The measurement of the thermal conductivity of solid aggregates using the transient plane source technique

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Abstract. In predicting the overall thermal conductivity of composite porous structures such as building materials or soils, the thermal conductivities of their solid components must be known in order to apply some theoretical models. Horai proposed a method to determine the thermal conductivity of solid particles. The material under study is first ground to a fine powder. This powder is then carefully saturated with a fluid and the thermal conductivity of this mixture is measured using the 'needle probe' method. Finally, the conductivity of the solid phase is evaluated using a mixing-law model, namely the mean of the so-called 'Hashin and Shtrikman bounds'. We have introduced a slightly different version of this method that uses the 'transient plane source' (TPS) technique recently developed by Gustafsson for simultaneously measuring both the thermal conductivity and the thermal diffusivity of solids or fluids. An adapted experimental device has been designed and our approach has been validated through measuring, at room temperature, the thermal conductivities of the well-known minerals quartz, calcite and kaolinite. Afterwards, while considering that our method had become fully operational, we studied the thermal conductivity of the solid aggregates of a light-weight, energy-saving concrete whose solid phase is a mixture of natural clay, hardened cement paste and wood aggregates in various proportions.

1. Introduction and basic principles

Determining the effective thermal conductivity of composite materials, such as building materials and soils, has been the subject of much research. This has turned out to be a difficult problem because the transfer property is a complex function of many other parameters, such as the thermal conductivities of each phase, their relative proportions, the size of the solid particles and the contact areas and distribution within the medium. Many models, based on different assumptions and with various degrees of realism, have been developed in order to predict the effective thermal conductivity of these heterogeneous systems: see, for example, Luikov *et al* (1968), Crane *et al* (1977), Vachon *et al* (1973), Dawson and Briggs (1981) and Jackson and Black (1983).

The main difficulty in using the corresponding analytical expressions of these models stems from the fact that these expressions generally require the knowledge of several parameters. In particular, the thermal conductivity of the solid phase has be estimated. To overcome this potential obstacle, Ki-Iti Horai (Horai and Simmons 1969, Horai 1971) developed a method to determine the thermal conductivity of minerals at ordinary temperature and pressure. In this method, the solid particles are first ground to a diameter of less than 80 μm and then mixed with a fluid, typically distilled water. The thermal conductivity of this saturated mixture is measured by the 'needle probe' or 'line-source' technique. The thermal conductivity of the solid particles is thus evaluated using the arithmetic mean $(\lambda_U + \lambda_L)/2$ of the so-called 'Hashin and Shtrikman bounds' λ_U and λ_L, which can be written as follows for two phases (Hashin and Shtrikman 1962):

$$\lambda_U = \lambda_S + \frac{\theta}{\frac{1}{\lambda_w - \lambda_S} + \frac{1-\theta}{3\lambda_S}} \tag{1}$$

$$\lambda_L = \lambda_w + \frac{1 - \theta}{\frac{1}{\lambda_S - \lambda_w} + \frac{\theta}{3\lambda_w}} \quad (2)$$

where λ_s is the conductivity of the solid phase to be determined and λ_w and θ are the thermal conductivity and volume fraction of water, respectively.

Horai and Baldridge (1972a, 1972b) demonstrated the applicability of this method to the measurement of thermal conductivities of rocks, which are commonly mixtures of different kinds of minerals. By using this method, Horai (1971) and Horai and Baldridge (1972a, 1972b) determined the thermal conductivity of 166 rock-forming minerals and 19 igneous rocks. In spite of this significant amount of data, there is still a lack of information concerning many natural and artificial materials, such as clay minerals, hardened cement paste, concrete and bricks.

More recently, Gustafsson (1990, 1991) developed a new method for measuring the thermal properties of materials, known as the 'transient plane source' (TPS) technique. This technique is based on recording the temperature rise of a plane source heating the surrounding material to be measured. One original feature of the corresponding sensor is the fact that the conducting pattern is used both as a heat source and as a temperature sensor. For this purpose, the temperature-measurement method employed utilizes the temperature dependence of the resistivity of the heating element's alloy. The initial electrical resistance R_0 of the TPS element is first balanced in a Wheatstone bridge and, during these measurements, the unbalanced voltage drop $\Delta E(t)$ is recorded versus the time t using a high-impedance digital voltmeter (figure 1). Basically, $\Delta E(t)$ has the following expression (Gustafsson 1991):

$$\Delta E(t) = \frac{R_S R_0^2}{R_0 + R_s} \frac{I_0^3 \alpha}{\pi^{3/2} a} \frac{D(\tau)}{\lambda} \quad (3)$$

where R_s is the standard resistance in the other branches of the Wheatstone bridge, I_0 is the heating current, α is the temperature coefficient of the TPS element, a is the outer radius of the heating element, λ is the thermal conductivity of the material and $D(\tau)$ is a function that can be tabulated for a particular TPS element as a function of the adimensional parameter $\tau = (\kappa t/a^2)^{1/2}$ where κ is the thermal diffusivity of the material. One can find additional details on the theoretical expression of this function as well as on the methods for determining λ and κ using (3) in Gustafsson's original papers.

In this paper, we will first describe the manner in which we have integrated the TPS technique into the original Horai method to determine the thermal conductivity of solid aggregates. Then, validation results for pure minerals will be presented. Finally, measurements on mixtures of clay, cement paste and wood aggregates will be reported and comparisons drawn between these measurements and values generated from the application of different models.

2. The experimental set-up and procedures

The solid particles were first crushed in a grinder (Controlab Company) to a diameter of less than 80 μm. The mineral powder was then mixed with distilled water. In order to
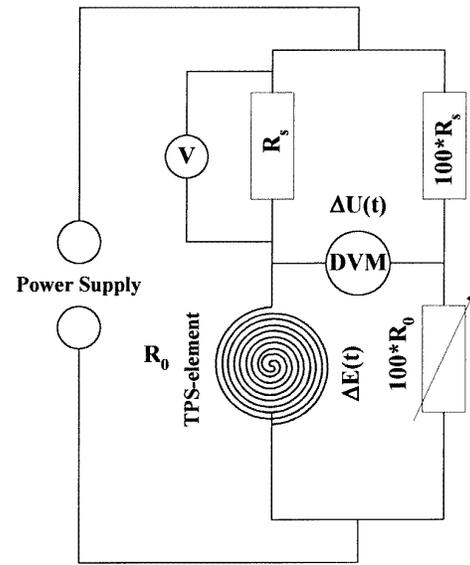

Figure 1. A schematic diagram of the bridge circuit.

obtain a homogeneous mixture consisting strictly of two phases, air bubbles were carefully removed. Three reasons for why small fine sizes are preferred (Horai 1971) can be advanced:

(i) to prevent convective heat transfer through the mixture,

(ii) to obtain a macroscopically homogeneous and isotropic mixture and

(iii) to allow one to consider the structure of the mixture to be totally random.

For the experimental determination of the thermal conductivity of the saturated mixture, we used a plastic cell of inner dimensions 7 cm × 7 cm × 6.5 cm, yielding a useful volume of about 300 cm^3. Half of the water-saturated mixture was first placed inside the cell. Then, the TPS element was positioned on top of this first layer while the rest of the mixture was carefully poured in. In this manner, the thermal contact between the mixture and the TPS element obtained was very good; however, great care had to be taken to ensure that the TPS element remained horizontal. The thickness Δ_e of each layer (approximately 30 mm) and, consequently, the overall dimensions of the cell were chosen to satisfy the 'probing-depth condition' defined by Gustafsson et al (1984), namely

$$\Delta_e \geq 1.42(\kappa t_{max})^{1/2} \quad (4)$$

where t_{max} is the total time of the transient recording which, in our case, is 200 s.

Successive weighings of the cell–empty, filled with the dry powder and filled with the water-saturated mixture– were performed in order to determine the weight fraction w of water in the mixture accurately. Calculating the corresponding volume fraction of water θ from w is quite straightforward using the following formula:

$$\theta = \frac{w}{w + (1 - w)\rho_s/\rho_w} \quad (5)$$

where ρ_w and ρ_s are the densities of water and of the solid particles, respectively. ρ_s can be measured by pycnometry, for example. An accurate determination of θ turns out to be very important. As a matter of fact, if we were to consider that the thermal conductivity λ_m of the mixture is given by

$$\lambda_m = \frac{\lambda_L(\lambda_s, \lambda_w, \theta) + \lambda_U(\lambda_s, \lambda_w, \theta)}{2} \qquad (6)$$

where the bounds λ_L and λ_U are evaluated by equations (1) and (2), respectively, a simple derivation would show that the sensitivity $\mathrm{d}\lambda_s/\mathrm{d}\theta$ of the λ_s determination to this parameter θ is very high; see figure 2. Furthermore, it should also be pointed out that the sensitivity to the measurement of λ_m is even higher! This means that the accuracy of the TPS measurement is also crucial. We will come back to this point later on.

The TPS element used for these measurements was composed of 10 μm thick nickel foils with a 30 μm thick Kapton insulation layer on each side, as designed by Gustafsson (1990). Before the start of the measurement procedure, the bridge was balanced using a small current so that the variation in resistance was negligible. After heating, the voltage drop over the TPS sensor was measured by using a HP 34401A digital multimeter. The temperature coefficient α of the sensor had previously been determined by measuring its resistance in the temperature range $0°$–$60\,°C$. A third-order polynomial has been developed to represent the $\alpha(T)$ evolution. For the 30 mm diameter element, α was about 4.6×10^{-3} K^{-1} at $20\,°C$.

The current supplied to the TPS element was monitored by measuring the voltage drop across a standard 10 Ω precision resistance using a precision multimeter (Schlumberger Solatron 7151). The dissipated power was adjusted according to the type of material so that the rise in the sensor's mean temperature did not exceed one kelvin at the end of a transient recording. We commonly used intensities in the range 150–250 mA. One channel of a data acquisition system (Schlumberger Solatron ORION 3530) is devoted to the control of the voltage applied by the power supply connected to the TPS sensor.

This entire system allowed measurements to be taken automatically at regular intervals. The corresponding digitized values were transferred for analysis to a personal computer through an HPIB interface.

To determine the mixture's thermophysical properties λ_m and κ_m, we fitted equation (3) to the measured $(t, \Delta E(t))$ points using a nonlinear estimation technique known as the 'gradient algorithm' (Press *et al* 1992). In determining the thermal conductivity of the saturated mixture, the voltage drop was deleted from the first measurement to eliminate the effect of the insulating layers (Gustafsson 1990, 1991). Lastly, the thermal conductivity of the solid particles λ_s was evaluated using the values of λ_m and θ found by solving equation (6) numerically with equations (1) and (2). Despite its apparent complexity, this whole procedure only took about 200 s on a standard personal computer.

Table 1. A comparison of thermal conductivity values for monomineralic aggregates.

Mineral	Line-source technique (LST) (W m^{-1} K^{-1})	Transient plane source technique (TPS) (W m^{-1} K^{-1})	Reference study (W m^{-1} K^{-1})
Quartz	7.71[a]	7.85	7.74[a]
Kaolinite	2.60[b]	2.60	2.62[b]
Calcite	3.60[a]	3.66	3.37[a]

[a] Data from Horai (1971).
[b] Data from Brigaud and Vasseur (1989).

3. The validation of the method

As a means of testing our method and evaluating its accuracy, measurements were first performed at ambient temperature ($20 \pm 0.5\,°C$) on pure quartz, calcite and kaolinite powders for which the thermal conductivity had already been determined by the line-source technique; see Horai (1971) and Brigaud and Vasseur (1989). Table 1 presents our results in comparison with those of Horai and of Brigaud and Vasseur, as well as in comparison with values found in the literature. It is obvious that all of these results are in very good agreement with one another. Our reported values represent the mean of three measurements taken on the same sample of each mineral; the standard deviation of these three tests was less than 2%, which can be considered satisfactory. The uncertainties in the θ and λ_m determinations are estimated to be less than $\pm 2\%$ and $\pm 2\%$, respectively. In propagating these uncertainties in our λ_s estimation procedure with the corresponding sensitivities (figure 2), we can state that the overall accuracy of our method was quite close to $\pm 5\%$ for these cases.

4. Measurements of the thermal conductivity of wood–light-weight-concrete aggregates

We are interested in the study of a new building material with improved thermal performance obtained by the incorporation of wood particles. This material is basically a 'light-weight concrete'–its binder is effectively a hydrated cement paste and its density of 500–1300 kg m^{-3} is much lower than that of standard concrete ($\rho = 2400$ kg m^{-3}), but its formulation has also been designed for recycling clayey waste from quarries.

We have undertaken a comprehensive study of the various factors that can influence the thermal conductivity of this material (Bouguerra, 1997). In this framework, using the estimation models mentioned in section 1, we naturally had to determine the thermal conductivity of the solid phase of this material, which is obviously highly heterogeneous. For this purpose, we proceeded in two steps.

First, using the method presented in section 2, measurements on water-saturated mixtures of natural clay, crushed hardened cement paste (HCP) and wood aggregates taken alone were performed. The corresponding results are presented in table 2.
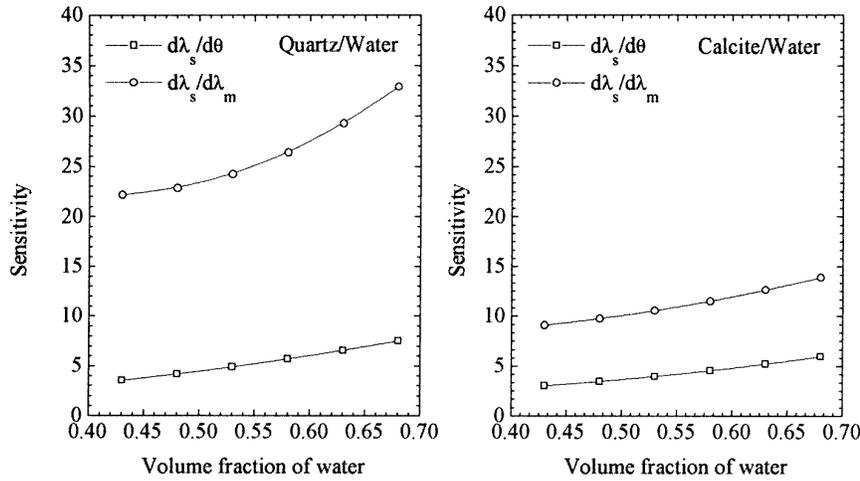
Figure 2. The sensitivities of the thermal conductivities of the solid particles estimated using Ki-Iti Horai's method of thermal conductivity measurement for a mixture and the volume fraction of water.

Table 2. Thermal conductivity measurements for natural clay, hardened cement paste and wood aggregates using the TPS method.

Material	Density	Experiment	Thermal conductivity (W m^{-1} K^{-1})
Natural clay	2.65	1	3.45
		2	3.40
		3	3.48
		Mean value	3.44
		SD (%)	0.44
Hardened cement paste	2.10	1	2.83
		2	2.85
		3	2.86
		Mean value	2.85
		SD (%)	0.54
Wood aggregates	1.52	1	1.15
		2	1.14
		3	1.12
		Mean value	1.14
		SD (%)	1.34

SD = Standard deviation.

Then, we developed water-saturated mixtures with powders from our various types of 'wood–light-weight concrete'. Using these heterogeneous mixtures incorporating powdered clay, HCP and wood in the proportions of our formulations, we measured with our method and always at room temperature the corresponding mean solid particle conductivities λ_s; see figure 3. The ratio of the mass of cement to the combined mass of clay and wood was held constant (at $\frac{1}{3}$) and increasing amounts of wood were tested. The proportion of wood aggregates was characterized by the ratio of the mass of wood to the combined mass of wood and clay, calculated with respect to the raw materials.

To analyse these results, we also tried to estimate λ_s by other models utilizing both the individual thermal conductivities measured previously (table 2) and the
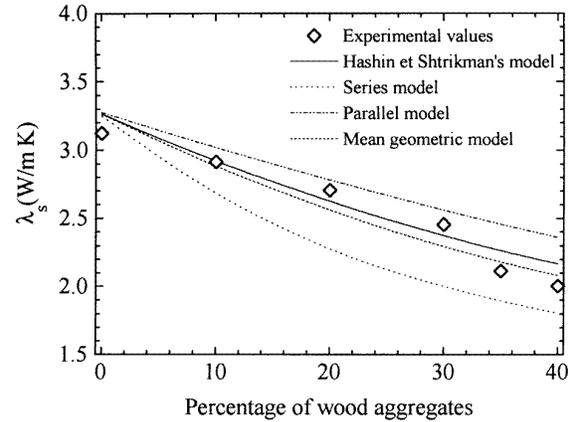

Figure 3. A plot of the thermal conductivity of clay–cement–wood aggregates in solid particles versus the percentage of wood aggregates.

corresponding proportion θ_i for each solid component i with $i = 1$, 2 and 3 for HCP, clay and wood, respectively.

The models considered were:

(i) a form of the Hashin and Shtrikman bounds generalized to n phases (Horai 1971):

$$\lambda_U = \lambda_{max} + \frac{A_{max}}{1 - \alpha_{max} A_{max}} \qquad \lambda_L = \lambda_{min} + \frac{A_{min}}{1 - \alpha_{min} A_{min}}$$

$$\lambda_{max} = \max(\lambda_1 \ldots \lambda_n) \qquad \lambda_{min} = \min(\lambda_1 \ldots \lambda_n)$$

$$\alpha_{max} + (3\lambda_{max})^{-1} \qquad \alpha_{min} = (3\lambda_{min})^{-1}$$

$$A_{min} = \sum_{i_{k_i=k_{min}}} \theta_i[(\lambda_i - \lambda_{min})^{-1} + \alpha_{min}]^{-1}$$

$$A_{max} = \sum_{i_{k_i=k_{max}}} \theta_i[(\lambda_i - \lambda_{max})^{-1} + \alpha_{max}]^{-1} \qquad (7)$$

(ii) the 'geometric mean' model (Dawson and Briggs 1981):

$$\lambda_S = \prod_{i=1}^{n} \lambda_i^{\theta_i} \qquad (8)$$

(iii) the well-known 'Wiener bounds' (Birch and Clark 1940a); for which the lower bound (serial) is

$$\lambda_S = \sum_{i=1}^{n} \left(\frac{\theta_i}{\lambda_i} \right)^{-1} \tag{9}$$

and the upper bound (parallel) is

$$\lambda_S = \sum_{i=1}^{n} \theta_i \lambda_i. \tag{10}$$

It can be shown from figure 3 that both (7) and (8) provide good estimates of λ_s. *A posteriori*, this justifies once again the use of the mean of the Hashin and Shtrickman bounds in our λ_s measurement method and indicates that a 'geometric mean' model would probably have been suitable. From another standpoint, this also serves to demonstrate that both of these models can be used to predict the solid-phase conductivity of such materials. The Wiener bounds (9) and (10) are only useful in setting the interval within which the value of λ_s should be found.

5. Conclusion

In this paper, we have described a method for measuring the thermal conductivity of solid aggregates. The experiments conducted on known minerals have demonstrated the applicability of the transient plane source (TPS) technique in combination with the Horai method designed for this purpose. We have used this modified method to determine the thermal conductivity of hydrated cement pastes, clayey waste and wood aggregates, all of which are components of a light-weight concrete being studied in our laboratory. Additional measurements carried out on mixtures of these aggregates, with increasing percentages of wood, were in very good agreement with the theoretical predictions of the *n*-phase 'Hashin and Shtrikman' and 'geometric mean' models.

The main advantages of using the TPS technique are, first, that it allows simultaneous measurement of the thermal conductivity and thermal diffusivity from only one transient recording and, second, that the experiments are performed with extremely small temperature perturbations of the materials. We now intend to capitalize on these advantages in order to extend this method to the determination of the thermal diffusivities of solid minerals and thermal properties of liquids.

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