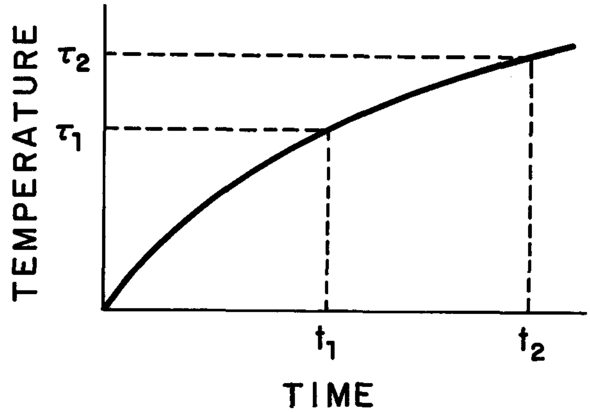

Figure 1. Rise of temperature of heating wire of probe of Quick Thermal Meter during a measurement. Thermal conductivity is calculated on basis of rate of temperature rise between times t_1 and t_2.

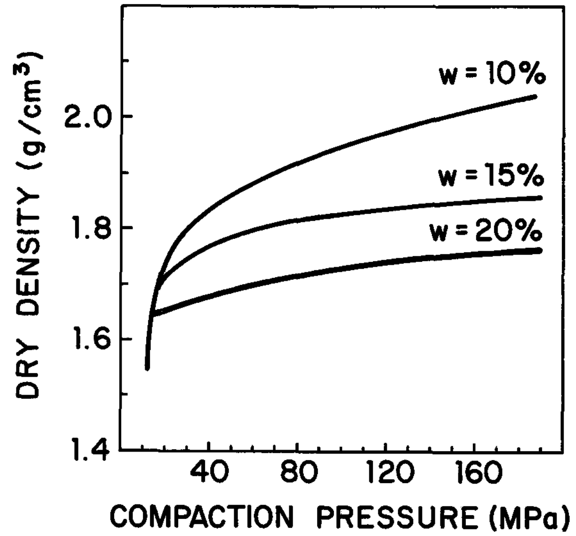

Figure 2. Density of compacted clay (Ca-smectite) having different water contents (w) as function of compaction pressure.

Denko. This instrument is based on the hot wire method (see Parrot and Stuckes, 1975). The measuring probe consists of a heating wire (10-cm length) and an attached thermocouple. During the measurement, electrical current flows from the QTM to the heating wire at a constant voltage. Because the measuring surface of the probe is kept in close contact with the clay, the temperature of the heating wire rises in proportion to the thermal conductivity of the clay, as shown in Figure 1. The thermal conductivity is calculated on the basis of the constants K and H of the probe and the rate of temperature rise, as shown in the formula established by Carslaw and Jaeger (1959) for a heating wire in a medium having cylindrical symmetry:

$$\lambda = K\frac{\log(t_2/t_1)}{\tau_2/\tau_1} + H$$

(t_1, t_2, τ_1, τ_2 are defined in Figure 1). The measurement accuracy is 0.02 W/m·K, as estimated from the accuracy and the reproducibility of measurements on the reference plates and on some clay samples.

For the measurement of thermal conductivity as a function of temperature, the probe and the block of clay were put in an oven (the probe may be used at temperatures ≤200°C).

MATERIALS

Four natural clays were studied: a Na-smectite from Wyoming, a Ca-smectite from the Paris Basin (France), an illite from central France, and a palygorskite from southeastern France. Mineralogical analyses of the whole clays were carried out by X-ray powder diffraction; microchemical analyses of the clay fractions were carried out with a Camebax electron microprobe. Analytical methods and results were developed by Coulon *et al.* (1987); the principal data are presented on Table 1.

Clay powders (mean particle size = 150 μm) were compacted in an isostatic press to obtain high density samples. As shown in Figure 2, the resultant density depended on compaction pressure and the water content of the clay. To obtain the desired water content, the clay powder was placed in an oven at a controlled temperature and humidity. After 1 wk, equilibrium is reached between the air humidity and water content of the clay. Figure 3 shows the relationship between the water content of the Ca-smectite and the relative humidity of the air.

Table 1. Mineralogical composition of the four studied clays.

			Clay (%)					
Material	SiO_2 (%)	Calcite (%)	Sm	Ill	Kaol	I/S	Paly	Associated Minerals
Ca-smectite	7	3	90		Tr	Tr		
Na-smectite	15	2	83					Feldspar
Illite	7	1		69	18	5		Feldspar
Palygorskite	10	6	Tr	Tr			86	

Sm = smectite; Ill = illite; Kaol = kaolinite; I/S = interstratified illite/smectite; Paly = palygorskite; Tr = trace.

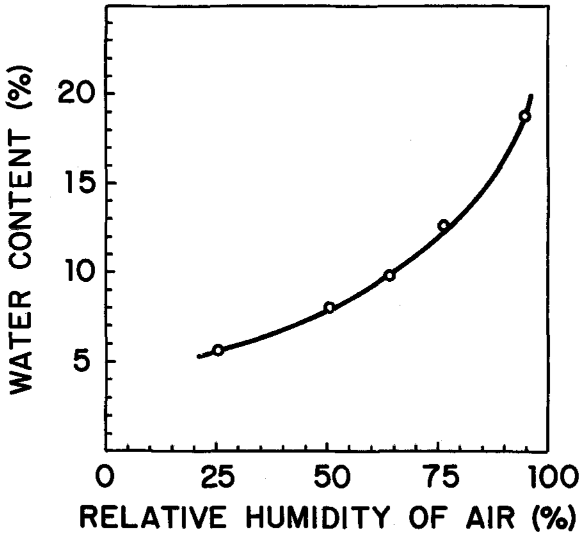

Figure 3. Water content of clay powder (Ca-smectite) as a function of relative humidity of air, after 1 wk in oven at controlled temperature and humidity of air.

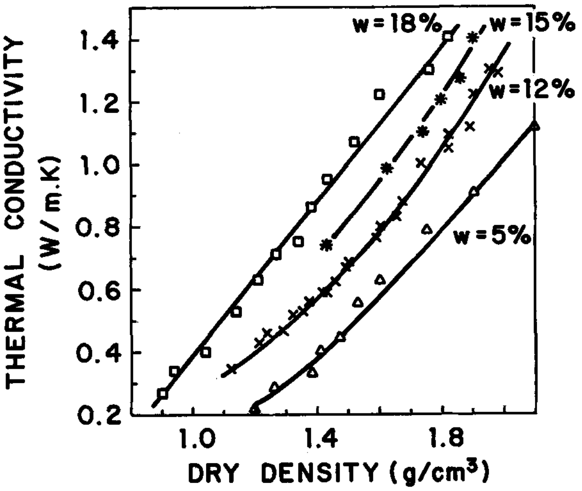

Figure 4. Thermal conductivity of Ca-smectite as function of dry density for several water contents (w) at ambient temperature.

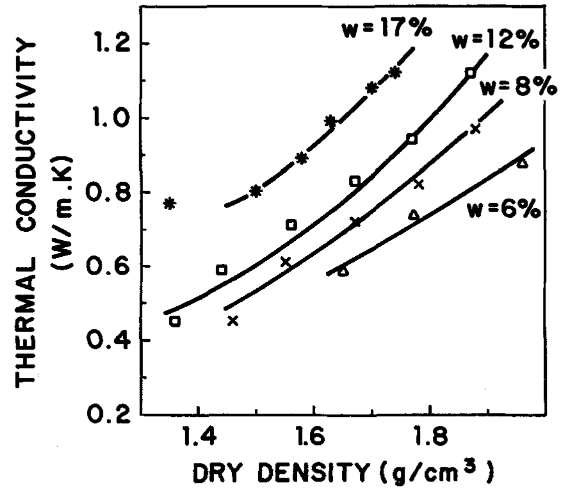

Figure 5. Thermal conductivity of Na-smectite as function of dry density for several water contents (w) at ambient temperature.

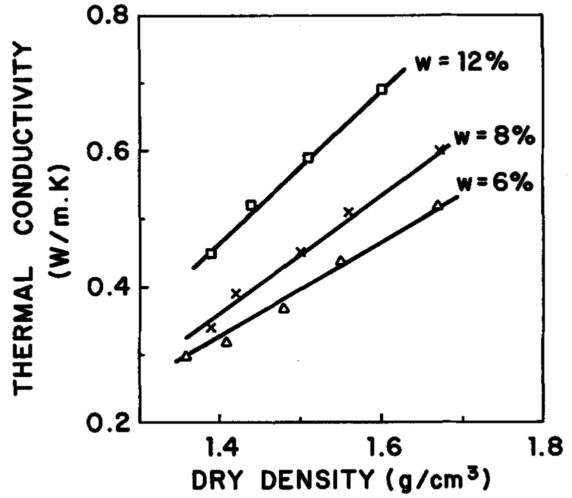

Figure 6. Thermal conductivity of palygorskite as function of dry density for several water contents (w) at ambient temperature.

RESULTS

Thermal conductivity data at ambient temperature are presented in Figures 4–7 as a function of dry density, ρ_D. Dry density is defined by the relationship:

$$\rho_D = \rho_H \frac{1}{1 + w/100},$$

where ρ_H is the bulk density and w is the water content (weight of water referred to the weight of dry material heated at 105°C for 24 hr). The thermal conductivity of the Ca-smectite at different temperatures is presented in Figure 8 (for a sample having a dry density of 1.99 g/cm^3 and a water content of 0%).

DISCUSSION AND CONCLUSIONS

As shown in Figures 4–7 thermal conductivity increased with density and water content. For example, the thermal conductivity of the Ca-smectite having a water content of 11% increased from 0.5 to 1.3 W/m·K as the density increased from 1.30 to 1.95 g/cm^3. The thermal conductivity of samples having a dry density of 1.6 g/cm^3 increased from 0.6 to 1.2 W/m·K as the water content increased from 5 to 18%.

These results can be explained by considering the respective values of the specific thermal conductivity

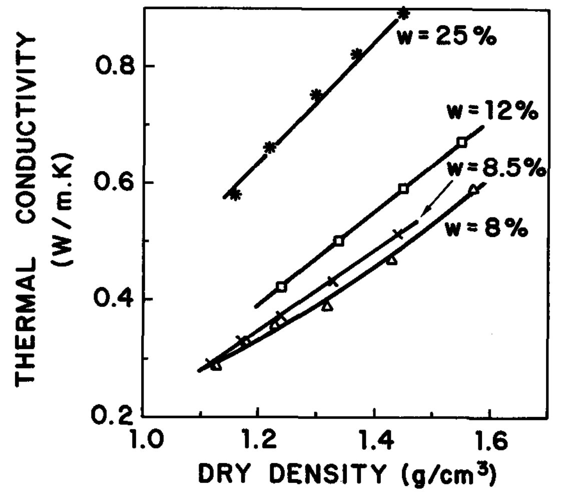

Figure 7. Thermal conductivity of illite as function of dry density for several water contents (w) at ambient temperature.

Table 2. Thermal conductivity of the four studied clays as a function of water content (w) and dry density (ρ_D) at ambient temperature.

w (%)	ρ_D (g/cm³)	Thermal conductivity (W/m·K) Ca-smectite	Na-smectite	Palygorskite	Illite
6	1.6	0.63	0.52	—	0.48
	1.8	0.83	0.74	—	—
	2.0	1.01	0.88	—	—
12	1.4	0.57	0.50	0.54	0.45
	1.6	0.80	0.74	0.71	0.69
	1.8	1.08	1.01	—	0.87
17	1.2	0.63		0.51	—
	1.4	0.86	0.79	0.69	—
	1.6	1.22	0.92	0.90	—
	1.8	1.32	1.20	—	—

of the three phases forming the clay material (solid, water, air): λ_{solid} = 2–3 W/m·K, λ_{water} = 0.6 W/m·K, and λ_{air} = 0.024 W/m·K. The thermal conductivity of air is about 25 times less than that of water; hence, if air is replaced by water in the clay (i.e., if the water content increases), the bulk thermal conductivity increases.

If dry density increases (at constant water content), the volume fraction of air decreases, and the volume fraction of solid increases. Consequently, the bulk thermal conductivity increases. Moreover, if dry density increases, solid particles move closer one to another, and the thermal contact resistance between clay particles decreases.

On the basis of the data in Table 2, the thermal conductivity of the studied clays having the same density and water content may be compared. The variation of thermal conductivity from one clay to the other is apparently less than 30%. These variations are smaller than those obtained if water content and density are allowed to vary. The four studied clays may be ranked as follows on the basis of increasing thermal conductivity:

illite < palygorskite < Na-smectite < Ca-smectite.

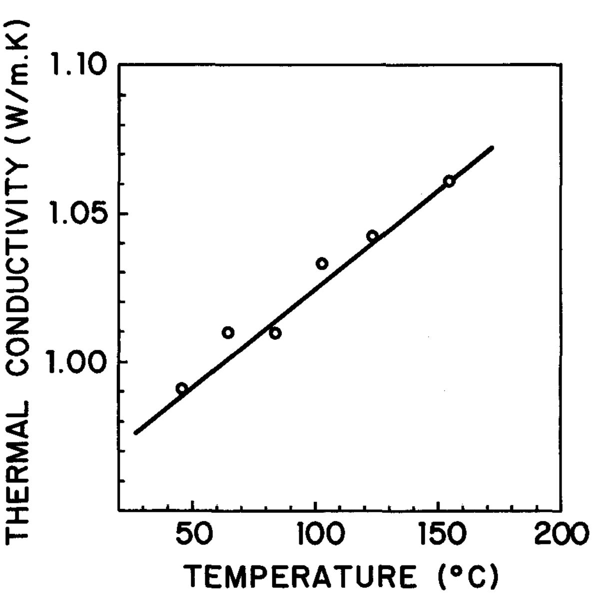

Figure 8. Thermal conductivity of dry sample of Ca-smectite as function of temperature (sample dry density = 1.99 g/cm³, water content = 0%).

Heating to 188°C apparently did not change the thermal conductivity significantly; an increase of about 10% was observed when temperature rose from 50° to 188°C. In this temperature range thermal conductivity of crystallized materials decreases with temperature, whereas for noncrystalline materials, it increases. The studied clays seem to have behaved more like noncrystalline amorphous materials, although they possess crystalline structures. In fact, the compacted clay appears to be a porous material in which heat was transported by gaseous phase movement, thereby increasing the apparent thermal conductivity.

In conclusion, the important parameters for heat transfer in clays appear to be water content and density; temperature and mineralogical type appeared to be of less importance.

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