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Proceedings of the 5th International Conference on Inverse Problems in Engineering: Theory and Practice, Cambridge, UK, 11-15th July 2005

EXPERIMENTAL ESTIMATION OF THERMAL DISPERSION COEFFICIENTS IN GRANULAR MEDIA THROUGH WHICH A GAS IS FLOWING: POROUS VERSUS NON POROUS GRAIN

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Abstract - A technique for estimating the thermal dispersion coefficients in a granular medium (packed bed of monodisperse glass beads), originally developed for water as a flowing fluid, has been adapted for an air flow. Thermocouples in the downstream neighbourhood of a line heat source measure the temperature response to a step power input. The estimation of the different parameters of the model (one-temperature model) is made by least squares minimization of the temperature residuals term that is augmented by a regularization term based on the prior approximate knowledge of the thermocouple locations. It is shown, both theoretically and experimentally that, contrary to the water case, both dispersion coefficients (longitudinal and transverse) as well as the Darcy velocity, can be reached for air as a working fluid. The same technique has been tested for solid grains with internal micro-porosity. The first experimental temperature responses do not obey the previous model. The presence of adsorbed water in micro-pores of the grains, whose evaporation is caused by the decrease of the relative humidity of air, constitutes an additional negative heat source term. An experimental work, where ambient air is replaced by dry nitrogen, allows the preliminary drying of the grains. Correction of the strong temperature drifts caused by initial non-equilibrium, yields temperature signals that can be inverted with satisfactory residuals.

1. INTRODUCTION

Thermal dispersion, that is heat transfer in a porous medium through which a fluid is flowing, occurs in many natural situations or industrial applications. In the case of process engineering, modelling of this phenomenon is very important for controlling temperature in granular catalyst beds since chemical conversion and/or catalyst lifetime strongly depend on temperature. Thermal dispersion in a porous model is a complex phenomenon resulting from diffusion in the solid phase and convection and diffusion in the moving fluid. Experimental works exist for studying this phenomenon in the porous media and chemical engineering literature [12, 4, 5] but to our knowledge inverse experimental methods have not yet been applied. The simplest homogeneous model that can be used in such a situation is based on a local mean temperature that is a average between the local solid and fluid temperature that are weighted by their respective heat capacities [10]. This reduced model requires the definition of a thermal dispersion tensor, whose coefficients can be considered as pseudo-conductivities that depend on the local Darcy's (or filtration) velocity.

Metzger [6] and Metzger *et al*. [7, 8] have shown experimentally that this model could be used in the case of water flowing through a bed of glass beads. They estimated the dependence of the longitudinal thermal dispersion coefficient on the reduced Darcy's velocity (the Peclet number). In that water/glass beads case they could only yield rough estimates of the transverse dispersion coefficient.

The same approach is implemented here for a gas (air or nitrogen) flowing through two different granular beds, without (same glass beads bed) or with (catalyst support) internal porosity.

Changing from water to an air flow through glass beads produces a large change of the solid to fluid conductivity ratio (λ_s / λ_f), see Table 1, that passes from 2 to 40. The same is true for the volumetric heat capacities (ρc_p) of both solid and fluid phases whose ratio ($(\rho c_p)_s/(\rho c_p)_f$) passes from 0.5 to 1600. So one challenge in the characterization of thermal dispersion in such a solid/fluid medium was to test the validity of the one-temperature model through an inverse experimental approach.

2. MODEL AND EXPERIMENTAL SETUP

Model

The one-temperature model, for a fluid (f) flowing through a granular solid medium (s) is a reduced model where the two phase medium is replaced by a homogeneous medium.

It is given by the following convection-diffusion equation:

$$\left(\rho c_p\right)_t \frac{\partial \langle T\rangle}{\partial t} = \nabla . \left(\boldsymbol{\lambda} \nabla \langle T\rangle\right) - \left(\rho c_p\right)_f \mathbf{u}_D . \nabla \langle T\rangle + s \tag{1}$$

where s is a volumetric heat source (heat power per unit volume) that dissipates heat in the homogenized granular medium and the total volumetric heat $(\rho c_p)_t$ of the medium is given by a mixing law based of the volumetric heat of both phases :

$$\left(\rho c_p\right)_t = \varepsilon \left(\rho c_p\right)_f + (1-\varepsilon) \left(\rho c_p\right)_s \tag{2}$$

and where ε is the granular medium porosity, that is the fluid volumetric fraction. In equation (1) $\boldsymbol{\lambda}$ is the thermal dispersion tensor and $\mathbf{u_D}$ Darcy's velocity (also called filtration or superficial velocity). The average enthalpic temperature $\langle T\rangle$ in a point P of the medium is a local weighted average of temperature in a sphere of volume V, centred in P, of radius R , that is large with respect to the characteristic size of the granular medium (the grain diameter), but small with respect to the representative system length (the width of the bed here):

$$\langle T\rangle(\mathrm{P},t) = \frac{1}{(\rho c_p)_t \, V} \int_V \rho c_p(\mathrm{P}') \, T(\mathrm{P}',t) \, \mathrm{d}V \tag{3}$$

If the granular medium can be considered as structural isotropic and if the fluid flows in the x direction with a uniform filtration velocity, eqn. (1) becomes:

$$\left(\rho c_p\right)_t \frac{\partial T}{\partial t} = \left(\lambda_x \frac{\partial^2 T}{\partial x^2} + \lambda_y \frac{\partial^2 T}{\partial y^2} + \lambda_z \frac{\partial^2 T}{\partial z^2}\right) - \left(\rho c_p\right)_f u_D \frac{\partial T}{\partial x} + s \tag{4}$$

where the triangular brackets around temperature T have been omitted. Let us note here that the three dispersion coefficients, λ_x, λ_y et λ_z, depend on the structure of the porous medium, on the nature of the fluid and of the filtration velocity u_D.

Solution of the direct problem

We consider here a fixed granular bed shown in Figure 1 and through which a fluid downwards flows with uniform filtration velocity $u = u_D$. We assume initial thermal equilibrium ($T = T_0$). An electric heating wire is set in the z direction normal to the x - y plane of the figure and at the origin ($x = y = 0$) of the coordinate system. It dissipates heat with a power step of lineic power intensity Q (W.m^{-1}) at time $t = 0$. One considers here the medium as infinite, which means that the temperature response to this excitation is $\Delta T = T - T_0$ and is equal to zero at large distances from the source. It can be calculated using the two-dimensional Green's function [2]:

$$\Delta T(x,y,t) = \frac{Q}{4 \pi \sqrt{\lambda_x \lambda_y}} e^{\frac{(\rho c_p)_f u x}{2 \lambda_x}} \int_0^{\frac{(\rho c_p)_f^2 u^2 t}{4 (\rho c_p)_t \lambda_x}} e^{-\left(\frac{x^2}{\lambda_x} + \frac{y^2}{\lambda_y}\right) \frac{(\rho c_p)_f^2 u^2}{16 \lambda_x} \frac{1}{\theta} - \theta} \frac{d\theta}{\theta} \tag{5}$$

This integral can be calculated through a numerical quadrature.

Experimental setup

A fixed bed, see Figure 1, is constituted of monodisperse glass beads of diameter d = 2 mm, and porosity ε = 0.365. Either water or air can flow downwards through it. Table 1 gives the thermal properties of both phases. The initial bench [8] (water flow) has been modified for a gas flow. A fan is located in a cylindrical duct downstream the setup and aspires air from a high volume room upstream the lab through a second upstream cylindrical duct. The large volume of the upstream room allows a quasi-constant temperature from the air input.

The heating wire is perpendicular to the air flow. Thirteen thermocouples, of type E and of 127 µm diameter, set parallel to the wire and located mainly downstream the heating wire, measure the temperature response of the medium to the power step. Thermocouples 12 and 13 allow to check the zero temperature variation upstream while thermocouples 8 and 11 allow to verify that the heated zone does not reach the wall and that the assumption of an infinite medium is valid. The air velocity is measured by a hot wire anemometer in the downstream cylindrical duct.

	Water	*Air*	*Glass*	*Equivalent bed properties*	
				Water/glass beads	*air/glass beads*
ρc_p (KJ/m^3)	4170	1.2	2080	2840	1320
λ (W/m/K)	0,607	0,026	1	0.831	0.2

Table 1. Thermal properties of the two phases of the granular medium.

The heating level Q is chosen in order not to modify the thermophysical properties of both fluid and solid (maximum temperature rise of the order of one Celsius degree). Measurements have been made for Peclet numbers ($Pe = (\rho\, c_p)_f\ \ u\ d\ /\lambda_f$) from 10 to 70 in the case of air flow, which corresponds to maximum filtration velocities close to 0.7 m/s. For water flow the Peclet number varied between 10 and 130, with maximum filtration velocities of the order of 7 mm/s.

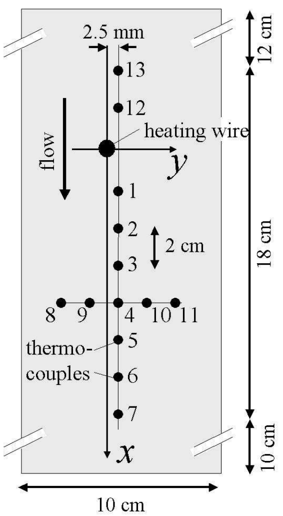

Figure 1. Dimensions of the granular media and positions of thermocouples.

3. PARAMETER ESTIMATION TECHNIQUE

Parameter estimation

The most classical parameter estimation technique relies on the minimization of the ordinary least square sum :

$$S_T(\boldsymbol{\beta}) = \sum_i \sum_k \left(T_{\exp,\, ik} - T(x_i, y_i, t_k, \boldsymbol{\beta})\right)^2 \quad (6)$$

where $T_{\exp,\, ik}$ is the temperature measured at the location (x_i, y_i) of the i^{th} thermocouple and t_k, the k^{th} time of measurement and $T(x_i, y_i, t_k, \boldsymbol{\beta})$ the corresponding theoretical temperature given by solution (5) of the direct problem, which depends of the different parameters to be estimated. If the exact locations of the thermocouples are known, it is possible to estimate the two dispersion coefficients as well as the filtration velocity, that is vector $\boldsymbol{\beta} = [\lambda_x, \lambda_y, u]^t$. In practice that is not the case and it is necessary to estimate not only λ_x, λ_y and u but also the unknown locations (x_i, y_i). This comes from the fact that, in order to have the less intrusive character and the highest possible response times, the thermocouple wires are very thin (127 µm diameter) and are first tightened in the empty bed. The posterior construction of the bed by filling the 2mm glass beads into the box of the setup (see Figure 1) causes a displacement of the hot junctions of the thermocouples (in the $z = 0$ plane), which means that their exact locations (x_i, y_i) differ from their nominal locations (x_i^{nom}, y_i^{nom}).

In order to estimate the new parameter vector $\boldsymbol{\alpha}_5 = [\lambda_x, \lambda_y, u, (x_i, y_i)_{i\,=\,1,\,Ntc}]^t$, with Ntc the number of thermocouples used, the prior knowledge of these nominal location is incorporated in the new objective function:

$$S'(\alpha_5) = \frac{1}{\sigma_T^{\;2}} \sum_i \sum_k \left(T_{ik}^{\exp} - T_{ik}(\alpha_5)\right)^2 + \frac{1}{\sigma_{pos}^{\;2}} \sum_i \left(x_i^{nom} - x_i\right)^2 + \frac{1}{\sigma_{pos}^{\;2}} \sum_i \left(y_i^{nom} - y_i\right)^2 \quad (7)$$

where σ_{pos} is the standard deviation of the location in x and y of the thermocouple hot junctions in the bed and σ_T the standard deviation of the temperature measurement.

The experimental temperature standard deviation can be measured in a steady state situation ($\sigma_T = 0.02$°C), that is without any excitation Q, and it can be assumed that the standard deviation of the location of a hot junction, that is a measure of its displacement, is of the order of one bead radius ($\sigma_{pos} = 1$ mm).

This type of estimator has been studied previously in the simpler two parameter case (slope and intercept) of the straight line model with stochastic errors on both dependent (ordinate) and independent (abscissa) variables [6]. It is called a *measurement error model* in the statistical literature [3, 11] and qualified as a *functional* model in the case where the model is exact and where the exact values of the independent variable(s), also called *incidental* variables, x_i and y_i in our case – are deterministic. When the incidental variables are independently distributed stochastic variables of common variance, the statistical model is called *structural* or *ultrastructural* model, depending whether they are identically distributed random variables or not.

Minimisation of sum (7) with respect to parameter vector $\boldsymbol{\alpha}_5$ constitutes a modified version of the Total Least Square estimator (which is obtained for $\sigma_T = \sigma_{pos} = 1$) or to orthogonal regression where both variables, here temperatures T_i and locations (x_i, y_i) are treated symmetrically.

It is also possible to show that this minimisation, which is made here through the Gauss-Newton algorithm, corresponds to the use of a Gauss-Markov estimator, with a minimum variance for the estimates of the coefficients of the parameter vector.

Metzger *et al.* have shown [6]-[9] that these estimated values depend quite weakly on the choice of the standard deviation σ_{pos}, as soon as it is bigger than a fraction of a millimetre. A low value σ_{pos} (smaller than one micrometer) leads to very poor temperature residuals with estimated locations close to their nominal values ($\hat{x}_i \approx x_i^{nom}$; $\hat{y}_i \approx y_i^{nom}$); values of σ_{pos} in-between one micrometer and a few tenth of millimetre lead to a decrease of the residuals and a variation of the estimated values. As soon as $\sigma_{pos} = 1$ mm, both residuals and estimates become good and do not vary any more. At last, for $\sigma_{pos} > 1$mm, one nearly meet the case of ordinary least squares where temperatures only are fitted and the non linear inversion algorithm does not converge anymore. Let us notice that multiplication of sum S' by $\sigma_T^{\,2}$ show that this minimisation can also be considered as Tikhonov zeroth order regularization where the regularization coefficient is $(\sigma_T/\sigma_{pos})^2$.

Monte Carlo simulations

It is possible to make Monte Carlo simulations of inversion [1]: the true temperature response of model (5) is noised with an independent additive normal random noise of zero mean and standard deviation σ_T, which yields the simulated experimental temperatures $T_{\exp,\, ik}$. The same technique is implemented with both exact thermocouple coordinates (x_i, y_i) that are noised the same way with a noise of standard deviation σ_{pos} to produce the nominal locations (x_i^{nom}, y_i^{nom}). A Gauss-Newton minimization of S' yield an estimation $\hat{\boldsymbol{\alpha}}_5$ of the parameter vector. If 400 simulations of this type are made with the corresponding inversions, 400 estimates $\hat{\alpha}_j^{(n)}$ are available for the j^{th} parameter of $\hat{\boldsymbol{\alpha}}_5$, n being the inversion number. It is then possible to reach the statistical distribution of each estimated parameter (its histogram) and to calculate the dispersion (standard deviation s_j) of each estimate as well as its bias b_j, that are :

$$b_j = \bar{\hat{\alpha}}_j - \alpha_j \quad \text{and} \quad s_j = \frac{1}{400}\sum_{n=1}^{400} \left(\hat{\alpha}_j^{(n)}\right)^2 - \left(\bar{\hat{\alpha}}_j\right)^2 \quad \text{with} \quad \bar{\hat{\alpha}}_j = \frac{1}{400}\sum_{n=1}^{400} \hat{\alpha}_j^{(n)} \tag{8}$$

Such estimates are given in Table 2 for air or water flow through the glass beads. They correspond to the locations of thermocouples 2 to 7 in Figure.1 and to a time step of 0.15 s, with a final time of 900 s for air, the corresponding values being 0.15 s and 45 s for water. One can use here the $(|b_j| + s_j)/\alpha_j$ ratio (relative error) as an index of quality of inversion for parameter α_j. The λ_x estimations have the same quality for air and water with "relative errors" smaller than 3 %: bias is larger for air but it is compensated by a lower dispersion. For λ_y estimations the "relative error" is still acceptable for air (5 %) but too large for water (21 %) to yield precise values. For both fluids the filtration velocity is the parameter that is estimated with the maximum precision (relative errors lower than 2 %). This confirms the possibility of estimating the transverse dispersion coefficient for air, which was not possible for water.

It is interesting to notice here that the estimation bias on the different parameters, that is caused by the non linear character of the estimator here, can be of the same magnitude or even higher than the standard deviation, see the $|b_j|/s_j$ column in Table 2.

	j	parameter	exact value α_j	average estimation $\bar{\hat{\alpha}}_j$	estimation bias b_j	estimation standard deviation s_j	bias/dispersion $\|b_j\|/s_j$	« relative error » $(\|b_j\|+s_j)/\alpha_j$
air	1	λ_x (W.K^{-1}.m^{-1})	0.962	0.984	+0.022	0.008	275 %	3 %
	2	λ_y (W.K^{-1}.m^{-1})	0.256	0.246	-0.010	0.003	336 %	5.2 %
	3	u (m.s^{-1})	0.353	0.355	+0.002	0.004	50 %	1.7 %
water	1	λ_x (W.K^{-1}.m^{-1})	60	60.321	+0.321	1.009	32 %	2.2 %
	2	λ_y (W.K^{-1}.m^{-1})	3	2.681	-0.329	0.310	106 %	21 %
	3	u (mm.s^{-1})	6.288	6.306	+0.018	0.033	55 %	0.8 %

Table 2. Monte Carlo simulations of inversion for air or water flow through a bed of glass beads.

4. EXPERIMENTAL RESULTS AND RESIDUALS FOR GLASS BEADS

The experimental and recalculated thermograms (thermocouples 2 to 7) as well as the temperature residuals are shown in Figure 2a for water flow and in Figure 2b for air flow through the 2mm glass beads. The Peclet numbers are close for the two cases ($Pe \cong 30$). The temperature residuals $\left(T_{\exp,\,ik} - T_{ik}(\hat{\boldsymbol{\alpha}})\right)$ are plotted as functions of time for each thermocouple. Their quadratic mean is lower than 30 mK and identical for the two fluids. The low level and the non correlated shape of the residuals show that the one-temperature model fits very well the experimental curve for both flows. It is interesting to notice that the measurement duration is about 10.

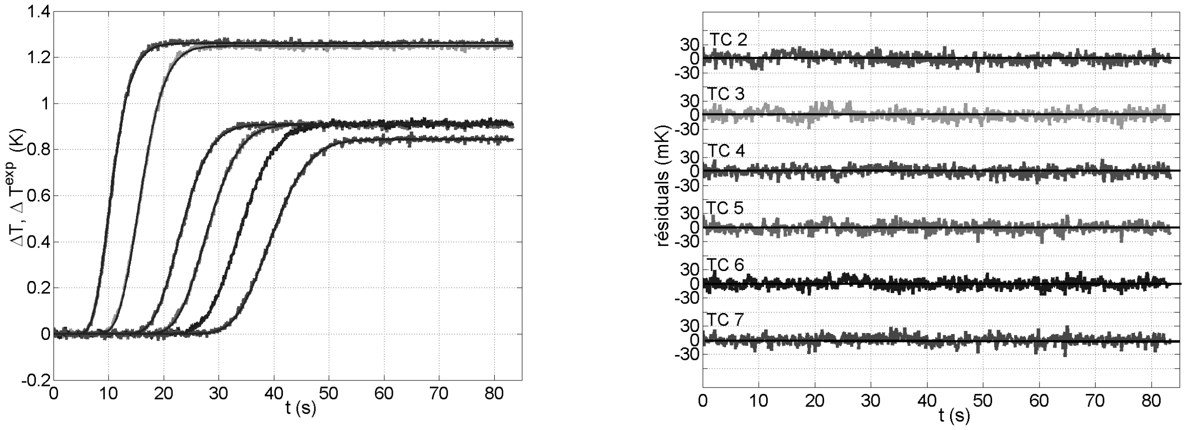

Figure 2a. Transient temperature response and residuals for the case of water for Pe ≈ 30.

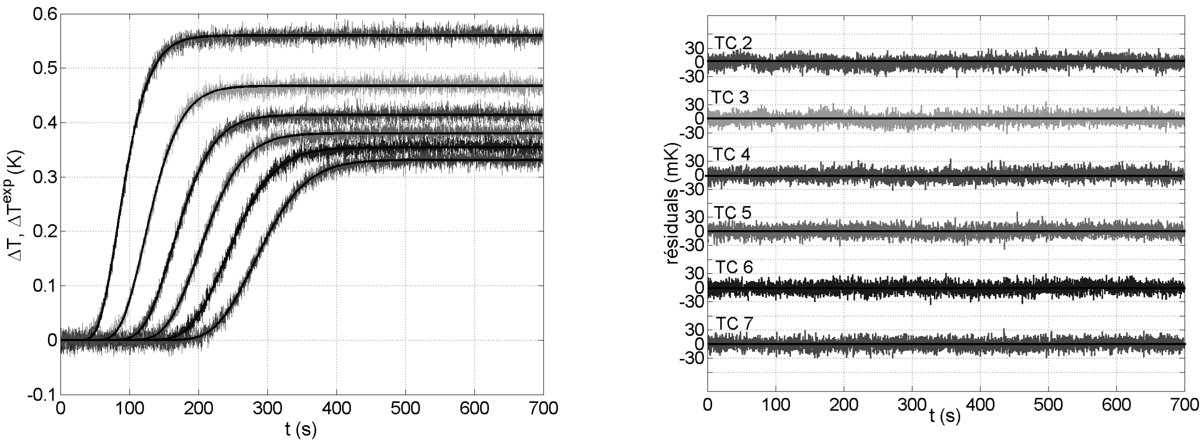

Figure 2b. Transient temperature response and residuals for the case of air Pe ≈ 30.

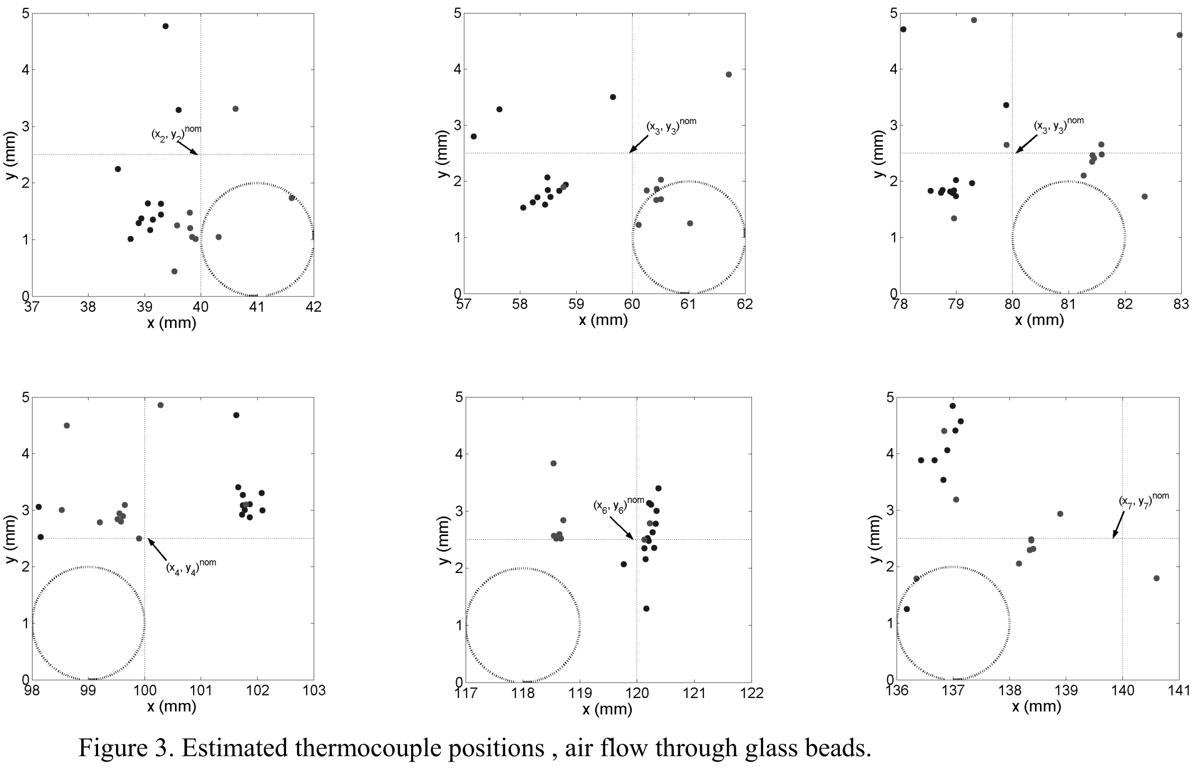

Figure 3. Estimated thermocouple positions , air flow through glass beads.

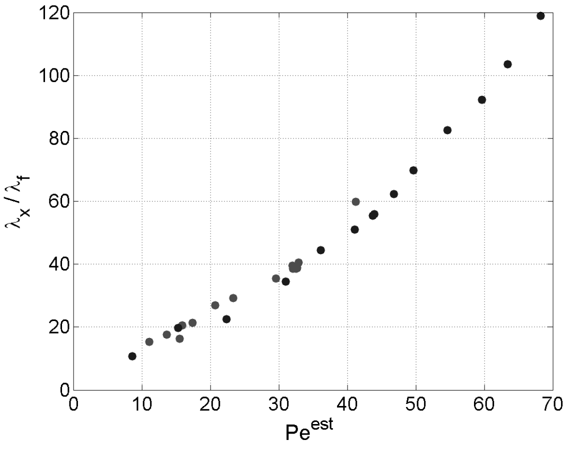

Figure 4. The result of estimation of longitudinal thermal dispersion coefficient.

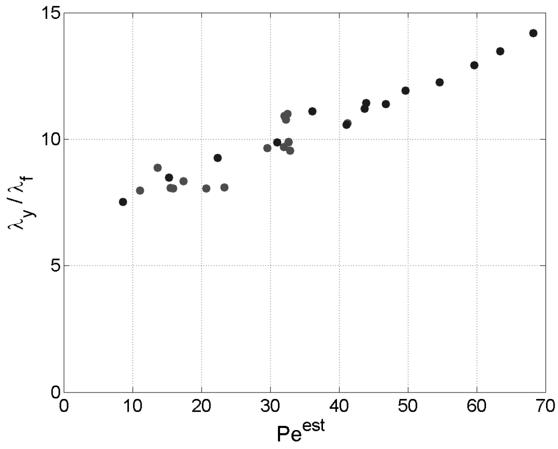

Figure 5. The result of estimation of transversal thermal dispersion coefficient.

The experimental location residuals are presented for air flow in Figure 3 for Peclet number ranging from 10 to 70 : the nominal location of each thermocouple is located at the center of each (x, y) coordinate system while the corresponding estimated location , for a given Peclet number, is set in the same figure. The size of a bead is represented by a circle in each figure. It is interesting to notice that the estimated locations are distributed uniformly, in terms of angular position and polar radius, around the nominal location point, which seems to show that there is no large experimental bias in the location estimation procedure. The corresponding values of the estimations of λ_x and λ_y are shown in Figures 4 and 5. The estimates have not been subjected to a bias correction here. One can notice that the dispersion of the estimations of λ_x agree with the theoretical Monte Carlo simulations shown in Table 2. That is not the case for the estimations of λ_y that are more dispersed at low velocities where some velocity fluctuations during the experiment are suspected (a fan with a non optimal efficiency was used).

5. EXPERIMENTAL RESULTS FOR SOLID GRAINS WITH INTERNAL POROSITY

Experiments with a bed of glass beads, through which air flows, have allowed both the measurement of the dispersion coefficients and the validation of the one-temperature model in this situation for this type of granular academic medium. We have tried to use the same inverse technique for a change of solid grains : the solid grain used (a ceramics, with approximately spherical grains of granulometry between 2 and 4 mm) has an internal porosity, which was not the case for the glass beads. The thermal properties of the ceramics grains (conductivity, volumetric heat) differ from those of the glass beads and the porosity of the ceramics grain/air medium differ from the porosity of the glass beads/air system.

The temperature responses of the new medium that has been filled in the setup box are shown in Figure 6. They are very different from the glass/air thermograms and our model, eqn.(5) does not fit the experimental curves.

Presence of liquid adsorbed in the micro-pores of the grains, that are hygroscopic in the presence of humid air, is suspected for this deviation: this water can evaporate because of the decrease in relative humidity of air that has been heated by the heating wire. This could cause a negative non linear heat source in the heat equation. This source depends of course of the saturation curve of the grain and can not be integrated in our model. It was therefore necessary to make the experiments with both dry grains and dry gas. Air flow was replaced by dry nitrogen that expanded out of N_2 cylinders through a gas regulator into the setup. Nitrogen flowed during more than one hour through the bed, in order to dry it, before heating started.

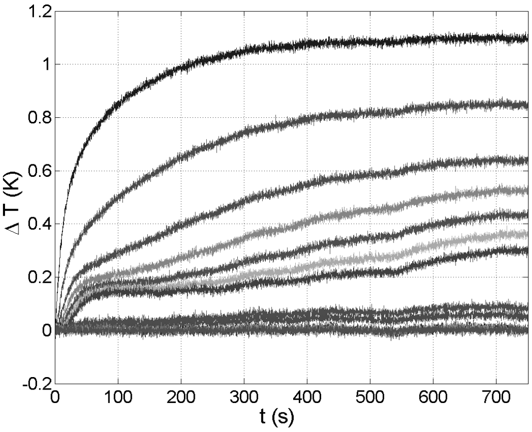

Figure 6. Temperature response signal for air / porous grains.

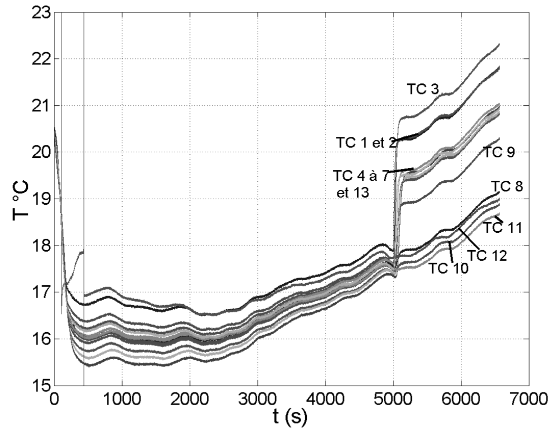

Figure. 7. Temperature response signal for nitrogen / porous grains.

Thermograms are shown for this type of ceramics grain/nitrogen flow in Figure 7, for a filtration velocity of 0.5 m/s. Heating starts here at time t_s = 5000 s. The medium is not at initial uniform temperature and the inlet gas temperature varies with time. This is why a strong temperature drift with time can be noticed. A temperature correction of the thermograms, $T_i(t)$, is necessary . It uses a control thermocouple set in the free gas flow upstream the set up and yield an experimental reference thermogram $T_{réf}(t)$. A moving average is made with three points of the reference thermogram and this temperature is subtracted from each thermogram:

$$T_i'(t_k) = T_i(t_k) - \overline{T}_{ref}(t_k) \qquad \text{with} \qquad \overline{T}_{ref}(t_k) = \frac{1}{3}\sum_{m=-1}^{1} T_{ref}(t_k + m\ \Delta t) \tag{9}$$

where the time step is $\Delta t = 0.15$ s.

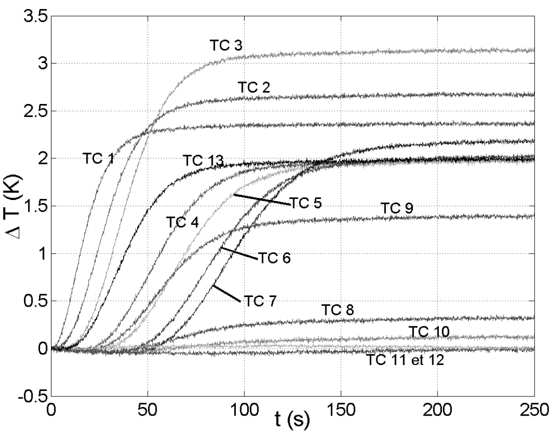

Figure 8. Temperature response signal for nitrogen/porous grain after corrections.

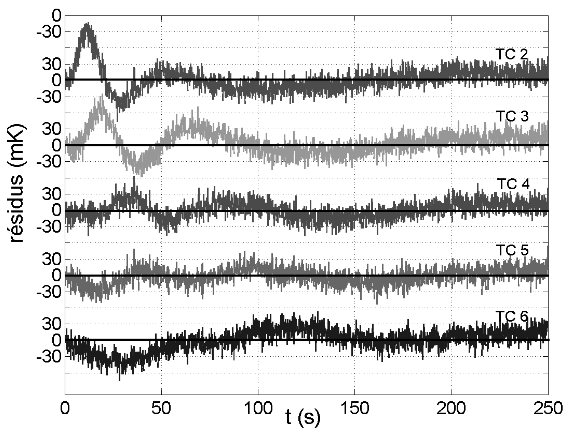

Figure 9. Residuals for nitrogen/porous grains.

This correction is not sufficient because a drift remains before the excitation. A second correction of slope and intercept is calculated for the 30 seconds before excitation:

$$T_i''(t_k) \;=\; T_i'(t_k) - a_i(t_k - t_{start}) - T_i'(t_{start}) \tag{10}$$

where $a_i = \dfrac{1}{\sum_{t\langle t_k}\left(t_k - t_{start}\right)^2} \sum_{t\langle t_k}\left(t_k - t_{start}\right)\, T_i^{'}\left(t_k\right)$ (correction of the drift of each individual temperature by a one-parameter linear regression).

Figure 8 shows the T_i'' thermogram after these two corrections, with a new time origin $t = 0$ set at former time t_s. Starting from these thermograms, it is possible to invert the responses of thermocouples 2 to 6. Residuals are shown in Figure 9. They are higher and more correlated than in the glass beads/air case but since heating has been higher than in the preceding case (3 K instead of 1 K), the residual over signal ratio remains the same. The filtration velocity that can be deduced from the free stream velocity measurement by the hot wire anemometer is 0.50 m/s while the estimated velocity is 0.52 m/s: this agreement is excellent in spite of the correction made. The parameter estimation is still possible.

6. CONCLUSIONS

The quality of the temperature residuals that have been presented show that the one-temperature model can be used for modelling heat transfer in a bed of glass beads through which either water or air flows, in spite of the very different thermophysical properties of both phases in the latter case: conductivity ratio, solid over gas (air) of 40 and a corresponding volumetric heat ratio of 1600. The use of a modified least squares sum (total or orthogonal least squares estimation, after normalization of the temperature and location residuals), that takes into account the uncertainty on the exact locations of the thermocouple hot junctions, made these results possible. A point that deserves to be underlined here is the precision with which the filtration or Darcy's velocity has been estimated. This shows that velocities can be measured indirectly by temperature measurements in the "thermal wake" of a heat source in inverse problems based on a convection-diffusion equation. This model has also been extended to cases where the granular phase is also characterized by an internal micro-porosity, thanks to a mastering of the effects of humidity adsorption.

NOMENCLATURE

b_j	bias
c_p	heat capacity, J.K^{-1}.kg^{-1}
d	particle diameter, m
Pe	Peclet number, $u_D\, d\, /\, a_f$
Q	linear heating power, W.m^{-1}
s	volumetric heat source, W.m^{-3}
s_j	standard deviation
T	temperature, K
t	time, s
u_D, u	Darcy's velocity, m.s^{-1}
x, y, z	space coordinates, m
Greek symbols	
$\boldsymbol{\alpha}$	parameter vector
α_j	parameter
ε	porosity
λ	thermal conductivity, W.m^{-1}.K^{-1}
λ_x, λ_y	thermal dispersion coefficients, W.m^{-1}.K^{-1}
ρ	density, kg.m^{-3}
Subscripts	
f	fluid phase
s	solid phase
Superscripts	
^	estimated value
_	average value

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