Nano-PCMs for passive electronic cooling applications

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Abstract. The present work aims at investigating a new challenging use of oxide (TiO₂, Al₂O₃, etc.) nanoparticles to enhance the thermal properties: thermal conductivity, specific heat, and latent heat of pure paraffin waxes to obtain a new class of Phase Change Materials (PCMs), the so-called nano-PCMs. The nano-PCMs were obtained by seeding different amounts of oxide nanoparticles in a paraffin wax having a melting temperature of 45°C. The thermophysical properties such as latent heat and thermal conductivity were then measured to understand the effects of the nanoparticles on the thermal properties of both the solid and liquid PCM. Finally, a numerical comparison between the use of the pure paraffin wax and the nano-PCM in a typical electronics passive cooling device was implemented. Numerical simulations were carried out using the Ansys-Fluent 15.0 code. Results in terms of solid and liquid phase temperatures, melting time and junction temperature were reported. Moreover, a comparison with experimental results was also performed.

1. Introduction

The solid-liquid phase change process is an interesting possibility to reject even high heat loads, especially when they are intermittent. The term Phase Change Materials (PCMs) commonly refers to those materials, which use the solid-liquid phase change process to adsorb and then release heat loads [1]. The use of the latent heat absorption phenomenon associated with melting of a suitable PCM can be considered an effective way to delay or modify the temperature rise of a surface subjected to high and intermittent heat fluxes. The intrinsic advantage of the PCM systems is related to their simplicity and reliability; however, the reversible phase change process must be carefully analysed in order to avoid any unmanageable situations during the real operation of these devices.

As reported in several comprehensive reviews [2-4] published in the open literature, PCMs have been widely suggested for thermal storage applications due to their capability of storing and releasing large amounts of energy with a small PCM volume and a relatively low temperature variation. A large number of PCMs (organic, inorganic, and eutectic) are available in any required temperature range. An ideal PCM should exhibit a suitable phase-transition temperature, high latent heat of fusion, and high thermal conductivity; it should be characterized by high density associated with a small volume change during the melting process and low vapour pressure in the melt; moreover, it should be chemically stable over a long period of time, non-toxic and non-hazardous, and compatible with the constructional materials. Finally, it should be abundant, available, and cost effective.
Paraffin waxes have been found to exhibit many desirable characteristics, such as high latent heat, low vapour pressure in the melt, chemically inert and stable, non-toxic. However, they also have a very low thermal conductivity and a high volume change during the melting process. As such, several researchers have focused their attention to different enhancement techniques proposed to improve the thermal conductivity of the PCMs; Jegadheeswaran and Pohekar [5] and Fan and Khodadadi [6] have reviewed most of these efforts.

A new challenging opportunity to improve the thermophysical properties of the paraffin waxes is represented by the addition of nanoparticles to a base PCM. For example, Khodadadi and Hosseinizadeh [7] seeded PCMs with nanoparticles to improve thermal energy storage by increasing heat conductivity. Recent research (e.g. Shin and Banerjee [8], Chieruzzi et al. [9]) showed that nanoparticles could enhance the heat capacity of the molten salt PCM. Zhichao et al. [10] measured 8% total heat storage capacity enhancement dispersing TiO$_2$ nanoparticles at 0.2 vol% in erythritol as PCM. Jiang et al. [11] studied PCM paraffin microcapsules modified with different percentages of nano-Al$_2$O$_3$ obtaining enhanced thermal transfer and phase change properties. Karkri et al. [12] used a PCM based on high-density polyethylene (HDPE) mixed with micro-encapsulated paraffin wax. The paraffin wax acted as a high-latent-heat material, whereas the HDPE matrix ensured the compact shape. The authors observed a decrement on the PCM thermal conductivity and diffusivity, and an enhancement on the latent, sensible and total heat of the PCM increasing the paraffin content. He et al. [13] studied thermophysical properties of nanofluids as PCM, BaCl$_2$–H$_2$O PCMs, in low temperature cool storage, using TiO$_2$ nanoparticles. Thermal conductivity increased up to 16.7% suggesting the possible use of PCM in the industries of low temperature energy storage, as refrigeration and air conditioning systems, solar energy systems and heating and cooling of buildings.

This paper presents some preliminary experimental and numerical results on two nano-PCMs obtained by seeding different amounts, 0.5% and 1.0%wt, of Al$_2$O$_3$ nanoparticles in a pure paraffin wax with a melting temperature of 45 °C, called RT45. The thermal conductivity, the specific heat of solid and liquid and the latent heat of the three PCMs are experimentally measured and critically discussed.

A numerical analysis is performed in order to compare the different heat transfer characteristics of the three PCM in a typical passive cooling system for electronic thermal management. In particular, the case study consists of an aluminium heat sink of 40x40 mm and 50 mm high filled up with the PCMs and then heated from the bottom to simulate a hot spot to be cooled. Different heat flow rates were imposed (10, 20 and 30 W); the results are presented in terms of melting time and maximum interfacial temperature.

2. Materials

A commercial paraffin wax with a melting point around 45 °C, RUBITHERM® RT45, was purchased from Rubitherm Technologies GmbH (Germany). Al$_2$O$_3$ nanopowder (Nanostructured & Amorphous Materials) has a nominal diameter of 10 nm, 99% purity and 160 m$^2$/g of Specific Surface Area (SSA). Nanoparticles were added to the liquid paraffin sample, to reach mass fractions of 0.5% and 1.0%, using the analytical balance Mettler PM6100 (FS 6100 g, ± 0.01 g). Corresponding volume fractions are 0.10% and 0.21%. The nano-PCMs were mechanically stirred for 20 min at 600 rpm and then they underwent to an ultrasonic procedure using a probe sonicator (Hielscher UP200s), in order to uniformly disperse nanoparticles. Temperature of the nano-PCMs tends to increase during the sonication process, therefore, in order to maintain the sample temperature under 70 °C, the samples were immersed in a thermostatic bath set at 50 °C, sonicated for at least 150 min with the temperature of the PCMs monitored every 30 min. Then, the samples were quickly solidified and used for thermal conductivity and latent heat measurements.

3. Thermal Conductivity

Thermal conductivity was measured using a ThermTest TPS2500S, an instrument based on the Hot Disk technique. It can measure thermal properties of a wide range of materials (solids, liquids,
powders, pastes and foams). The hot disk technique uses the transient plane source mode and is applicable in the transitional regime. The plane sensor (Figure 1) consists of a nickel thin metal strip wound in a double spiral on a plane support. The spiral is enclosed between two Kapton layers printed on it. Kapton material is a poor electrical conductor, but a good thermal conductor. This disk is then placed between two samples of the same material, as shown in figure 2, and works as both a transient heater, to increase the temperature of the sample, and a precise temperature reader, to record the time-dependent temperature increase. The heat transfer is assumed purely conductive and the thermal transport properties are determined by the instrument, considering the constant electric current supplied by the sensor and the temperature change as a function of time, by monitoring the total resistance of the hot disk sensor. The declared instrument uncertainty is 5%; however, it has been calibrated with a standard stainless steel sample showing a deviation lower than 1.5%. Pure and nano-PCMs were measured at around 24 °C.

Figure 1. Hot disk sensor.  
Figure 2. Thermal conductivity setup. The sensor is placed between two paraffin samples.

4. Specific Heat

The measurement of specific heat was performed using a Differential Scanning Calorimeter (DSC), Setaram Instrumentation (Figure 3). The DSC measures specific heat capacity by heating a sample and by measuring the temperature difference between the sample and a reference sample. Three measurements must be repeated with two vessels, under an identical programmed variation of temperature, between an initial and final value, and with a predefined velocity.

Figure 3. Differential scanning calorimeter, detailed calorimetric cells.

The first run was performed with the two empty vessels and the baseline heat flux was obtained; the second run was performed with one empty vessel and a mass of reference sample in the measuring vessel, providing the reference heat flux; the third run, which imposes the heat flux into the sample,
was carried out with one empty vessel and a mass of the unknown sample in the measuring vessel. Sapphire is the reference sample and the instrument uncertainty is 2%.

5. Experimental Results
The values of the thermal conductivity of solid PCM and nano-PCMs were measured at 24 °C, i.e. at temperature more than 20 K lower than the reference melting temperature of the base paraffin wax RT45. As listed in Table 1, the experimental value of the thermal conductivity of RT45 was $0.2415 \pm 1.5\%$ W/mK, while the measurements carried on the nano-PCM paraffin waxes gave $0.2506 \pm 1.5\%$ and $0.2512 \pm 1.5\%$ W/mK, at 0.5 and 1.0 wt%, respectively. As an outcome, the addition of Al$_2$O$_3$ nanoparticles, up to 1 wt%, seems to lead to an overall enhancement of around 4%.

| Type                  | $\lambda$ (W/mK) | Variation (%) |
|-----------------------|------------------|---------------|
| RT45                  | 0.2415 ± 1.5%    | -             |
| RT45 Al$_2$O$_3$ 0.5wt% | 0.2506 ± 1.5%    | +3.8          |
| RT45 Al$_2$O$_3$ 1.0wt% | 0.2512 ± 1.5%    | +4.0          |

Latent heat of PCMs and nano-PCMs was measured imposing a programmed temperature variation, between 27 °C and 87 °C, and with a predefined velocity of 30 °C/h. Figure 4 compares the response signals (i.e. specific heat) of the three paraffin waxes as a function of temperature. The peaks represent the solid-liquid phase change. At a glance, it clearly appears that the nano-PCM paraffin waxes exhibit higher peaks as compared to the base paraffin wax. The values of the latent heat of fusion of the investigated materials were determined by applying a numerical integration of the recorded curve.

Figure 4. Specific heat capacity of paraffin waxes RT45 and its nano-PCMs, as a function of temperature.

Figure 5. Phase change latent heat of the three studied materials.

In the case of nano-PCMs, only the paraffin wax undergoes a phase change, while the Al$_2$O$_3$ nanoparticles remain solid. Thus, in the presence of nanoparticles before integrating the peak area, the contribution of the Al$_2$O$_3$ on the measured specific heat capacity ($c_{p,tot}$) was accounted and subtracted from the overall measured one.
Specifically, knowing the specific heat of Al$_2$O$_3$ ($c_{p,np} = 880$ J/kgK) and the mass fraction ($\omega$), specific heat capacity of the only paraffin wax part ($c_{p,wax}$) was calculated from the total measured specific heat capacity ($c_{p,tot}$) as:

$$c_{p,wax} = \frac{c_{p,tot} - \omega c_{p,np}}{1 - \omega}$$

The results of the numerical integration are presented in figure 5. It was observed that the latent heat slightly increases when Al$_2$O$_3$ nanoparticles are added: 2.3% and 2.8% for 0.5 wt% and 1.0 wt%, respectively. Those enhancements are lower than ones obtained for example by Shaikh et al. [14], which measured a maximum increase of around 13% in the fusion heat of paraffin waxes with single wall carbon nanotubes at 1 vol.% and Al$_2$O$_3$ nanoparticles at 1.0 and 1.5 wt%, respectively. Moreover, in a previous work (Colla et al. [15]), authors measured enhancements in the latent heat of 7.5 and 13.7% with a nanoparticles mass concentration of 1.0% in different paraffin waxes. Assuming that the intermolecular attraction in the paraffin wax/Al$_2$O$_3$ dispersion explains the enhancement of the latent energy, as proposed by Shaikh et al. [14] according to the Lennard-Jones theory of intermolecular forces, different paraffin waxes have different surface interaction with the Al$_2$O$_3$ nanoparticles. Therefore, the change in latent heat of PCM due to the inclusion of nanoparticles strongly depends on the type of the PCM and the nanoparticles.

6. Physical and Numerical Models

The numerical analysis simulates an electronic passive cooling device, in particular an aluminium box filled up with pure paraffin wax or nano-PCM with 0.5% and 1.0% weight of nanoparticles. The geometry is a rectangular box with height equal to 50 mm and base equal to 40 mm. A two dimensional domain is considered, as depicted in figure 6.

![Figure 6. Domain and geometrical size.](image)

The boundary conditions are given by an assigned heat flux at the bottom, to simulate the heat transfer dissipation of an electronic device, and by convective heat transfer on the top, while the sides are adiabatic. The heat flux is related to the heat transfer rate, equal to 10 W, 20 W and 30 W, i.e. typical values in an electronic device. The melting of paraffin wax is modelled with the enthalpy-porosity method [16], given that paraffin wax does not melt at a single temperature value, but in a temperature range. In this temperature range, solid and liquid phases coexist, and the “liquid fraction” is defined as a parameter indicating the liquid fraction in a simulation model cell. This parameter is expressed with $\beta$ and its value changes from 0, when the domain is completely solid, to 1 when it is totally liquid:
\[
\begin{align*}
\beta &= 0 \quad \text{for } T < T_{\text{solidus}} \\
\beta &= \frac{T - T_{\text{solidus}}}{T_{\text{liquidus}} - T_{\text{solidus}}} \quad \text{for } T_{\text{solidus}} < T < T_{\text{liquidus}} \\
\beta &= 1 \quad \text{for } T > T_{\text{liquidus}}
\end{align*}
\]

where \( T \) is the local temperature of the cell, \( T_{\text{liquidus}} \) is the temperature upper which the domain is completely liquid and \( T_{\text{solidus}} \) is the temperature below which it is completely solid. The mixed solid-liquid zone exists in a temperature range between \( T_{\text{liquidus}} \) and \( T_{\text{solidus}} \). The gravitational acceleration is taken into account along the y-axis and the Boussinesq approximation is assumed in order to take into account the buoyancy force due to natural convection. The solid part of the domain is modelled by adding a source term in the momentum equation. The single-phase model to simulate the nano-PCM in liquid phase is assumed. In the following, the mass fraction of the nano-PCM is \( \psi \) and it indicates the ratio between the mass of nanoparticles and the total mass of the paraffin wax and nanoparticles. The properties of the materials used for the simulations are listed in the table 2.

The specific heat, the thermal conductivity and the latent heat are calculated from the experimental results, while the following properties are calculated.

The density is evaluated by:

\[
\rho_{\text{NANOPCM}} = (1 - \psi) \rho_{\text{PCM}} + \psi \rho_{\text{PARTICLES}}
\]

where \( \rho_{\text{NANOPCM}} \) is the density of the nano-PCM, \( \rho_{\text{PCM}} \) is the density of pure paraffin wax, \( \rho_{\text{PARTICLES}} \) is the density of the nanoparticles (aluminum oxide) and \( \psi \) is the mass fraction of the nanoparticles.

The values of specific heat and thermal conductivity are obtained from the experimental results for the liquid phase while for the solid phase it used the value provided by the supplier.

The viscosity is calculated from the Maxwell equation:

\[
\mu_{\text{NANOPCM}} = \frac{\mu_{\text{PCM}}}{(1 - \psi)^{2.5}}
\]

where \( \mu_{\text{PCM}} \) is the viscosity of the pure paraffin wax, \( \mu_{\text{NANOPCM}} \) is the viscosity of the nano-PCM.

**Table 2.** Thermal properties of the materials, i.e. pure paraffin wax, nano-PCM at 0.5%wt and nano-PCM at 1.0%wt. \(^a\) from supplier data sheet, \(^b\) calculated, \(^c\) measured.

| Properties                  | Pure PCM | Nano-PCM 0.5%wt | Nano-PCM 1%wt |
|-----------------------------|----------|-----------------|---------------|
| Density [kg/m\(^3\)]       | Solid    | 880 \(^a\)     | 895 \(^b\)    | 911 \(^b\) |
|                             | Liquid   | 770 \(^a\)     | 786 \(^b\)    | 802 \(^b\) |
| Specific heat [J/kg·K]      | Solid    | 3028 ± 2\(^c\) | 2943 ± 2\(^c\) | 3032 ± 2\(^c\) |
|                             | Liquid   | 2333 ± 2\(^c\) | 2322 ± 2\(^c\) | 2376 ± 2\(^c\) |
| Thermal Conductivity [W/m·K]| Solid    | 0.241 ± 1.5\(^c\) | 0.251 ± 1.5\(^c\) | 0.251 ± 1.5\(^c\) |
|                             | Liquid   | 0.2\(^a\)     | 0.203 \(^b\)   | 0.206 \(^b\)   |
| Viscosity [kg/m·s]          |          | 0.0256 \(^b\) | 0.0259 \(^b\)   | 0.0263 \(^b\)   |
| Thermal expansion factor [1/K]|        | 1.25 · 10\(^4\)\(^a\) | 1.22 · 10\(^4\)\(^b\) | 1.19 · 10\(^4\)\(^b\) |
| Latent Heat [J/kg]          |          | 1.397·10\(^5\) ± 2\(^c\) | 1430·10\(^5\) ± 2\(^c\) | 1.436·10\(^5\) ± 2\(^c\) |
| \( T_{\text{solidus}} \) [K] |          | 308            | 308            | 308            |
| \( T_{\text{liquidus}} \) [K] |          | 322            | 322            | 322            |

The thermal expansion coefficient is calculated from [17]:

\[
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\[ \beta_{\text{NANOPCM}} = \frac{(1 - \psi)(\rho \beta)_{\text{PCM}} + \psi(\rho \beta)_{\text{PARTICLES}}}{\rho_{\text{NANOPCM}}} \]  

(5)

The latent heat is obtained from experimental data. It is worth underlying that with the increasing of mass fraction the latent heat of fusion tends to increase. The thermal conductivity in liquid phase is calculated from the Maxwell equation [17].

The governing equations of the model are:

\[ \nabla \cdot (\rho \vec{V}) = 0 \]  

(6)

\[ \rho \left( \frac{\partial \vec{V}}{\partial t} + (\nabla \cdot \vec{V}) \vec{V} \right) = \mu \left( \nabla^2 \vec{V} \right) - \nabla \vec{p} + \vec{S} \]  

(7)

\[ \frac{\partial (\rho \beta)}{\partial t} + \nabla \cdot (\rho \beta \vec{V}) + L \left[ \frac{\partial (\rho \beta)}{\partial t} + \nabla \cdot (\rho \beta \vec{V}) \right] = k \left( \nabla^2 T \right) \]  

(8)

where \( \vec{V} \) is the velocity vector of the material in the liquid phase, \( \mu \) is the viscosity of the material, \( p \) is the pressure and vector \( \vec{S} \) is a source term given as [18]:

\[ \vec{S} = \frac{(1 - \beta)^2}{\left( \beta^3 + \pi \right)^3} A_{\text{mush}} \vec{V} - \rho g \gamma (T - T_0) \hat{y} \]  

(9)

**Figure 7** Validation test: a) data from Krishnan et al. [21] b) data from present numerical model.

The first term models the presence of the solid part in the mixed (liquid and solid) region, where \( \pi \) is a small number (0.001) to avoid division by zero [18] and \( A_{\text{mush}} \) is the mushy zone constant which describes the kinetic process in the mushy zone [19]. In these simulations, the value of mushy zone constant is set to \( 10^7 \). The second term models the natural convection in the liquid phase of the material according to the Boussinesq approximation. The energy equation uses the enthalpy of the material according to enthalpy-porosity method [20].

The employed grid is a set of quadrilateral control volumes with a coarse grid in the centre and fine grid near the boundary of the domain. The test carried out for the mesh independence solution consists
of four different grids: 20 \times 25, 40 \times 50, 60 \times 75, and 80 \times 100. The mesh 60 \times 75 represents the best trade-off for the present study, as good compromise between the accuracy and the computational time. The validation of the model is made by comparison with the work of Krishnan et al. [21]. The same characteristics are employed and then compared. The results are shown in figure 7.

**Figure 8.** Mean values of liquid fraction vs time, for heat flux equal to: a) $6.25 \times 10^3$ W/m$^2$ (10 W) b) $12.5 \times 10^3$ W/m$^2$ (20 W), c) $18.75 \times 10^3$ W/m$^2$ (30 W).

**Figure 9.** Mean values of temperature vs time, for heat flux equal to: a) $6.25 \times 10^3$ W/m$^2$ (10 W) b) $12.5 \times 10^3$ W/m$^2$ (20 W), c) $18.75 \times 10^3$ W/m$^2$ (30 W).
7. Numerical Results

The numerical results are shown in terms of average liquid fraction, average temperature of the whole domain, melting time, and the max interfacial temperature at different heat flow rates (10, 20 and 30 W) and mass fractions (pure material, 0.5%wt and 1.0%wt). The maximum interfacial temperature corresponds at the temperature of the bottom taken at time of completed melting.

In figure 8, there are three plots on the average liquid fraction for pure material, 0.5%wt and 1.0%wt concentration, as functions of time, and a), b) and c) refer to 10 W, 20 W and 30 W of imposed power, respectively. When the concentration of nanoparticles increases it seems that the liquid fraction tends to rise more slowly. In fact, the liquid fraction for pure PCM, respect to liquid fraction for the 0.5%wt or 1.0%wt concentrations, moves up more quickly. The only exception happens with a power supplied of 10 W, where the liquid fraction of the material with 0.5%wt of nanoparticles grows faster. This behaviour can be explained by the smaller value of latent heat at decreasing the concentration of nanoparticles into PCM. Table 2 reports the latent heat of the materials that slightly increases as the concentration of nanoparticles is increased.

In figure 9 there are three plots about the average temperature of the pure material, and of the PCM with 0.5%wt and 1.0%wt nanoparticles concentration, as functions of time, and a), b) and c) refer to 10 W, 20 W and 30 W imposed power, respectively.

The temperature path can be divided in three ranges: in the first range the temperature sharply increases in linear way. In the second stage, longer than the first, temperature follows the same trend, even if increasing less suddenly. This stage is important, because the melting occurs in a large part of its domain, leading to a slow increase of the average temperature. In the third range, the melting of whole domain is almost completed and the temperature returns to quickly rise. Table 3 reports the melting time and the corresponding maximum interfacial temperature for the three supplied powers.

| POWER SUPPLIED | Material   | 10 W  | 20 W  | 30 W  |
|----------------|------------|-------|-------|-------|
|                | Time [s]   | Max interfacial temperature [K] | Time [s]   | Max interfacial temperature [K] | Time [s]   | Max interfacial temperature [K] |
| Pure PCM       | 1906       | 413.7 | 1154  | 477.8 | 900  | 541.3 |
| Nano-PCM at 0.5%wt | 1935   | 412.6 | 1192  | 476.5 | 889  | 528.9 |
| Nano-PCM at 1.0%wt | 2020   | 413.4 | 1239  | 480.1 | 965  | 554.6 |

8. Conclusion

This paper presents an experimental and numerical study focused on the possible application of nano-PCM for electronics cooling. A commercial paraffin wax, having the solid-liquid phase transition at 45°C, was selected and used to develop two nano-PCM, by seeding Al₂O₃ nanoparticles at 0.5% and 1.0%wt, respectively. The three PCMs were then experimentally characterized in order to measure their solid thermal conductivity, solid and liquid heat capacity, and latent heat of fusion. The results show that the nanoparticles lead to a very low increase of both the thermal conductivity and latent heat. Furthermore, a numerical analysis was conducted to understand the effects of the nanoparticles during the solid-liquid phase change process in a passive electronics cooling device. In particular, a rectangular box 50 mm high, with a base equal to 40 mm, filled up with the three PCMs, was simulated when heated from the bottom by imposing 10, 20 and 30 W, respectively. The results show that the nano-PCMs are able to delay the melting process with a globally lower mean temperature.
Additional experimental, theoretical, and numerical work is surely needed to understand the heat transfer mechanisms on the basis of the performance of the paraffin waxes, in order to develop rigorous procedure for the nano-PCMs development and characterization.

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