Measurement of thermal conductivity of granular materials over a wide range of temperatures. Comparison with theoretical models.

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Abstract. This study is devoted to the measurement of thermal conductivity of granular materials. The materials studied are foundry sand and steel shot. These measurements are performed at high temperature (up to 640°C) by an unsteady state method using a cartridge heater as heating element. Then, experimental results are faced with three analytical models giving the effective thermal conductivity of granular materials. The thermal effusivity of these granular materials is also estimated.

1. Introduction
Thermal data for granular materials over a wide range of temperatures, including thermal conductivity, are not common in literature. However, these data become critical when one wishes to model and simulate thermal phenomena in industrial processes with high levels of temperature.

Experimental methods developed for the measurement of these properties can be subdivided into two main families: steady state methods (guarded hot plate, radial heat flow, ...) and unsteady methods (hot plane, hot wire, ...) [1-2]. The choice of a method is based on several parameters such as the nature of the studied material (granular, porous or solid, insulating or conducting) or the simplicity of implementation [1].

Steady state methods are particularly suitable for the characterization of insulating materials. These methods make the assumption of a material under stable thermal gradient subjected to a steady heat flow over time. The studied material is instrumented at various points and Fourier's law is used for analysis. Concerning unsteady methods, their use is widespread as these methods are suitable for all types of material. They are based on the exploitation of the temperature response of the material during the transient.

The aim of the present study is to measure the thermal conductivity and bulk density of two granular materials (foundry sand and steel shot) in order to semi-empirically determine their thermal effusivity. Measurements were carried out over a wide range of temperatures between room temperature and 640°C. In order to reach this goal, an unsteady state method was used. This method,
using a cartridge heater as heating element, is similar to the hot wire method. Then, these measurements are faced with three analytical models (Maxwell [3], Frey [4], Kunii & Smith [5]) for calculating the effective thermal conductivity of granular materials and deduce their thermal effusivity.

2. Experimental method

The hot wire method is generally used for liquids and gases but can also be applied to solid materials. The method involves immersing a wire, as thin as possible, in the material to be studied. Electric power is then applied to this wire (U=50.0V; I=0.281A), which is heated by Joule effect. A linear heat flux is generated in the surrounding material. In this method, heat transfer is assumed to occur only by conduction.

Let define: \( \Delta T(r,t) = T(r,t) - T_0 \), with \( T_0 \) : a reference temperature. For a wire of radius \( r_0 \), the temperature difference at the surface of the wire is given by

\[ \Delta T(r_0,t) = \frac{Q}{4\pi r_0^2} \ln(t) + K \]

By plotting the temperature difference \( \Delta T(r_0,t) \) as a function of \( \ln(t) \), one will get a straight line whose slope is directly related to the thermal conductivity of the studied material.

In the present work, experiments will be conducted by replacing the hot wire by a cartridge heater as heating element (Figure 1). This results in a similar method that needs to take into account the radius \( R \) of the heater. Conditions are such that the cartridge can be considered as a thermally thin body (diameter of 6.5mm vs a length of 110mm) at temperature \( T_1(t) \). The temperature of the cartridge is measured by a thermocouple. In addition, a second thermocouple is placed in the studied granular material at a distance of 5 mm from the cartridge in order to measure the granular material temperature. The first thermocouple (\( T_1(t) \)) is used to estimate \( mC_p \) value of the cartridge and the thermal conductivity of the material. The second thermocouple (\( T_2(t) \)) is used to estimate the distance \( r \) between this thermocouple and the cartridge, and the thermal conductivity of the material. Then, we check that the two values of thermal conductivity of the material estimated are the same. The volume of the granular material is around 600 mL and the measurements last about 400s.

![Figure 1. Sketch of the experimental method](image)

The cartridge is thus considered as a thermally thin body at temperature \( T_1(t) \) and the granular material is considered as a semi-infinite medium at temperature \( T_2(r,t) \). The equations to be solved are firstly that of the cartridge:

\[ mC_p \frac{dT_1(t)}{dt} = Q - \varphi \cdot S_c \]

(1)
wherein \( Q \) (in W) is the electric power dissipated and \( \varphi \), the surface heat flux (in W.m\(^{-2}\)) transmitted to the granular material, and, secondly, the 1D radial diffusion equation of heat in the sand:

\[
\frac{\partial^2 T_2(r,t)}{\partial r^2} + \frac{1}{r} \frac{\partial T_2(r,t)}{\partial r} - \frac{1}{a_s} \frac{\partial T_2(r,t)}{\partial t} = 0 \tag{2}
\]

Boundary and initial conditions are:

- \( r = R \) and \( t > 0 \): \( T_1(t) = T_2(R,t) \) \( \tag{3} \)
- \( r = R \) and \( t > 0 \): \( \varphi, S_c = -k_s \frac{\partial T_2(r,t)}{\partial r} \times 2\pi \times R \times lg \) \( \tag{4} \)

where \( lg \) is the heated length of the cartridge heater.

- \( r \to \infty, T_2(\infty,t) \) is finite \( \tag{5} \)
- \( t = 0 \): \( T_1(0) = T_2(r,0) = T_0 \) (Reference temperature) \( \tag{6} \)

This equation is solved in the Laplace domain. The temperature is then expressed as a function of the Laplace variable \( p \): \( T_2(r,p) \). Equation (2) thus admits as solution:

\[
T_2(r,p) = AI_0(\alpha r) + BK_0(\alpha r) \quad \text{with} \quad \alpha = \sqrt{\frac{p}{a_s}} \tag{7}
\]

As we consider a semi-infinite granular medium, condition (5) gives \( A = 0 \) and equation (7) becomes:

\[
T_2(r,p) = BK_0(\alpha r) \tag{8}
\]

Using Equation 4 gives:

\[
mCp \frac{dT_1(t)}{dt} = Q + k_s \frac{\partial T_2(r,t)}{\partial r} \times 2\pi \times R \times lg \tag{9}
\]

Solving in the Laplace domain, we obtain:

\[
\frac{Q/p + k_s \times Sc[-\alpha BK_1(\alpha R)]}{mCp \times p} = BK_0(\alpha R) \tag{10}
\]

Boundary condition (3) leads to:

\[
\frac{Q/p + k_s \times Sc[-\alpha BK_1(\alpha R)]}{mCp \times p} = BK_0(\alpha R) \tag{11}
\]

Therefore, the constant \( B \) is given by:

\[
B = \frac{Q/p}{mCp \times p \times K_0(\alpha R) + k_s \times Sc \times K_1(\alpha R)} \tag{12}
\]
Finally, the cartridge temperature $T_1(p)$ and the granular material temperature $T_2(r, p)$ in the Laplace domain write:

$$T_1(p) = \frac{Q}{p} \left[ \frac{K_0(\alpha R)}{mC_p \times p \times K_0(\alpha R) + \alpha k_s \times Sc \times K_1(\alpha R)} \right]$$ (13)

and

$$T_2(r, p) = \frac{Q/p}{mC_p \times p \times K_0(\alpha R) + \alpha k_s \times Sc \times K_1(\alpha R)} K_0(\alpha r)$$ (14)

Temperatures in the real domain are obtained using Stehfest’s algorithm [6] (equation 15):

$$f(t) \cong \frac{\ln 2}{t} \sum_{j=1}^{20} V_j F\left(\frac{j \ln 2}{t}\right)$$ (15)

This model calculates the thermal response of the studied material. Then, a parameter identification procedure, based on a Gauss-Newton algorithm, has been set up to retrieve the thermal conductivity of the studied material, the “$mC_p$” value of the cartridge heater and the effective distance between the cartridge and the thermocouple placed in the granular material.

3. Analytical models

In order to feed the databases on material properties for models and numerical simulations, it is advantageous to have a theoretical or empirical model to estimate the apparent thermal conductivity of a granular material. The relatively complex structure of this type of material (seen as an empty set filled with solid grains) involves a certain difficulty in the measurements. Indeed, the effective thermal conductivity is highly sensitive to the particular material porosity (degree of compaction), moisture (change of interstitial fluid) and the morphology of the grains. Nevertheless, assuming that the experimental procedure is adequately controlled, one may consider that these parameters remain constant from one measurement to another. Here are described three models that will be faced with experimental measurements obtained with the cartridge heater.

3.1. Maxwell model [3]

Many models are derived from Maxwell’s scheme, such as the Hamilton & Crosser [7] or the Hashin & Shtrikman [8] models. This type of model assumes a discontinuous solid phase, that is to say, suspended in the fluid phase (gas or liquid). Maxwell’s model is particularly suitable for environments with regular stacking of particles.

Therefore, the heat flow ($q$) through such a medium can be written:

$$q = \epsilon q_f + \alpha q_s$$

with $\epsilon$, porosity of the medium; $q_f$, heat flow in the fluid phase; $q_s$, heat flow in the solid phase and $\alpha = (1-\epsilon)$.

The relative density of a granular medium is expressed as the ratio between the effective density of the granular material and the density of the “pure” solid material. It can be written:

$$\alpha = \frac{\rho_{eff}}{\rho_{solid}} = 1 - \epsilon$$ (16)
By expressing the average heat flux as a function of the effective thermal conductivity of the granular medium studied, we obtain:

\[
k_{\text{eff}} = \frac{\varepsilon k_f (2k_f + k_s) + 3\varepsilon k_s k_f}{\varepsilon (2k_f + k_s) + 3\varepsilon k_f}
\]

(17)

with \( k_{\text{eff}} \): effective thermal conductivity of the granular medium; \( k_s \), thermal conductivity of the solid phase; \( k_f \), thermal conductivity of the fluid phase.

It is therefore important to know the thermal conductivities, both of the solid and the fluid phase, and their variation with temperature. Accurate measurement of the bulk density is also necessary to determine the porosity of the granular medium.

3.2 Frey model [4]

This model does not derive from the Maxwell model. It assumes a periodic medium. The effective thermal conductivity is thus determined by discretizing the medium in a series of basic cells. Then, two approaches are possible:

- Electrical analogy.
- Resolution of the heat equation in each basic cell. The medium studied in this case is modeled by a series of resistors in parallel or in series.

The effective thermal conductivity writes:

\[
k_{\text{eff}} = k_f \left[ \frac{1 - \alpha^{1/3} + \alpha}{1 - \alpha^{1/3} + \frac{k_f}{k_s} \alpha^{1/3}} \right] \frac{k_f}{k_s} (1 - \alpha^{1/3} - \alpha)
\]

(18)

3.3 Kunii & Smith model [5]

Unlike previously developed models, the model of Kunii and Smith considers the heat transfer through the contact points between solid particles. This is a contact model. In their model, Kunii and Smith consider a packing of spheres of equal diameter and assume that the heat transfer occurs at the contact points in the fluid and the solid.

The apparent thermal conductivity is given by:

\[
k_{\text{eff}} = k_f \left[ \varepsilon + \frac{\alpha \beta}{k_f} \right]
\]

(19)

with : \( \beta = \frac{\Delta L}{D_p} = \frac{2}{3} ; \gamma = \frac{l_s}{D_p} = 1 ; \varphi = 0.058 \)

where \( \varphi \) is a function of the porosity and of the ratio \( k_s/k_f \). In the present case, \( \varphi \) is empirically set to 0.058 to fit the sand measurements. \( l_s \) is the thickness of a cylinder having the same volume as the solid sphere. \( D_p \) is the diameter of the spherical particle and \( \Delta L \) is the centre-to-centre distance between two solid particles. For densely packed spherical particles, \( \beta \) is set to 0.895.
4. Results and discussion

4.1 Thermal conductivity

Two types of granular materials, foundry sand and steel shot, were characterized over a wide temperature range. Initially, averaged values of thermal conductivity between room temperature and 500°C were obtained. In order to refine these values, the same measurements were repeated but with "temperature plateaux", thus providing average values of thermal conductivity on narrower temperature ranges and improving the accuracy. These measurements were carried out between room temperature and 640°C and provide a first trend of the variation of the thermal conductivity with temperature.

Effective thermal conductivities deduced from the experiments and the three analytical models are plotted in Figure 2 for foundry sand and in Figure 3 for steel shot. Measurements carried out by "temperature plateaux" allowed obtaining an average value of thermal conductivity on relatively narrow temperature ranges (100-150°C). These experimental values can then be compared to analytical models.

For foundry sand, the experimentally determined effective thermal conductivities are ranging from 0.3 W.m\(^{-1}\).K\(^{-1}\) at 100°C to 0.415 W.m\(^{-1}\).K\(^{-1}\) at 500°C. Concerning steel shot, experimental measurements give an effective thermal conductivity ranging from 0.483 W.m\(^{-1}\).K\(^{-1}\) at 70°C to 0.778 W.m\(^{-1}\).K\(^{-1}\) at 640°C.

The uncertainty in the estimated thermal conductivities and effusivities is primarily due to the uncertainty in the measurement of the electrical current and tension applied as well as in the effective density of the granular materials. The maximum uncertainty in the effective thermal conductivities and thermal effusivities are estimated at 3.5% and 1% respectively.

4.2 Thermal effusivity

The thermal effusivities \((b = \sqrt{k\rho C_p})\) of foundry sand and steel shot can be estimated from the effective thermal conductivities given in section 4.1 and the measurement of the effective (or bulk) density \((\rho_{\text{eff}})\) of the granular materials. Properties of the two studied granular materials are given in Table 1. As a first approximation, one can consider \(C_{\text{p,eff}} \approx C_{\text{p,solid}}\) as \(\rho_{\text{solid}} >> \rho_{\text{fluid}}\) in the present study. Thermal effusivities deduced from the experiments and the three theoretical models are plotted in Figure 4 for foundry sand and in Figure 5 for steel shot.
Granular materials properties

| Material     | $\rho_{\text{solid}}$ (kg.m$^{-3}$) | $\rho_{\text{eff}}$ (kg.m$^{-3}$) | $C_{p_{\text{solid}}}$ (J.kg$^{-1}$.K$^{-1}$) | Porosity ($\varepsilon$) | $\alpha$ = $\frac{\rho_{\text{eff}}}{\rho_{\text{solid}}}$ |
|--------------|-----------------------------------|-----------------------------------|-----------------------------------------------|--------------------------|-----------------------------------------------|
| Steel shot   | 7200                              | 4180                              | 450                                           | 0,419                    | 0,581                                          |
| Sand         | 2200                              | 1480                              | 1000                                          | 0,327                    | 0,673                                          |

For foundry sand, the experimentally determined thermal effusivities are ranging from 668J.K$^{-1}$m$^{-2}$.s$^{1/2}$ at 70°C to 785J.K$^{-1}$m$^{-2}$.s$^{1/2}$ at 500 °C, with an effective density measured at 1480 kg.m$^{-3}$. For steel shot, the experimentally determined thermal effusivity is ranging from 953J.K$^{-1}$m$^{-2}$.s$^{1/2}$ at 70°C to 1209J.K$^{-1}$m$^{-2}$.s$^{1/2}$ at 640 °C, with an effective density measured at 4180 kg.m$^{-3}$.

Despite a lower specific heat, the better thermal effusivity observed for steel shot (+40% compared to foundry sand) can essentially be attributed to a higher effective density as well as higher effective thermal conductivities.

Discussion

In both cases, one may note that the theoretical models allow a partial description of the variation of apparent thermal conductivity. The increase in thermal conductivity with temperature is found, but with a steeper slope observed for the theoretical models. These models also take into account the impact of the interstitial fluid (air). Indeed, the relative difference in thermal effusivity between the two materials is less important in the granular state. Finally, one may also note that theoretical models better describe the variation of the apparent thermal conductivity in the case of foundry sand than in the case of steel shot. This difference may be explained by the difference in grain morphology. Although the averaged grain size is about the same for foundry sand and steel shot, the morphology and the particle size distribution change. Unlike steel shot grains which are spherical in shape, foundry sand is a granular material whose grains (crushed) have a random angular geometry. Therefore, the conductive heat transfer will preferentially occur by contact points between grains for steel shot, whereas it will occur through surfaces rather than points in the case of foundry sand.

Conclusion

The effective thermal conductivity of two granular materials (foundry sand and steel shot) was measured over a wide range of temperatures. An unsteady state method using a cartridge heater as heating element was used and associated with an inverse method model to identify the effective
thermal conductivity and deduce their thermal effusivity. The increase in thermal conductivity with
temperature was confirmed, but with a steeper slope observed for the theoretical models. It is however
important to note the difficulty of these measurements and precautions to be taken when analyzing
results. Indeed, the measured effective thermal conductivity values are highly dependent on the
interstitial fluid (here air), i.e. on the porosity of the medium. This porosity will depend on the degree
of compaction of the material. In fact, the effective thermal conductivity of a granular medium will
increase by decreasing the degree of porosity.

The differences observed between theoretical models and experimental values of effective thermal
conductivity of steel shot (differences that we do not find with sand) tend to highlight the importance
of grain morphology, a parameter that is not included in the three theoretical models presented in this
study. To further this study, the investigation of the influence of different morphologies and different
grain sizes for the same base material (e.g., sand) could be considered.

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