T6 Thermal characterization of an insulating material through a tri-layer transient method

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Abstract. The three layers transient method is dedicated to the thermal properties measurement of insulating materials. The three layers experimental device (brass/sample/brass) and the principle of the measurement based on a pulsed method will be first presented. The three dimensional modelling of the system will be developed and used for a sensitivity analysis. The estimation method will be described and its application to simulated noisy measurements realized with COMSOL will be presented. During the workshop, several experiments will be carried on different materials and the experimental temperature records will be used to estimate the thermal properties of the tested samples. Some improvements to the initial model such as taking into account a parallel or series thermal resistance will be discussed.

6.1 Introduction

The three layers method [1] has been specially developed for thermal conductivity measurement of small samples of low-density insulating materials since the existing methods are not suited to this type of materials. The contact transient methods using plane or linear heating element: hot disk [2-3], hot wire [4], hot plate [5], hot strip [6-7] cannot measure precisely thermal conductivity of low density insulating materials for the following reasons:

- The thermal capacity and the thermal resistance of the heating element (often heterogeneous and made of a metal wire inserted between two plastic films) is not known with precision and is often taken into account by a simplified model.
- The sensitivity of the measured temperature to the thermal capacity of the heating element is very high if the thermal capacity of the insulating material is low (case of a low density material)
- The thermal conductivity of the heating element is higher than the conductivity of the insulating material. The longitudinal heat transfer (parallel to the contact surface between the heat source and the sample) in the heating element that is not taken into account in the models may be and lead to estimation errors.
- The Flash method [8] is difficult to use for the following reasons:
- The insulating materials are often semi-transparent to the radiations of the Flash lamp,
- It is very difficult to measure precisely a surface temperature on a low density material,
- The heat transfer on the heated face is often very different of the heat transfer on the other faces (very important temperature differences).

To avoid the first two disadvantages, the sample may be inserted between two heat conducting plates; a device based on this principle has already been used for the liquids [9]. For very low density materials, one can show that the sensitivity of the unheated face temperature to the thermal conductivity is highly correlated to the sensitivity to the convective losses and that the sensitivity to the thermal diffusivity is low.

The aim of this work was to develop a new method suited to the thermal conductivity (and eventually diffusivity) measurement of low and very low density insulating materials.

6.2 Principle and experimental device

The experimental device includes a cylindrical sample (R = 2cm, e = 5 to 10mm) of the material to be characterized inserted between two brass discs with a thickness e_b = 0,4mm and the same radius (cf. figure 1). Two type K thermocouple with wire diameter 0,05mm are welded on the external face of each brass disc by the technique of the separated contact (with a distance of 5mm between the two wires). The lower disc is in direct contact with a plane circular heating element having the same diameter and set on an insulating material. A pressure is applied on the unheated brass disc by four PVC (chosen for its low thermal conductivity) tips with a very low contact surface area. The upper surface of the unheated brass disc exchanges with the ambient air by natural convection and radiation.

A heat flux is applied during a few seconds to the heating element and the temperatures $T_{b1}(t)$ and $T_{b2}(t)$ of the brass discs are recorded. A three-dimensional model associated to an inverse method is then used to estimate the thermal conductivity and the thermal diffusivity of the insulating material inserted between the two brass discs.

The heat flux is produced by a plane-heating element during a few seconds instead of being produced by a flash lamp (device initially tested) during a few milliseconds for the following reasons:

- The temperature increasing of the heated face must not be too fast to be compatible with the thermocouple response time.
- A uniform pressure may be easily applied on the brass discs through the plane heating element.
- A light part of the flash may reach the lateral surfaces by reflection, this disadvantage is avoided with a plane heating element.

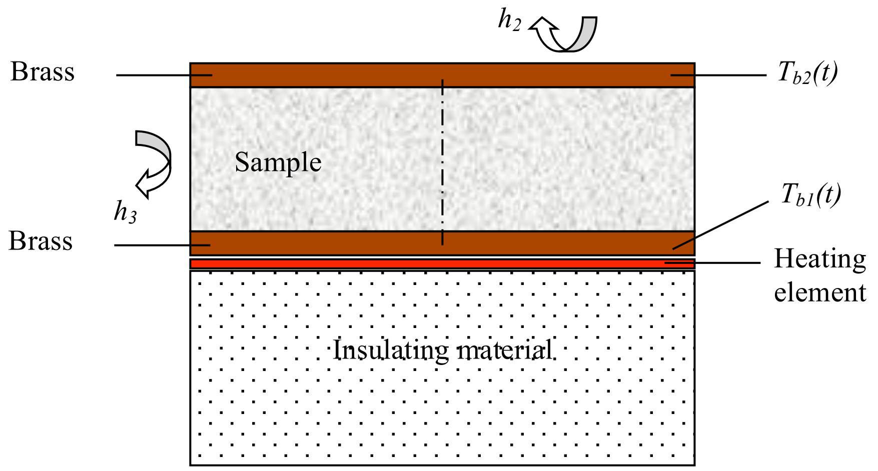

Figure 1. Experimental device

6.3 Model and estimation method

Assumptions:

- The temperatures T_{b1} and T_{b2} in the brass discs are uniform
- The thermal contact resistances (typically 10^{-4} $m^2.K.W^{-1}$) between the sample and the brass discs are negligible in comparison with the thermal resistance of the sample (greater than 5 x 10^{-2} $m^2.K.W^{-1}$ for the tested samples).

As a first step, the following case is considered: a unique sample with heat transfer by convection with the ambient air on all its faces receives a direct and short heating on one face (no brass discs as represented in figure 2).

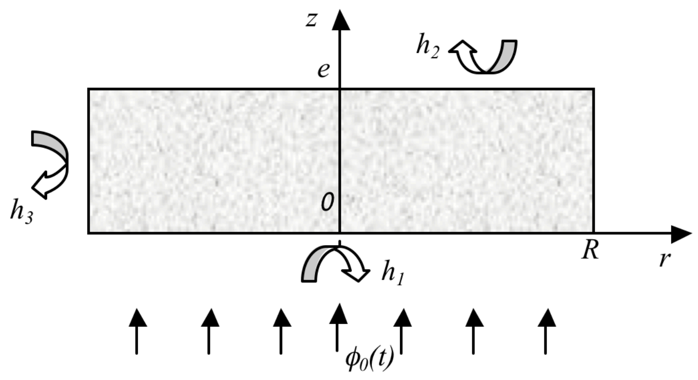

Figure 2. Experiment schema for a unique sample

Setting $\overline{T}(r,z,t) = T(r,z,t) - T_e$
The equation of heat becomes:

$$\frac{\partial^2 \overline{T}(r,z,t)}{\partial r^2} + \frac{1}{r}\frac{\partial \overline{T}(r,z,t)}{\partial r} + \frac{\partial^2 \overline{T}(r,z,t)}{\partial z^2} = \frac{1}{a}\frac{\partial \overline{T}(r,z,t)}{\partial t} \tag{6.1}$$

Initial and boundary conditions may be written as:

$$z = 0 \rightarrow \lambda \frac{\partial \overline{T}(r,0,t)}{\partial z} = h_1 \,\overline{T}(r,0,t) - \phi_0(t) \tag{6.2}$$

$$z = e \rightarrow -\lambda \frac{\partial \overline{T}(r,e,t)}{\partial z} = h_2 \overline{T}(r,e,t) \tag{6.3}$$

$$r = 0 \rightarrow \frac{\partial \overline{T}(0,z,t)}{\partial r} = 0 \tag{6.4}$$

$$r = R \rightarrow -\lambda \frac{\partial \overline{T}(R,z,t)}{\partial r} = h_3 \,\overline{T}(R,e,t) \tag{6.5}$$

$$t = 0 \rightarrow \overline{T}(r,z,0) = 0 \tag{6.6}$$

The Laplace transform applied to relation (1) with $L\left[\overline{T}(r,z,t)\right] = \theta(r,z,p)$ leads to:

$$\frac{\partial^2 \theta(r,z,p)}{\partial r^2} + \frac{1}{r}\frac{\partial \theta(r,z,p)}{\partial r} + \frac{\partial^2 \theta(r,z,p)}{\partial z^2} = \frac{p}{a}\theta(r,z,p) \tag{6.7}$$

The Laplace transforms of the boundary conditions are:

$$\lambda \frac{\partial \theta(r,0,t)}{\partial z} = h_1 \theta(r,0,t) - \Phi_0(t) \tag{6.8}$$

$$-\lambda \frac{\partial \theta(r,e,t)}{\partial z} = h_2 \theta(r,e,t) \tag{6.9}$$

$$\frac{\partial \theta(0,z,t)}{\partial r} = 0 \tag{6.10}$$

$$-\lambda \frac{\partial \theta(R,z,t)}{\partial r} = h_3 \, \theta(R,e,t) \tag{6.11}$$

$$\theta(r,z,0) = 0 \tag{6.12}$$

Setting: $\theta(r,z,p) = R(r,p)\, Z(z,p)$
One obtains:

$$\theta(r,z,p) = \sum_{n=1}^{\infty} A_n J_0(\alpha_n r) \, \{\beta_n \, ch[\gamma_n (e-z)] + H_2 \, sh[\gamma_n (e-z)]\} \tag{6.13}$$

Where:

$$A_n = \frac{2\Phi_0(p) \frac{e}{\lambda}}{\omega_n \left(1 + \frac{\omega_n^2}{H_3^2}\right) J_1(\omega_n) \left[\left(\beta_n^2 + H_2 H_1\right) sh(\beta_n) + \beta_n (H_2 + H_1) ch(\beta_n)\right]} \tag{6.14}$$

ω_n is solution of: $\omega J_1(\omega) = H_3 \, J_0(\omega)$, $\beta_n = \sqrt{\frac{p\, e^2}{a} + \left(\frac{e}{R}\right)^2 \omega_n^2}$, $H_1 = \frac{e\, h_1}{\lambda}$, $H_2 = \frac{e\, h_2}{\lambda}$,
$H_3 = \frac{h_3 \, R}{\lambda}$

The mean temperatures for $z = 0$ and $z = e$ may be calculated by integration of relation (13) between $r = 0$ and $r = R$:

$$\theta_{moy}(0,p) = \sum_{n=1}^{\infty} \frac{4\, \Phi_0(p) \frac{e}{\lambda} \, [\beta_n ch(\beta_n) + H_2 sh(\beta_n)]}{\omega_n^2 \left(1 + \frac{\omega_n^2}{H_3^2}\right) \left[\left(\beta_n^2 + H_2 H_1\right) sh(\beta_n) + \beta_n (H_2 + H_1) ch(\beta_n)\right]} \tag{6.15}$$

$$\theta_{moy}(e,p) = \sum_{n=1}^{\infty} \frac{4\, \Phi_0(p) \frac{e}{\lambda} \beta_n}{\omega_n^2 \left(1 + \frac{\omega_n^2}{H_3^2}\right) \left[\left(\beta_n^2 + H_2 H_1\right) sh(\beta_n) + \beta_n (H_2 + H_1) ch(\beta_n)\right]} \tag{6.16}$$

The next step is the study of the three layers device represented in figure 3.
The relations (1), (4), (5) and (6) remain valid in this case.
The local thermal balance at radius r on the heated brass disc (supposed at uniform temperature $T_{b1}(t)$) is:

$$\pi R^2 e_1 \, \rho_b c_b \frac{\partial T_{b1}}{\partial t} = \phi_0(t) \pi R^2 - h_1 \pi R^2 \, T_{b1} - h_3 \, 2\pi R \, e_1 \, T_{b1} + \int_0^R 2\pi r \, dr \, \lambda_s \, e \frac{\partial T(r,0)}{\partial z} \tag{6.17}$$

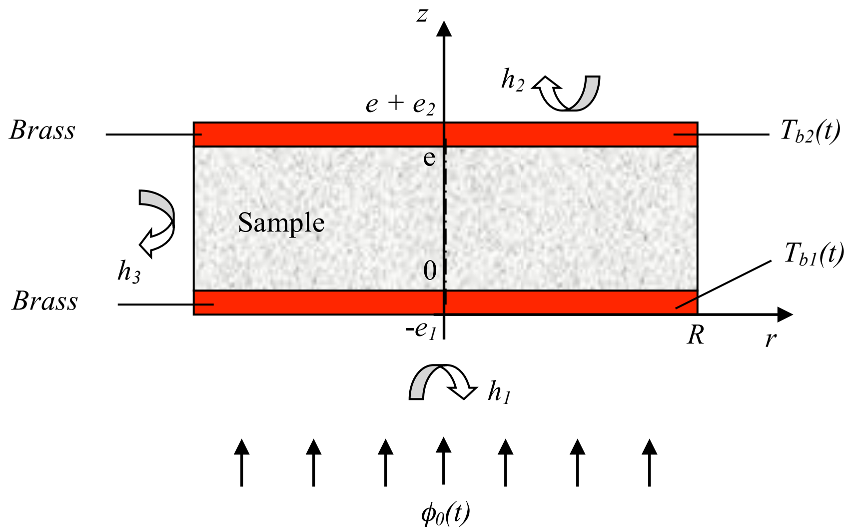

Figure 3. Experimental schema of a three layers device

The integration of this local balance between $r = 0$ and $r = R$ with a Laplace transformation leads to:

$$-\lambda \frac{\partial \theta_{moy}(e,p)}{\partial z} = \Phi_0(t) - \left[\rho_b\, c_b\, e_1\, p + \left(h_1 + \frac{2 h_3 e_1}{R}\right)\right] \theta_{b1} \tag{6.18}$$

This equation is similar to relation (8) considering the mean temperature for a given value of z instead of the local temperature at (r,z) and replacing h_1 by a corrected coefficient:

$$h_{1c} = \rho_b\, c_b\, e_1\, p + \left(h_1 + \frac{2 h_3 e_1}{R}\right) \tag{6.19}$$

The thermal balance of the unheated disc leads similarly to a corrected coefficient:

$$h_{2c} = \rho_b\, c_b\, e_2\, p + \left(h_2 + \frac{2 h_3 e_2}{R}\right) \tag{6.20}$$

By considering the mean temperature at z instead of the local temperature at (r,z), this boundary conditions are the same as for a unique sample. So that for the three layers system brass/sample/brass, the expressions (18) and (19) of the mean temperatures respectively at $z = 0$ and $z = e$ for a unique sample remains valid if h_1 is replaced by h_{1c} and h_2 by h_{2c} in relation (17).
The transfer function $H(p)$ of the system may be written as:

$$H(p) = \frac{\theta(e,p)}{\theta(0,p)} \tag{6.21}$$

And:

$$T_{b2}(t) = T_{b1}(t) \otimes L^{-1}[H(p)] \tag{6.22}$$

These two relations are true whatever the boundary condition on the heated brass disc is, particularly if this disc exchange heat by conduction with an insulating material (as described in figure 1) rather than by convection with air.
The principle of the method is to estimate the transfer function $H(p)$ by estimating the values of the three parameters a, λ and $h = h_2 = h_3$ that minimize the sum of the quadratic errors between the experimental

values of $T_{b2}(t)$ and those calculated by relation (25) with experimental values of $T_{b1}(t)$. The number of parameters to be estimated is the same that in the classical flash method (heat flux density ϕ, thermal diffusivity a and convection heat transfer coefficient h. The minimization is realized by using the Levenberg-Marquart method.

One of the advantage of the method is that it is not sensitive to the heat transfer on the heated face; in the case of a convective heat transfer with important variations of temperature on this face, the hypothesis of a constant convection coefficient being the same on the heated and on the unheated face is not totally true. It will be shown that this model approximation can lead to errors in the estimated parameters particularly in the case of a measurement realized on an insulating material where the heated face temperature may reach several decades degrees and where the resistance to the external transfer (convection) is of the same order of magnitude that the resistance to the internal transfer (conduction).

Compared to the limits (described in introduction) of the classical contact methods and of the Flash method for the thermal conductivity measurement of low density material, the proposed method has the following advantages:

- The temperature measurements are more precise since they are done on a heavy conductive material (brass),
- The thermal capacity of the two discs (homogeneous) is known precisely and taken into account,
- The longitudinal heat transfers in the discs are taken into account without approximation in the model (boundary condition of uniform temperature).

6.4 Sensitivity analysis

The reduced sensitivities of the transfer function H(p) have been calculated and represented in figure 4 for a three layers system with the following characteristics :

- Brass discs: e_b = 0.4mm, D = 40 mm, λ =100.04 $W.m^{-1}.K^{-1}$, a =3.14 x 10^{-5} $m^2.s^{-1}$;
- Samples:
  - Insulating material: λ = 0.02 $W.m^{-1}.K^{-1}$, a = 4 x 10^{-6} $m^2.s^{-1}$, e = 5mm et e = 10mm
  - Polystyrene: λ = 0.035 $W.m^{-1}.K^{-1}$, a = 8 x 10^{-7} $m^2.s^{-1}$, e = 10mm
  - Cellular concrete: λ = 0.15 $W.m^{-1}.K^{-1}$, a = 5 x 10^{-7} $m^2.s^{-1}$, e = 10mm

It can be noticed that:

- For a given material, if the sample thickness decreases then the sensitivity to the thermal diffusivity increases and the sensitivity to the thermal conductivity decreases.
- For a given sample thickness, if the thermal capacity of the sample increases then the sensitivity to the thermal diffusivity increases
- The sensitivity to the thermal diffusivity is quite low for very low density materials so that the thermal diffusivity will not be measurable with this method. Nevertheless, this sensitivity is high enough to estimate the thermal diffusivity of insulating material with higher density such as polystyrene or cellular concrete.

The sensitivity to the thermal conductivity is high in each case and is not correlated with the sensitivities to the convection coefficient and to the thermal diffusivity. The thermal conductivity is thus estimable by this method for all type of insulating materials.

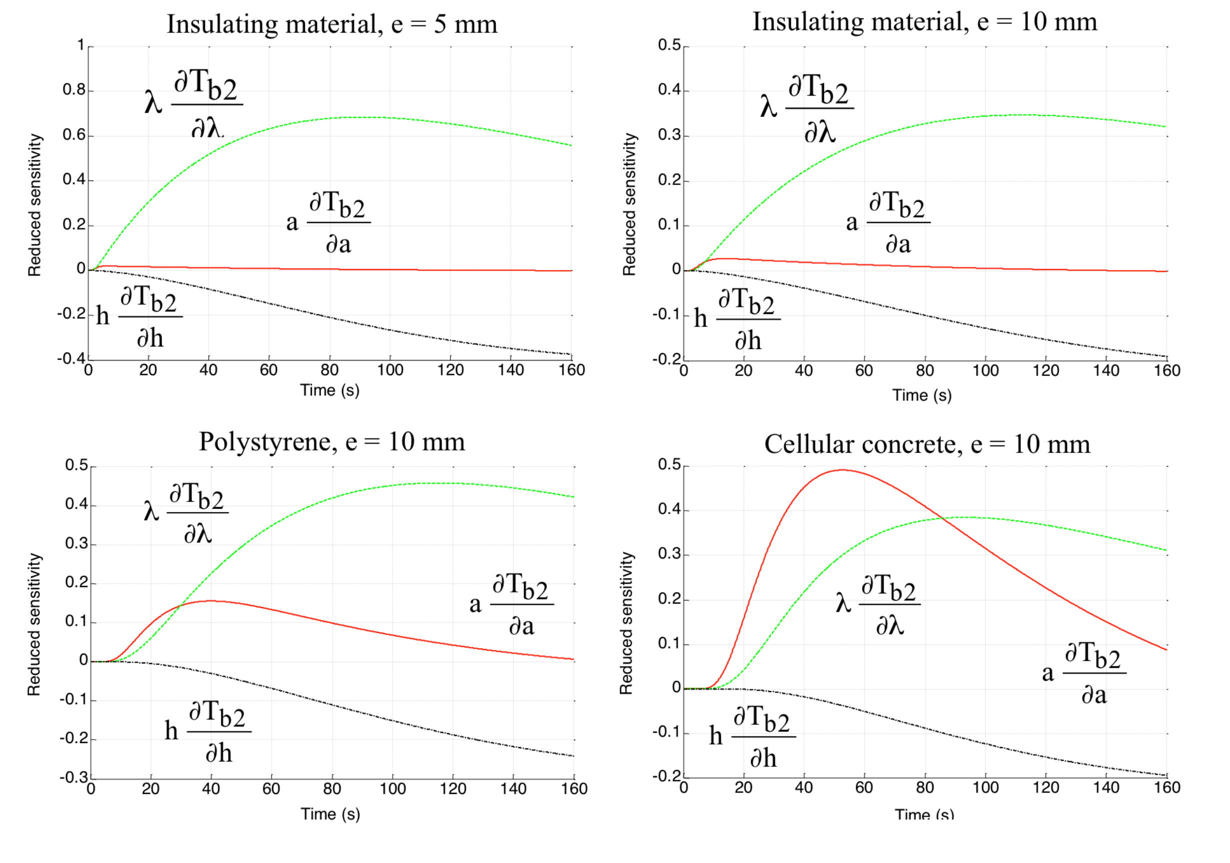

Figure 4. Reduced sensitivities of the transfer function H(p) calculated by relation (25)

6.5 Estimation from numerical simulations

The temperatures in a three layers device have been simulated with COMSOL. The following data has been considered:

- Sample: diameter = 35mm, thickness = 5.6mm, thermal properties: $\lambda = 0.02$ W.m^{-1}.K^{-1}, $\rho c = 5000$ J.m^{-3}.K^{-1}, $a = 4 \times 10^{-6}$ m^2.s^{-1}
- Metallic discs (copper): diameter = 35mm, thickness = 0.4 mm, thermal properties: $\lambda = 397.5$ W.m^{-1}.K^{-1}, $\rho = 8940$ kg.m^{-3}, $c = 384.9$ J.kg^{-1}.K^{-1}.

First, the simulation of the temperatures has shown that the hypothesis of uniform temperature in the two metallic discs is valid.

Then, the simulated temperatures $T_{b1}(t)$ and $T_{b2}(t)$ have been considered as experimental data and an estimation parameter has been applied according to the two following methods:

1. The unheated face temperature $T_{b2}(t)$ is the only experimental data considered as in the classical flash method. An inversion method is used to estimate the heat flux density ϕ_0, the thermal diffusivity a, the thermal conductivity λ and the convection coefficient h that minimize the sum of the quadratic errors between the experimental curve and the simulated curve $T_{b2}(t)$ calculated by relation (19) with the corrected convection coefficients given by relations (22) and (23).
2. The temperature $T_{b1}(t)$ of the heated face is considered as input experimental data of the system both with the temperature $T_{b2}(t)$. An inversion method is used to estimate the thermal diffusivity a,

the thermal conductivity λ and the convection coefficient h that minimize the sum of the quadratic errors between the experimental curve and the simulated curve $T_{b2}(t)$ calculated by relation (25) of the unheated face temperature.

The results of these two estimations are reported in table 1 where t_s is the upper bound of the estimation time interval.

Table 1. Results of the estimations realized by two different methods from temperatures simulated with COMSOL

$h_1 = h_2 = h_3 = 10$ W.m^{-2}.K^{-1}									
	$t_s = 100s$			$t_s = 200s$			$t_s = 300s$		
Estimation method	$10^6 a$	$10^3 \lambda$	h	$10^6 a$	$10^3 \lambda$	h	$10^6 a$	$10^3 \lambda$	h
1	4.18	17.7	11.0	4.19	16.4	11.3	4.2	16.5	11.3
2	4.01	19.8	11.1	3.95	19.8	11.1	3.8	19.9	11.2
$h_1 = 15$ W.m^{-2}.K^{-1} ; $h_2 = h_3 = 5$ W.m^{-2}.K^{-1}									
	$t_s = 100s$			$t_s = 200s$			$t_s = 300s$		
Estimation method	$10^6 a$	$10^3 \lambda$	h	$10^6 a$	$10^3 \lambda$	h	$10^6 a$	$10^3 \lambda$	h
1	4.12	33.8	7.4	4.11	33.8	7.4	4.1	34.0	7.5
2	3.95	19.9	5.4	3.93	19.9	5.4	3.9	19.9	5.5
$h_1 = 15$ W.m^{-2}.K^{-1} ; $h_2 = h_3 = 5$ W.m^{-2}.K^{-1} ; $dT_{b1} = dT_{b2} = 0.01$°C									
	$t_s = 100s$			$t_s = 200s$			$t_s = 300s$		
Estimation method	$10^6 a$	$10^3 \lambda$	h	$10^6 a$	$10^3 \lambda$	h	$10^6 a$	$10^3 \lambda$	h
1	4.16	27.0	8.4	4.22	31.7	7.7	4.1	33.7	7.5
2	3.88	20.1	5.3	3.85	20.5	5.6	3.8	20.6	5.7
$h_1 = h_3 = 10$ W.m^{-2}.K^{-1} ; $h_2 = 5$ W.m^{-2}.K^{-1}									
	$t_s = 100s$			$t_s = 200s$			$t_s = 300s$		
Estimation method	$10^6 a$	$10^3 \lambda$	h	$10^6 a$	$10^3 \lambda$	h	$10^6 a$	$10^3 \lambda$	h
2	3.92	19.4	6.2	19.3	16.4	6.1	4.01	19.3	6.1

It can be noticed that:

- The method 1, based on the 3D model and considering that the only known temperature is $T_{b2}(t)$, does not lead to a precise estimation of λ if the convection coefficient h_1 on the heated face is different of the coefficients h_2 and h_3 on the other faces.
- The method 2 gives in all cases a precise estimation of the thermal conductivity, even when a random noise with an amplitude of 0.01°C has been added to the temperatures simulated with COMSOL. The estimation of the thermal diffusivity becomes less precise if the measurement noise increases.
- A simulation based on method 2 with $h_2 = 5$ W.m^{-2}.K^{-1} and $h_1 = h_3 = 10$ W.m^{-2}.K^{-1} leads to an error of 3.5% on the estimated value of λ and of 2% on the estimated value of a. An extreme case has been considered ($h_3 = 2\ h_2$) so that the errors with experimental data will be less important.

The effect of an error on the supposed "known" value of the thermal capacity ρc_{mp} of the two metallic plates has also been investigated. It was found that an error of x% on ρc_{mp} leads to the same error on the estimated value of λ but has no significant influence on the estimated value of a, for $x < 5\%$. It has been verified that an error of 5% on the thermal conductivity of the metallic plates has no influence on the estimated values.

6.6 Experimental study

A first experiment will be carried on a sample of polyethylene foam with the following characteristics:
e = 3mm, D = 40mm. The estimated values will be compared to the following ones estimated by use of other methods: ρc = 80000 J.m^{-3} (measured by DSC) ; λ = 0.042 W.m^{-1}.K^{-1} (measured by the centred hot plate method [9]).
Other experiments will be carried on with the following materials and/or processed:

- Spaceloft (super-insulator) with e = mm
- PVC: ρc = 1.40x10^{6} J.m^{-3} (measured by DSC) ; λ = 0.184 W.m^{-1}.K^{-1} (measured by the tiny hot plate method [10]).

The analysis of the residuals may lead in some cases to use a modified model taking into account a parallel or a series thermal resistance.

6.7 References

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