Inverse Problem and a New Device to Estimate Thermal Conductivity of Composite Phase Change Material

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Abstract: A new experimental device has been developed in order to characterize the phase change material (PCM) thermal properties (thermal conductivity k, sensible and latent heat thermal energy storage, cp and Lf) in the solid phase, during the solid-liquid transition and in the liquid phase. It allows to measure cylindrical samples of maximum 60 mm radius and 10 mm thick. A typical measurement consists in imposing a vertical temperature gradient through the PCM sample driven by a heat source, monitoring during the experiment time all the boundary conditions (temperatures and heat fluxes) and measuring temperature evolution in three locations within the PCM sample. In this work, we will focus only on the solid thermal conductivity characterization. These experiment data are used to solve the inverse heat conduction problem by applying the conjugate gradient method and finally, to determine the PCM thermal properties. Two types of composite PCM have been thermally characterized: paraffin mixed with synthetic graphite (Timrex SFG75) and paraffin mixed with graphite waste.

Keywords: inverse problem, PCM composite, thermal conductivity.

1. Introduction

Due to the current energy crisis, the improvement of energy efficiency systems becomes more and more crucial for reducing fossil fuel consumption and CO2 emissions. One of the major research areas is dedicated to thermal energy storage (TES) systems or materials [1]. Indeed, energy savings are achieved if TES systems are used as energy provisions. They are extremely helpful especially when the supply of and demand for thermal energy do not occur in the same time. Many surveys are devoted to demonstrate the potential and feasibility of latent heat energy storage [2], [4].

In this regard, the phase-change materials (PCM) are attractive solutions because they can change their state (usually solid-liquid transitions) at relatively low temperatures while absorbing or releasing high amounts of heat [4]. Consequently, they have the ability to store energy in a narrow temperature range. Another advantage of PCM is their high-energy storage density due to the latent heat. For instance, melting 1 m3 of ice needs 84 kWh whereas heating up 1 m3 of liquid water by 1°C takes 1 kWh. In order to evaluate the opportunities for exploiting PCM in industrial applications, it is usual to perform numerical simulations describing and predicting the thermal system behavior. However, the numerical calculations require the thermal material properties as inputs. Their reliability depends strongly on the input accuracy. Consequently, a reliable experimental method is necessary to measure the PCM thermal properties with precision. As stated in [5], three widely used groups of methods are used to characterize PCM thermal properties: conventional calorimetry methods, differential thermal analysis (DTA) and differential scanning calorimetry (DSC). However, all these methods involve very small samples that can be significantly influenced by local heterogeneities. Another method, the T-history method, has been proposed in literature. A large sample size of organic, inorganic, encapsulated or composed PCM which can be measured by the T-history method. But, one important drawback is remained: the sample tested has to be homogeneous. For investigation of PCMs in real conditions, it is essential to design an experimental device to measure PCM at large scale. Recently PCM composite characterization was performed by Karkri et al. [1]–[5] using two different experimental techniques: thermal energy storage properties, such as latent heat and heat capacities, were investigated using a Transient Guarded Hot Plate Technique (TGHPT), whereas thermal conductivities and diffusivities were measured using a periodic temperature method. However, the previous works highlight the impact of PCM incorporation into building materials on their energy performance and do not focus exclusively on the only MCP. Another motivation justifies the design of a reliable experimental method to characterize PCM thermal properties. It can be a very helpful tool to perform pertinently thermal optimization of PCM by influencing their composition. Indeed, paraffin suffers from a low thermal conductivity (0.21-0.24 W.m-1.K-1). High thermal conductivity of both phases is a crucial thermodynamic criterion, so that the temperature gradients required for charging and discharging the storage material are small. Consequently, more working effort has been focused to improve the PCM thermal conductivity, by dispersing high conductive particles within the PCM [3]. Moreover, graphite particles have strong resistance to corrosion and chemical attacks which makes it compatible with most PCM. In this context, the new experimental method will be perfectly suited to control the optimization of PCM thermal conductivity. In the first section, the new experimental device capable of characterizing the PCM thermal properties will be described. In the second section, numerical studies will be lead to demonstrate how the PCM thermal properties can be determined by solving the inverse heat conduction problem (IHCP) with the gradient conjugate method [6] using the thermal measurement data from the new experimental device. In the third, two different PCM composites have been characterized by the new device and the results will be presented and discussed. In this work, we will focus only on the solid thermal conductivity characterization

2. Experimental device

A new experimental device has been developed in order to measure thermal properties of PCM composites. Figure 1 shows an overview of the experimental device. The device was designed in aluminium in order to conduct the heat flux upward and produce a significant temperature gradient through the PCM sample. The piston support (the "head") may be considered as a thermal fin which evacuates the heat. The whole cylindrical part of the measurement device has been isolated with mineral wool in order to reduce drastically thermal losses. The cavity bottom is maintained at a fixed temperature by a heating element and a coil heat exchanger in which an isothermal fluid is circulated by a thermo-regulated bath. The temperature and the heat flow below and above the PCM samples are respectively measured by thermocouples and heat flux meter and recorded by a LabView© application. The thermocouples have been calibrated and the heat flux meters have been calibrated by the manufacturer (the sensibility are 74074 $W.m^{-2}.V^{-1}$ and 129366 $W.m^{-2}.V^{-1}$ for the bottom and the upper heat flux sensors respectively). A typical measurement consists in imposing a vertical temperature gradient through the sample driven by a heat element, monitoring during the experiment time all the boundary conditions (temperatures T_1, T_2 and heat fluxes φ_1, φ_2) and measuring temperature evolution in 2 locations Y_1 and Y_2, in the thickness sample.

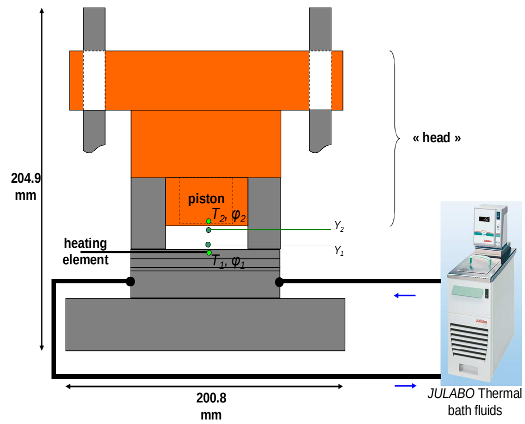


Figure 1. The experimental device

3. The conjugate gradient method

The conjugate gradient method is a straightforward and powerful iterative technique for solving linear and nonlinear inverse problems of parameter estimation. In the iterative procedure of the conjugate gradient method, at each iteration a suitable step size α is taken along a direction of descent w in order to minimize the objective function J. The direction of descent w is obtained as a linear combination of the negative gradient direction $-\nabla J^{(i)}$ at the current iteration with the direction of descent of the previous iteration $-\nabla J^{(i-1)}$. The linear combination is such that the resulting angle between the direction of descent w and the negative gradient direction $-\nabla J$ is less than 90° and the minimization of the objective function

J is assured. The application of the conjugate gradient method to estimate thermal parameters by solving the inverse heat conduction problem requires the computation of the following. The direct problem, adjoint and sensitivity problems [6]. The solution of the IHCP is obtained when the functional $J(k)$ is minimized :

$$J(k)=\sum_{i=1}^{M}\left\|T(x_i,y_i,z_i,t)-Y_i(x_i,y_i,z_i,t)\right\|^2 \qquad (1)$$

where $T(x_i,y_i,z_i,t)$ is the solution of the direct problem at (x_i,y_i,z_i), $Y_i(x_i,y_i,z_i,t)$ is the temperature measured by thermocouples located at (x_i,y_i,z_i) and M is the number of thermocouples.

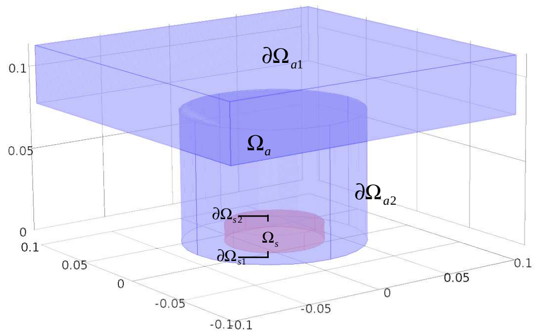


Figure 2. The heat transfer model geometry.

Figure 2 shows the geometry using for the modeling. The dimensions of the model geometry match those of the experimental device (Figure 1). Ω_s is the domain defining the sample volume, $\partial\Omega_{s1}$ and $\partial\Omega_{s2}$ are respectively the bottom and upper boundaries. Ω_a is the domain defining the aluminum volume, $\partial\Omega_{a1}$ and $\partial\Omega_{a2}$ delimit respectively the "head" and the "piston" boundaries. The direct problem is formulated as follows :

<u>In the sample:</u>

$$(\rho c_p)_s \frac{\partial T(x,y,z,t)}{\partial t}=\vec{\nabla}\cdot\left(k_s(T)\vec{\nabla}T(x,y,z,t)\right)$$
$$\forall(x,y,z)\in\Omega_s, \forall t\in\left]0,t_f\right] \qquad (2)$$

$$T(x,y,z,t)=T_2(t) \qquad \forall(x,y,z)\in\Omega_{s2} \qquad (3)$$

$$-k_s(T)\vec{\nabla}T(x,y,z,t)\cdot\vec{n}=\varphi_1(t) \quad \forall(x,y,z)\in\partial\Omega_{s1} \qquad (4)$$

$$T(x,y,z,t)=T_{0s}(x,y,z) \quad \forall(x,y,z)\in\Omega_s, t=0 \qquad (5)$$

In the aluminum:

$$(\rho c_p)_a \frac{\partial T(x,y,z,t)}{\partial t}=\vec{\nabla}\cdot\left(k_a(T)\vec{\nabla}T(x,y,z,t)\right)$$
$$\forall(x,y,z)\in\Omega_a, \forall t\in\left]0,t_f\right] \qquad (6)$$

$$-k_a(T)\vec{\nabla}T(x,y,z,t)\cdot\vec{n}=h_c\left(T(x,y,z,t)-T_{ext}\right)$$
$$\forall(x,y,z)\in\Omega_{a1} \qquad (7)$$

$$\vec{\nabla}T(x,y,z,t)\cdot\vec{n}=0 \qquad \forall(x,y,z)\in\partial\Omega_{a2} \qquad (8)$$

$$T(x,y,z,t)=T_{0a}(x,y,z) \quad \forall(x,y,z)\in\Omega_a, t=0 \qquad (9)$$

At the aluminum/sample interfaces, the continuity in temperatures and fluxes has been ensured. The convective transfer coefficient h_c is equal to 8 $W.m^{-2}.K^{-1}$. It is a common value for indoor natural convection. T_{ext} is supposed to be constant during the simulation duration and it is set to 22 *°C*. The initial temperature is supposed to be uniform over the entire geometry: $T_{0s}(x,y,z)=T_{0a}(x,y,z)=T_0$

The sensitivity problem

In order to derive the sensitivity problem for $k(T)$, we should perturb $k(T)$: it is assumed that when $k(T)$ undergoes a variation $\delta k(T)$, $T(x,y,z,t)$ is perturbed by $T+\delta T$. Then, replacing $k(T)$ by $k(T)+\delta k(T)$ and T by $T+\delta T$ in the direct problem, and subtracting from the resulting expressions the direct problem and neglecting the second-order terms, the following sensitivity problem for the sensitivity function δT is obtained :

In the sample:

$$(\rho c_p)_s \frac{\partial \delta T(x,y,z,t)}{\partial t}=$$
$$\vec{\nabla}\cdot\left(k_s(T)\vec{\nabla}\delta T(x,y,z,t)\right)+\delta k\vec{\nabla}T^2(x,y,z,t) \qquad (10)$$
$$\forall(x,y,z)\in\Omega_s, \forall t\in\left]0,t_f\right]$$

$$\delta T(x,y,z,t)=0 \qquad \forall(x,y,z)\in\Omega_{s2} \qquad (11)$$

$$-k_s(T)\vec{\nabla}\delta T(x,y,z,t)\cdot\vec{n}=\vec{\nabla}T(x,y,z,t)\cdot\vec{n}$$
$$\forall(x,y,z)\in\partial\Omega_{s1} \qquad (12)$$

$$\delta T(x,y,z,t)=0 \quad \forall(x,y,z)\in\Omega_s, t=0 \qquad (13)$$

In the aluminum:

$$(\rho c_p)_a \frac{\partial \delta T(x,y,z,t)}{\partial t}=\vec{\nabla}\cdot\left(k_a(T)\vec{\nabla}\delta T(x,y,z,t)\right)$$
$$\forall(x,y,z)\in\Omega_a, \forall t\in\left]0,t_f\right] \qquad (14)$$

$$-k_a(T)\overrightarrow{\nabla\delta T}(x,y,z,t)\cdot\vec{n}=h_c\delta T(x,y,z,t)$$

$\forall(x,y,z)\in\Omega_{a1}$ (15)

$$\overrightarrow{\nabla \delta T}(x,y,z,t)\cdot\vec{n}=0 \quad \forall(x,y,z)\in\partial\Omega_{a2} \qquad (16)$$

$$\delta T(x,y,z,t)=0 \quad \forall(x,y,z)\in\Omega_a, t=0 \qquad (17)$$

At the aluminium/sample interfaces, the continuity in temperatures and fluxes has been ensured. The relation between the search step size α and the sensitivity function δT is given by the equation:

$$\alpha=\frac{\int_0^{t_f}\sum_{i=1}^{M}\left[T(x_i,y_i,z_i,t)-Y_i(x_i,y_i,z_i,t)\right]\delta T(x_i,y_i,z_i,t)\,dt}{\int_0^{t_f}\sum_{i=1}^{M}\left[\delta T(x_i,y_i,z_i,t)\right]^2 dt} \qquad (18)$$

The adjoint problem

The Lagrangian multiplier $\psi(x,y,z,t)$ and the Lagrangian $L(T,k,\psi)$ are introduced [6] :

In the sapmle :

$$(\rho c_p)_s\frac{\partial\psi(x,y,z,t)}{\partial t}+\vec{\nabla}\cdot\left(k_s(T)\vec{\nabla}\psi(x,y,z,t)\right)= \sum_{i=1}^{M}2\left[Y_i(x_i,y_i,z_i,t)-T(x_i,y_i,z_i,t)\right]\delta(x-x_i)\delta(y-y_i)\delta(z-z_i) \qquad (19)$$

$$\psi(x,y,z,t)=0 \quad \forall(x,y,z)\in\Omega_{s2} \qquad (20)$$

$$\vec{\nabla}\psi(x,y,z,t)\cdot\vec{n}=0 \quad \forall(x,y,z)\in\partial\Omega_{s1} \qquad (21)$$

$$\psi(x,y,z,t)=0 \quad \forall(x,y,z)\in\Omega_s, t=t_f \qquad (22)$$

In the aluminum:

$$(\rho c_p)_a\frac{\partial\psi(x,y,z,t)}{\partial t}+\vec{\nabla}\cdot\left(k_a(T)\vec{\nabla}\psi(x,y,z,t)\right)=0$$

$$\forall(x,y,z)\in\Omega_a \qquad (23)$$

$$-k_a\vec{\nabla}\psi(x,y,z,t)\cdot\vec{n}=h_c\psi(x,y,z,t) \quad \forall(x,y,z)\in\Omega_{a1} \qquad (24)$$

$$\vec{\nabla}\psi(x,y,z,t)\cdot\vec{n}=0 \quad \forall(x,y,z)\in\partial\Omega_{a2} \qquad (25)$$

$$\psi(x,y,z,t)=0 \quad \forall(x,y,z)\in\Omega_a, t=t_f \qquad (26)$$

At the aluminum/sample interfaces, the continuity in temperatures and fluxes has been ensured. The adjoint problem is different from the standard initial value problems in that the final time condition at time $t=t_f$ is specified instead of the customary initial condition. Consequently, the resolution must be done in a retrograde way.

3.1. Model validation

This preliminary study aims to validate the direct problem (equations 2-9) using reference material (Table1) and the boundary condition $Y_1(t)$ and $Y_2(t)$ measured respectively by the lower and upper thermocouples inside the reference material. The temperature evolutions inside the material have been compared to those measured using the new device (Figure 3). We observe that the model temperatures fit correctly the experimental data: they stay inside the measurement error bars. We can conclude that the model is adapted to fit accurately the heat transfer phenomena occurring inside the experimental device.

	Density $\rho\left[kgm^{-3}\right]$	Heat capacity $c_p\left[Jkg^{-1}K^{-1}\right]$	Thermal conductivity $k\left[Wm^{-1}K^{-1}\right]$
Aluminum	2700	910	230
Paraffin	896	1987	0.233

Table 1: Aluminum and paraffin Thermal properties.

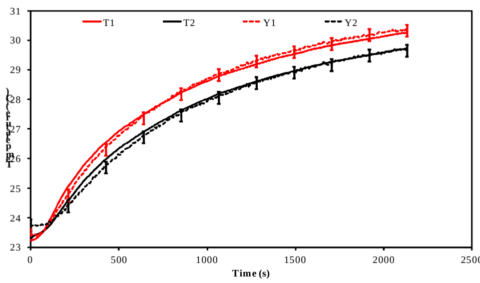


Figure 3. Model temperatures versus experimental temperatures with the error bars

3.2 The Conjugate Gradient Algorithm

A coupling between COMSOL Multiphysics® and MATLAB has been performed in order to solve the 3D inverse heat conduction problem by the conjugate gradient method. COMSOL Multiphysics® will be used to solve the direct problem, the adjoint problem and compute the gradient ∇J, and finally solve the sensitivity problem and compute the search step size α.

The algorithm is implemented in MATLAB and its structure is shown as follows [6].

3.3 Validation of the methodology

In order to validate the methodology (i.e. the coupling of MATLAB and COMSOL Multiphysics®), numerical experiments have been performed. The output data $Y_i(x_i,y_i,z_i,t)$ are first computed by the direct problem with a known ("target") thermal conductivity $k(T)$ over the time interval $[0, t_f]$. Then, these output data have been used to solve the inverse heat conduction problem by the conjugate gradient algorithm and identify the thermal conductivity. We will determine the thermal conductivity assuming that it depends on temperature. Two cases have been considered ($k(T)=a+bT$ (a) and $k(T)=ae^{\frac{T-27315}{b}}$ (b)).

a) Linearly temperature dependence

The target linear coefficients are set to: $a=0.5$ and $b=0.001$. The stopping criterion for the conjugate gradient algorithm is $\varepsilon=10^{-7}$ K^2. Figure 4 represents the convergence history. The number of iterations to reach the stopping criterion is 417. The model temperatures fit perfectly the output data, after convergence (Figure 5). The value of a and b obtained after the convergence are $a=0.499$ and $b=0.001002$. We observe that these values are very close to the target values.

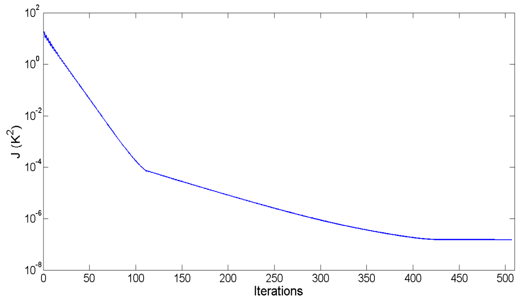


Figure 4. Evolution of the functional J over the iterations (linear case)

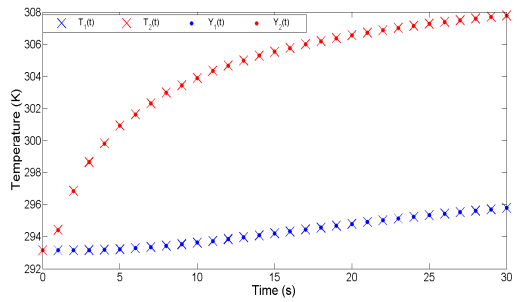


Figure 1. Model temperatures versus output data (linear case)

b) Exponential temperature dependent

The target exponential coefficients are set to: $a=0.5$ and $b=20$. The stopping criterion for the conjugate gradient algorithm is $\varepsilon=10^{-7}$ K^2. Figure 6 represents the convergence history. The number of iterations to reach the stopping criterion is 698. The model temperatures fit perfectly the output data, after convergence (Figure 7). The value of a and b obtained after the convergence are $a=0.499$ and $b=19.999$. We observe that these values are very close to the target values.

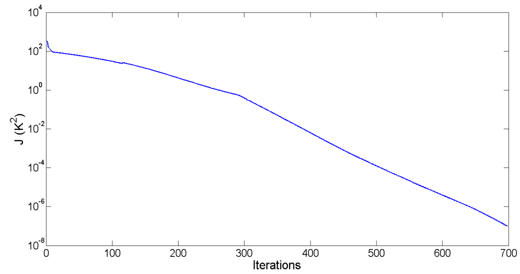


Figure 6. Evolution of the functional J over the iterations (exponential case)

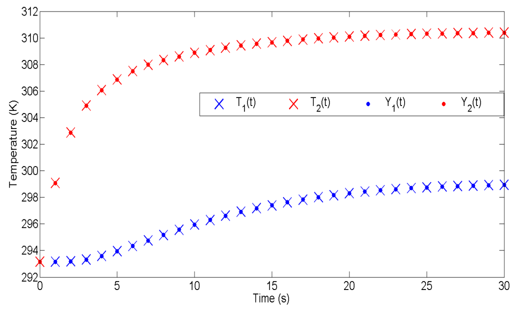


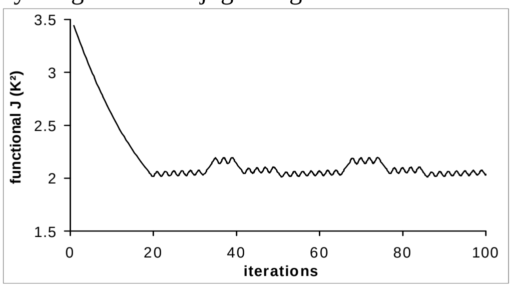
Figure 2. Model temperatures versus output data (exponential case)

4. Experimental results and discussions

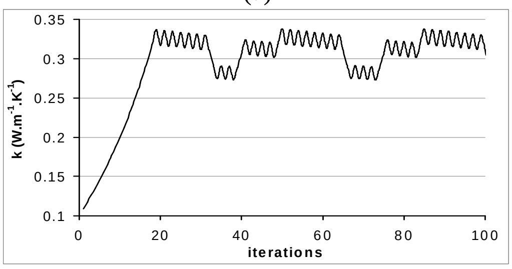
In the present work, the PCM to be characterized is a paraffin with melting temperature of 56-58 °C and with specific density of 900 $kg.m^{-3}$. The thermal conductivity is enhanced by addition of conductive graphite particles. Two different kinds of graphite were used in this study. One type is an industrial graphite "graphite waste". It comes from damaged tubular graphite heat exchangers [7]. The measured bulk density is 1936 $kg.m^{-3}$ with an average size of 85 μm. The second kind is the Timrex (SFG75) powder supplied by Timcal Graphite & Carbon at a bulk density of 2240 $kg.m^{-3}$. It is synthetic graphite with spherical shape and an average size of 75 μm, characterized by a well-aligned crystal structure and by a high thermal conductivity of the basal plane [8]. The elaboration method of the PCM composite is based on the cold uniaxial compression [8]. This technique leads to an anisotropic composite structure whose porosity is partially occupied by paraffin grains. The thickness and the diameter of all this specimens were 10mm and 60mm respectively (Figure 8).

Figure 8. Example of paraffin/graphite composite samples: (a) paraffin, (b) paraffin/graphite.

An experimental measurement on a PCM composite paraffin/graphite waste (5% wt. graphite waste) has been conducted by using our new experimental device. 2 thermocouples Y_1 and Y_2 have been inserted in 2 locations along the sample thickness. In this study, we assume that the thermal conductivity k is independent of the temperature. Figure 9 plots the convergence history and the evolution of k over the iterations by the gradient conjugate algorithm.



(a)



(b)

Figure 9. Evolution of the functional J (a) and the parameter k (b) over the iterations

The functional J reaches its minimum after 20 iterations. As previously stated, because of data noise, the minimum of the functional J does not have to be equal to zero. The standard deviation of the measurements can be evaluated to $\sigma = 0.09\,K$. The thermal conductivity oscillates around a mean value $k = 0.318$ $W.m^{-1}.K^{-1}$. Figure 10 plots temperature measurements Y_1 and Y_2 versus modeling temperatures T_1 and T_2 obtained by taking the inversion solution $k = 0.318$ $W.m^{-1}.K^{-1}$.

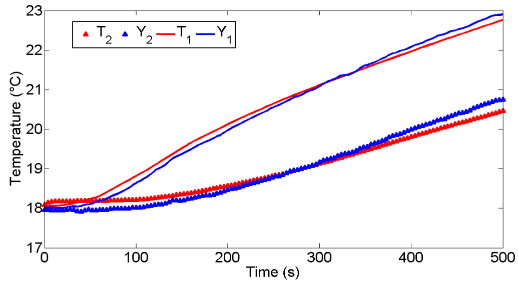


Figure 10. Model temperatures versus experimental data (paraffin/5% wt. graphite waste)

The modeling temperatures fit correctly the experimental data after convergence. A measurement of the same PCM composite performed by another experimental method [8] gives: $k = 0.289$ $W.m^{-1}.K^{-1}$.

Another experimental measurement on a PCM composite paraffin/SFG75 graphite (40% wt. SFG75 graphite) has been conducted. Figure 11 plots the convergence history and the evolution of k over the iterations by the gradient conjugate algorithm.

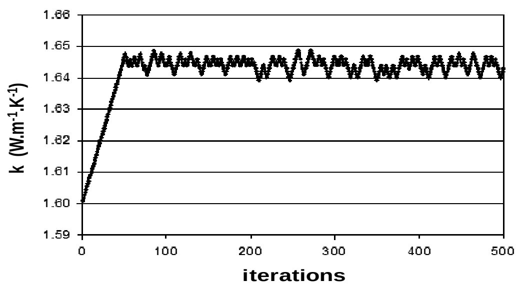


Figure 11. Evolution of the functional J and the parameter k over the iterations

The functional J reaches its minimum after 100 iterations. The standard deviation of the measurements can be evaluated to $\sigma = 0.02\,K$. The thermal conductivity oscillates around a mean value $k = 1.648$ $W.m^{-1}.K^{-1}$. Figure 12 plots temperature measurements Y_1 and Y_2 versus modeling temperatures T_1 and T_2 obtained by taking the inversion solution $k = 1.648\, W.m^{-1}.-K^{1}$.

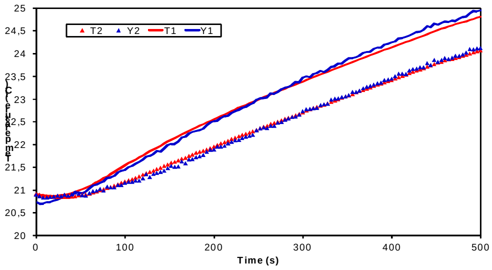


Figure 12. Model temperatures versus experimental data (paraffin/40% wt. SFG75 graphite)

The modeling temperatures fit correctly the experimental data after convergence. As stated in [7], thermal conductivity measurements have been performed on paraffin/SFG75 graphite composites with mass fraction of graphite varying from 0 to 20 wt.%. Although the authors did not make the measurement for 40% wt. SFG75 graphite, the thermal conductivity determined by the present experimental method follows the trend already established in [8].

From these experimental observations, we can conclude that our experimental device and the conjugate gradient method provide correct value of thermal conductivity.

5. Conclusions

A new experimental device to characterize the PCM thermal properties has been presented. A methodology consisting in coupling COMSOL Multiphysics® and MATLAB to determine the thermal conductivity $k(T)$ by solving inverse heat conduction problem with the conjugate gradient algorithm has been validated. The reliability and robustness of the methodology has been demonstrated through numerical experiments. Two PCM composites (paraffin/5% wt. graphite waste and paraffin/40% wt. SFG75 graphite) have been characterized by the new experimental device. The experimental results show that the experimental device measures correct values of thermal conductivity and the reliable applicability of the new device has been confirmed.

6. References

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