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Article

An enthalpy method for heat conduction in tube containing phase change material

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Abstract: We present an innovative enthalpy method for determining the thermal properties of phase change materials (PCM). The enthalpy-temperature relation in the “mushy” zone is modelled by means of a fifth order Obreshkov polynomial with continuous first and second order derivatives at the zone boundaries. The partial differential equation (PDE) for the conduction of heat is rewritten so that the enthalpy variable is not explicitly present, rendering the equation nonlinear. The thermal conductivity of the PCM is assumed to be temperature dependent and is modelled by a fifth order Obreshkov polynomial as well. The method has been applied to lauric acid, a standard prototype. The latent heat and the conductivity coefficient, being the model parameters, were retrieved by fitting the measurements obtained through a simple experimental procedure. Therefore, our proposal may be profitably used for the study of materials intended for heat-storage applications.

Keywords: enthalpy; moving boundaries; phase-change materials; nonlinear optimization; thermal conductivity; latent heat

1. Introduction

Phase Change Materials (PCMs), have been widely used in several applications [1–5] that exploit the released (or absorbed) energy during a phase transition. PCMs allow for latent heat utilization and have attracted the keen interest of the heat-storage engineering community. Naturally, the determination of the PCM thermal properties is of major importance for designing effective thermal depositories. Although the heat transfer equation could in principle describe accurately the charging (or discharging) process of a PCM, the temperature activated phase change, renders the problem highly nonlinear and its solution non trivial. This in turn, incommodes the determination of the specific and latent heat as well as of the thermal conductivity, i.e., the crucial parameters for the design of an effective heat-storage system. Indeed, significant effort has been devoted in developing techniques for the reliable determination of PCMs’ thermal properties, the most common ones being the Differential Scanning Calorimetry (DSC) [6], the T-History method [7] and its variants [8–12]. Nevertheless, these methods suffer from several problems regarding reproducibility, accuracy and robustness [13], qualities that become even more important in the case of mixed PCMs that are widespread in practical applications. In particular, the determination of PCM’s thermal conductivity, in the full operational temperature range, still remains a challenge.

The aim of the present work is to provide a framework facilitating the reliable determination of PCM's thermal properties, by means of simple experiments coupled to an analysis based on a theoretical model.

In the following, we describe the simple experimental setup, the associated heat conduction equation along with the proper boundary and initial conditions, and we detail the employed numerical technique.

2. Description of the approach

The procedure we follow consists of the following steps: *i*) The PCM is initially prepared to be in liquid state at a predefined temperature. Next, it is cooled by immersion in a heat-bath that is maintained at a lower temperature, and standard T-History measurements are performed to obtain the enthalpy—temperature curve within a temperature range where the liquid solidifies and is further cooled down to the heat-bath's temperature. From this curve we determine the liquid and solid regions, and using simple energy considerations (described by Yinping et al. and Hong et al. [7,8]), we obtain the corresponding specific heats for each region, c_p^l and c_p^s, by fitting the theoretical predictions to the experimental curves, provided that the corresponding densities are known. ii) From the obtained temperature versus time curves in the liquid and solid regions, and using the determined specific heats, we deduce the corresponding thermal conductivities, k_l and k_s. iii) The latent heat is calculated using the proposed models that are described below.

2.1. Thermal conductivity model, and latent heat enthalpy-based model

We consider a system of a long glass-tube containing a Phase Change Material (PCM). The tube's inner and outer radii are respectively denoted by R_1 and R_2 in **Figure 1**. Let T_l and T_s be the PCM's liquidation and solidification temperatures. The system is brought initially at a temperature $T_0 > T_l$, so that the PCM is in liquid state, and subsequently is immersed in a heat bath maintained at a constant temperature $T_1 < T_s$.

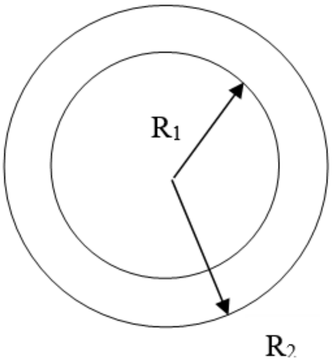

Figure 1. Geometrical representation of the model system.

The temperature "zone" $T \in [T_s, T_l]$, is referred to as the "mushy" zone, where the material is in a "mixed" state, and it is in fact the temperature region where the latent heat is released upon cooling, and therefore, it will be used for its determination. The temperature depends on both time and position. The position is measured from the tube's axis, and is denoted by r. Due to cylindrical symmetry there is no angular dependence, and since the tube is long compared to its radial extent, z-dependence may be neglected.

At $r = R_2$, the temperature is known to be $J(t), \forall t > 0$. Let the subscripts p and g refer to PCM and glass quantities correspondingly. Then, the governing equations for heat conduction may be written as:

$$\rho_p \frac{\partial H_p(r,t)}{\partial t} = k_p(T)\left(\frac{\partial^2 T(r,t)}{\partial r^2} + \frac{1}{r}\frac{\partial T(r,t)}{\partial r}\right) + \frac{dk_p(T)}{dT}\left(\frac{\partial T(r,t)}{\partial r}\right)^2, \quad \mathrm{r} \in [0, R_1] \tag{1}$$

$$\rho_g c_g \frac{\partial T(r,t)}{\partial r} = k_g\left(\frac{\partial^2 T(r,t)}{\partial r^2} + \frac{1}{r}\frac{\partial T(r,t)}{\partial r}\right), \quad \mathrm{r} \in [R_1, R_2] \tag{2}$$

where $H_p(r,t)$ is the enthalpy function of the PCM. Let the quantities c_p^s, c_p^l, L denote its specific heats in the solid and liquid phases and the latent heat per unit mass.

As the apparatus is immersed in the heat-bath, consisting of a tank filled with water, the temperature on the outer glass surface of the tube is not constant for a considerable amount of time. Hence this temperature $T(R_2,t)$ is measured and recorded. Also the temperature at the tube's axis, $T(0,t)$ is measured and recorded as well. Since analytical solutions for such a system do not exist, we will resort to numerical approximate solutions.

Heat conduction through PCMs is a highly nonlinear "moving boundary" problem. The boundary that is moving is the interface between the solid and liquid phase and on which special conditions must be satisfied.

"Front-tracking" methods, monitor the motion of the solid-liquid interface and require the satisfaction of the associated conditions on this moving boundary. A discretization grid is employed, and since the interface position will not always fall on a grid point, either interpolation, or a variable time step, or even a time-dependent grid is employed to accommodate this requirement. These approaches are complicated, have accuracy issues, and their implementation is quite cumbersome if feasible at all.

"Front-fixing" methods apply a variable transformation to immobilize the interface in the new coordinate system. However, the arising equations are even more complicated.

The enthalpy method is a "fixed-domain" method based on a reformulation of the heat conduction equation adopting a model for the enthalpy-temperature relationship. In this approach the solid-liquid interface does not explicitly appear and hence the difficulties due to the moving boundary are avoided. Based on this idea a variety of similar methods have been developed. We have chosen the enthalpy approach because it simplifies the numerical work and provides a direct modelling interpretation. The purpose of this study is to explore the possibility to reliably deduce PCM's properties (i.e., latent heat, thermal conductivity, specific heat, critical temperature, etc.) from the available measurements alone.

The enthalpy may be modelled as:

$$H_p(T)=\begin{cases} c_p^s T, & if\ T<T_s \\ \mathcal{H}_p(T), & if\ T_s\le T\le T_l \\ c_p^l(T-T_l)+c_p^s T_s+\mathrm{L}, & if\ T>T_l \end{cases} \tag{3}$$

where $H_p(T)$ is a fifth order polynomial in T [14], so as to match the enthalpy and its first and second derivatives at the endpoints of the mushy zone: $T\in[T_s,T_l]$.

$$H_p(T)=c_p^s T+A_3\left(\frac{T-T_s}{T_l-T_s}\right)^3+A_4\left(\frac{T-T_s}{T_l-T_s}\right)^4+A_5\left(\frac{T-T_s}{T_l-T_s}\right)^5 \tag{4}$$

Letting $\Delta=T_l-T_s$, one obtains:

$$A_3=10L-\Delta\left(6c_p^s+4c_p^l\right) \tag{5}$$

$$A_4=-15L+\Delta\left(8c_p^s+7c_p^l\right) \tag{6}$$

$$A_5=6L-3\Delta\left(c_p^s+c_p^l\right) \tag{7}$$

The derivative of the model function is given by:

$$Q_p(T)\equiv\frac{dH_p(T)}{dT}=\begin{cases} c_p^s, & if\ T<T_s \\ \dfrac{d\mathcal{H}_p(T)}{dT}, & if\ T_s\le T\le T_l \\ c_p^l, & if\ T>T_l \end{cases} \tag{8}$$

Note that in Equation (1) we can substitute: $\frac{\partial H_p(r,t)}{\partial t}=\frac{dH_p(T)}{dT}\frac{\partial T(r,t())}{\partial t}$

Therefore Equation (1) may be rewritten as:

$$\rho_p\frac{\partial T(r,\mathrm{t})}{\partial t}=\frac{k_p(T)}{Q_p(\mathrm{T})}\left(\frac{\partial^2 T(r,\mathrm{t})}{\partial t^2}+\frac{1}{r}\frac{\partial T(r,\mathrm{t})}{\partial r}\right)+\frac{1}{Q_p(T)}\frac{dk_p(T)}{\mathrm{dT}}\left(\frac{\partial T(r,\mathrm{t})}{\partial r}\right)^2,\forall r\in\left[0,R_1\right] \tag{9}$$

Now for the conductivity as a function of the temperature we use a fifth order polynomial for $T\in(T_s,T_l)$ that is continuous at the mushy zone endpoints along with its first and second derivatives. Namely:

$$k_p(T)=\begin{cases} k_p^s, ifT<T_s \\ K_p(T), ifT_s\le T\le T_l \\ k_p^l, ifT>T_l \end{cases} \tag{10}$$

With

$$K_p(T)=k_p^s+\left(k_p^l-k_p^s\right)\left[10\left(\frac{T-T_s}{T_l-T_s}\right)^3-15\left(\frac{T-T_s}{T_l-T_s}\right)^4+6\left(\frac{T-T_s}{T_l-T_s}\right)^5\right] \tag{11}$$

Adopting the following definitions:

$$f(T)\equiv\frac{k_p(T)}{\rho_p Q_p(T)}and g(T)\equiv\frac{dk_p(T)}{dT}\frac{1}{\rho_p Q_p(T)} \tag{12}$$

Equation (9) may be rewritten as:

$$\frac{\partial T(r,\mathrm{t})}{\partial t} = f(T)\left(\frac{\partial^2 T(r,\mathrm{t})}{\partial r^2} + \frac{1}{r}\frac{\partial T(r,\mathrm{t})}{\partial r}\right) + g(T)\left(\frac{\partial T(r,\mathrm{t})}{\partial r}\right)^2, \forall r \in [0, R_1] \quad (13)$$

2.2. Initial, boundary, and interface conditions

At the origin, due to azimuthal symmetry, the following Neumann condition holds:

$$\frac{\partial T(r,t())}{\partial r}|_{r=0}\Bigg| \quad (14)$$

At the PCM-glass interface, continuity of temperature and heat-flux requires that:

$$T(R_1 - \varepsilon, t) = T(R_1 + \varepsilon, t) \quad (15)$$

$$k_p(T)\frac{\partial T(r,t())}{\partial r}|_{r=R_1-\varepsilon}\, k_g(T)\frac{\partial T(r,t())}{\partial r}|_{r=R_1+\varepsilon}\Bigg\|\quad (16)$$

At the outer tube boundary, we have:

$$T(R_2, t) = J(t) \quad (17)$$

And initially, i.e., at t = 0:

$$T(r,0) = \theta_0, \forall \in [0, \mathrm{R}_2] \quad (18)$$

2.3. Discretization

Let $n+1$ points $r_i, i = 0,1,\dots,n$ be the grid for $r \in [0, R_1]$ which is the space filled by the PCM, and let $m+1$ points $r_{n+j}, j = 0,1,\dots,m$ be the grid over the glass part, i.e., for $r \in [R_1, \mathrm{R}_2]$.

$$r_i = ih \equiv \frac{i}{n}R_1, \forall i = 0,1,\dots,n \quad (19)$$

$$r_{n+j} = R_1 + j\delta \equiv R_1 + \frac{j}{m}(R_2 - R_1), \forall j = 0,1,\dots,m \quad (20)$$

Space derivatives are estimated by central differences as:

$$\frac{\partial T(r,t)}{\partial r} \approx \frac{T(r+s,t) - T(r-s,t)}{2s} \quad (21)$$

$$\frac{\partial^2 T(r,t)}{\partial r^2} \approx \frac{T(r+s,t) + T(r-s,t) - 2T(r,t)}{s^2} \quad (22)$$

And the time derivative by forward differences as:

$$\frac{\partial T(r,t)}{\partial t} \approx \frac{T(r,t+\tau) - T(r,t)}{\tau} \quad (23)$$

In order to solve Equations (2) and (9), we have chosen the implicit and unconditionally stable Crank-Nicolson scheme. We use the following notation:

$$T_i^j = T(r_i, j\tau), f_i^j = f\left(T_i^j\right), g_i^j = g\left(T_i^j\right), \text{ and } a_g = \frac{k_g}{\rho_g c_g}$$

At $r \to 0, \frac{\partial^2 T(r,t)}{\partial r^2} + \frac{1}{r}\frac{\partial T(r,t)}{\partial r} = \frac{4}{h^2}\left(T\left((h,t) - T(0,t)\right)\right) = \frac{4}{h^2}\left(T_1^j - T_0^j\right)$ for $t = j\tau$

The Crank-Nicolson scheme yields [15,16]:

$$T_i^{j+1} = T_i^j + \frac{\tau}{2}\left[f\left(T_i^{j+1}\right)\nabla^2 T_i^{j+1} + f\left(T_i^j\right)\nabla T_i^j\right] + \frac{\tau}{2}\left[g\left(T_i^{j+1}\right)\left(\frac{\partial T_i^{j+1}}{\partial r}\right)^2 + g\left(T_i^{j+1}\right)\left(\frac{\partial T_i^{j+1}}{\partial r}\right)^2\right], \forall i \in [0, n] \tag{24}$$

$$T_i^{j+1} = T_i^j + \frac{\tau a_g}{2}\left[\nabla^2 T_i^{j+1} + \nabla^2 T_i^j\right], \forall i \in [n, n+m] \tag{25}$$

For $i = 0$ Equation (24) may be rewritten as:

$$T_0^{j+1} - \frac{2\tau}{h^2} f_0^{j+1}\left(T_1^{j+1} - T_0^{j+1}\right) = T_0^j + \frac{2\tau}{h^2} f_0^j\left(T_1^j - T_0^j\right) \tag{26}$$

For $i = 1, 2, \ldots, n-1$ Equation (24) takes the form:

$$\begin{aligned}\left(1 + \frac{\tau}{h^2} f_i^{j+1}\right) T_i^{j+1} - \frac{\tau}{2h^2} f_i^{j+1}\left(\left(1 + \frac{1}{2i}\right) T_{i+1}^{j+1} + \left(1 - \frac{1}{2i}\right) T_{i-1}^{j+1}\right) \\ - \frac{\tau}{8h^2} g_i^{j+1}\left(T_{i+1}^{j+1} - T_{i-1}^{j+1}\right)^2 \\ = \left(1 - \frac{\tau}{h^2} f_i^j\right) T_i^j + \frac{\tau}{2h^2} f_i^j\left(\left(1 + \frac{1}{2i}\right) T_{i+1}^j + \left(1 - \frac{1}{2i}\right) T_{i-1}^j\right) \\ + \frac{\tau}{8h^2} g_i^j\left(T_{i+1}^j - T_{i-1}^j\right)^2\end{aligned} \tag{27}$$

For $i = n$ using the interface conditions (15,16) we obtain the relation:

$$k_p\left(T_n^{j+1}\right)\frac{T_n^{j+1} - T_{n-1}^{j+1}}{h} - k_g \frac{T_{n+1}^{j+1} - T_n^{j+1}}{\delta} = 0 \tag{28}$$

For $i = n+1, n+2, \ldots, n+m-1$ and with $x_i = i - n + \frac{R_1}{\delta}$

$$\begin{aligned}\left(1 + \frac{\tau a_g}{\delta^2}\right) T_i^{j+1} - \frac{\tau a_g}{2\delta^2}\left(\left(1 + \frac{1}{2x_i}\right) T_{i+1}^{j+1} + \left(1 - \frac{1}{2x_i}\right) T_{i-1}^{j+1}\right) \\ = \left(1 - \frac{\tau a_g}{\delta^2}\right) T_i^j + \frac{\tau a_g}{2\delta^2}\left(\left(1 + \frac{1}{2x_i}\right) T_{i+1}^j + \left(1 - \frac{1}{2x_i}\right) T_{i-1}^j\right)\end{aligned} \tag{29}$$

And finally for $i = n + m$ we have from the boundary condition Equation (17)

$$T_{n+m}^{j+1} = J\left((j+1)\tau\right) \tag{30}$$

So at each time step, we face a system of $n + m + 1$ nonlinear Equations (26)–(30), for the $n + m + 1$ unknowns, $T_{i=0,n+m}^{j+1}$. The solution procedure is based on nonlinear optimization and minimizes the sum of the squared residuals [17–21].

3. Results

Lauric acid is a well-studied PCM, and we have used it as a benchmark to test the proposed methodology. We have used the presented experimental setup to obtain the necessary measurements (**Figure 2**) and subsequently performed the suggested analysis to estimate the relevant Lauric acid properties.

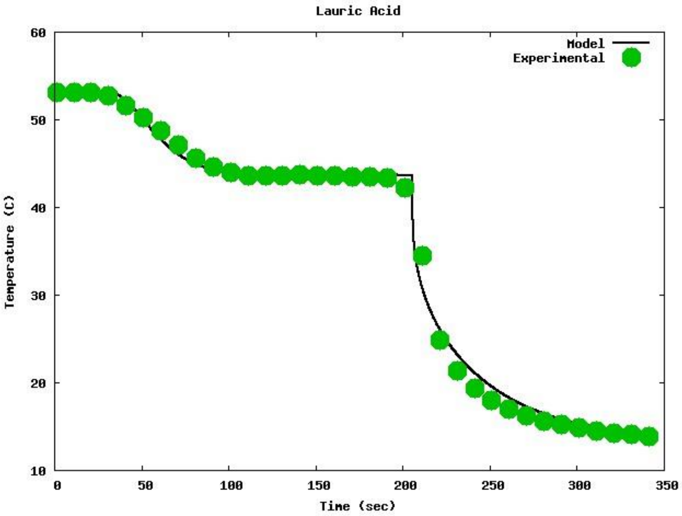

Figure 2. Application of the proposed method for the benchmark case of Lauric acid.

As it can be seen the solution reproduces satisfactorily the experimental data. Once this solution is achieved, Latent Heat, specific Heats and thermal conductivities in the three regions (Liquid, Mushy zone and Solid) are deduced. Our results are summarized in **Table 1**, and are compared to estimates from the relevant literature.

Table 1. Comparison of Lauric acid properties by our model, to values from the literature.

Latent Heat	**Specific Heat Solid Phase**	**Specific Heat Liquid Phase**	**Thermal Conductivity Solid Phase**	**Reference**
$L\ (KJ/Kg)$	$c_p^s\ (KJ/Kg\ K)$	$c_p^l\ (KJ/Kg\ K)$	$k^s\ (W/mK)$	
180 ± 7	2.14 ± 0.11	2.02 ± 0.08	0.23 ± 0.02	Present work
160	-	1.75	0.17	Ref. [7]
186 ± 10	2.81 ± 0.60	2.14 ± 0.46	-	Ref. [8]

4. Conclusions

We have presented a novel enthalpy-based method that provides a holistic description of the temperature evolution of a PCM upon cooling or heating, while the determination of their thermal properties can be deduced by performing two simple experiments. Note that as result, the thermal conductivity coefficients can be evaluated in the three temperature ranges of interest corresponding to liquid, solid and intermediate (mushy) phases. The method has been successfully applied to the Lauric acid prototype, estimating its thermal properties within reasonable error, and consequently it could be useful for the analysis and design of heat storage PCM-based systems.

Highlights

1) PCM thermal properties via simple measurements, coupled to an enthalpy model.
2) Non-linear heat conduction PDE, solved by optimization.
3) Test case: Lauric acid. Determination of thermal conductivity, latent & specific heats.

Author contributions: Conceptualization, GAE and IEL; methodology, IEL; software, IEL and DGP; validation, GAE and CP; formal analysis, IEL; investigation, GAE and CP; data curation, CP; writing—original draft preparation, GAE and IEL; writing—review and editing, GAE and IEL; supervision, GAE; project administration, GAE; funding acquisition, GAE. All authors have read and agreed to the published version of the manuscript.

Conflict of interest: The authors declare no conflict of interest.

Nomenclature

PCM	Phase Change Material
DSC	Differential Scanning Calorimetry
c_p^l	Specific heat of liquid
c_p^s	Specific heat of solid
k_l	Thermal conductivity of liquid
k_s	Thermal conductivity of solid
R_1	Inner radius of the glass tube
R_2	Outer radius of the glass tube
T_l	Liquidation temperature
T_s	Solidification temperature
T_0	Initial temperature
H_p	The enthalpy function of the PCM
L	Latent heat per unit mass
$T(R_2,t)$	Tube temperature at the outer surface at time t
T(R1,t)	Tube temperature at the inner surface at time t
$T(R_0,t)$	Tube temperature at the central axis at time t

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