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TEST OF TWO PHASE CHANGE MATERIALS FOR THERMAL ENERGY STORAGE

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Abstract. *Using a very simple experimental layout, some tests on the behavior of two phase change materials, during fusion and solidification processes, were accomplished. To do this, a system using thermal oil as the heat transfer carrier was used and the phase change material being tested operated in a fusion and solidification cycle. The heat transfer oil transferred heat to the phase change material during the fusion step and carried heat away from the change phase material during the solidification step. The influence of the mass flow rate of the heat transfer fluid, as well as of its inlet temperature, in the response of the phase change material, was studied. Axial and radial temperature profiles inside the phase change materials were obtained during the experiments. One of the tested phase change materials was a salt while the other was an organic.*

Keywords: *Thermal Energy Storage, Phase Change Materials, Heat Transfer*

1. INTRODUCTION

It is frequent to store energy available from renewable energy sources for its posterior use when necessary. This energy storage procedure can be of four types: chemical energy, electrical energy, mechanical energy and thermal energy (McLarnon and Cairns, 1989; Evans et al., 2012). The thermal energy storage is a very interesting option to store available solar energy as well as thermal energy being wasted in industrial processes. When the storage material does not suffer any phase change or chemical reaction the thermal storage technique is named sensible heat storage. If the storage medium undergoes any chemical reaction, there is thermochemical energy storage, whereas if there is any phase change of the material, there is latent heat storage (Pardo et al., 2014; Zhang et al., 2016).

The latent heat thermal energy storage uses a material that suffers a phage change of its molecular structure, be it solid-liquid, liquid-vapor or solid-solid (Sharma and Sagara, 2005). The solid-liquid phase changes are considered more efficient when compared with the liquid-vapor or the solid-solid transitions. Liquid-vapor transitions undergo a high volume variation while the solid-solid transitions have a very low latent heat of phase change (Sharma and Sagara, 2005; Zhang et al., 2016). The materials used for latent heat storage are called phase change materials (PCM) and because of the high latent heat value of phase change, the mass of material required per unit of stored energy is smaller than for sensible heat storage, leading to advantageous applications either in domestic or in industrial storage processes (Sharma and Sagara, 2005; Sharma et al., 2009). There are however some practical aspects that still require further development, like the heat transfer rate between the thermal energy transportation fluid and the heat storage material, or the PCM capacity of enduring a high number of operating cycles keeping the corresponding chemical and thermal

stability (Xu et al., 2015). The biggest disadvantage of the PCM's is the lower thermal conductivity, in the 0.2 to 0.8 W/(m K) range, and if these values could be increased an overall augmentation of the thermal efficiency of the storage process would be achieved through the improvement of the thermal energy charging and discharging processes (Zhang et al., 2016). The encapsulation of PCM's has solved some of the heat transfer limitations and is being used from solar plants to the thermal structure of buildings or even in textile fabrics, covering a wide range of operating temperatures (Xu et al., 2015).

To increase the charging and discharging efficiency different techniques to increase the heat transfer rate were studied (Fernandes et al., 2012; Kuravi et al., 2013; Haocheng, et al., 2014). The simplest methodology is to increase the heat transfer area through the application of fins to the heat transfer surfaces. Either radial or axial fins were evaluated (Zhang et al., 2016). Velraj et al. (1997) investigated the solidification of PCM's in a vertical cylindrical pipe with internal radial fins and concluded that this configuration with a V like geometry could keep the PCM inside it guarantying the best possible results for a finned geometry. Erek et al. (2005) studied numerically and experimentally the latent heat energy storage by means of radial fins. The stored energy increased with the fin radius and the reduction of inter fin spacing. Castell et al. (2008) added graphite longitudinal fins inside the PCM media to test the possible reduction of the solidification time. They found that, although the use of fins changed the natural convection currents inside the PCM, it was possible to reduce the solidification time of the material.

The majority of the commercially available PCM's are mixtures or impure materials, so the phase change does not take place at a fixed temperature value, but instead, in a range of temperature values. On the contrary, pure PCM's present phase changes at fixed temperatures (Zalba et al, 2003).

Low temperature PCM's, either organic, inorganic or eutectic are mainly used for the recuperation of wasted thermal energy in buildings, while inorganic or eutectic PCM's can be used in solar applications or other high temperature thermal energy storage applications (Zalba et al., 2003; Zhang et al., 2016).

The objective of the present study was to obtain data on the time evolution of the temperature of two commercially available PCM's during the fusion and solidification processes. This work is the first part of a study where heat transfer coefficients, during the PCM fusion and solidification were also measured.

2. LABORATORY SET-UP

To evaluate the fusion and solidification of the two PCM's under scrutiny a simple experimental procedure was established. The thermal oil Therminol 66 was used as energy transportation fluid, working either as a heating or as a cooling fluid. This oil circulated among the three main components of the experimental setup, namely, a test heat exchanger inside which there was the PCM, an oil heater equipped with electrical resistances, and a water cooled shell and tube heat exchanger. A centrifugal pump, capable of operating up to 170 ºC, was used to promote the thermal oil circulation. A 3D scheme and a picture of the experimental set-up are shown in Fig.1. The testing procedure was composed by a two-step cycle. In the first step (fusion) heat was supplied to the PCM, in the second step (solidification) heat was retrieved from the PCM. During the PCM fusion step the thermal oil was heated in equipment A and thermal energy was supplied to the PCM in the test exchanger C. During the solidification of the PCM, the thermal fluid received thermal energy from the PCM in the test exchanger C and rejected this thermal energy to the environment through the shell and tube heat exchanger B.

Figure 2 shows, with more detail, the thermal oil circulation cycle:

- During the PCM fusion step the heat transfer oil was heated by equipment A, Fig. 2, which was controlled by a PID *Eurotherm* controller according to the oil temperature in point K. The centrifugal pump C pumped the fluid towards the PCM heat exchanger D until the PCM was completely liquefied;
- During the PCM solidification step, the heat transfer oil extracted heat from the PCM heat exchanger D and transferred this energy towards the environment through the shell and tube heat exchanger B. Again, the centrifugal pump C was responsible for the oil impelling. This step ended with the complete solidification of the PCM.

The PCM heat exchanger was designed according to the following specifications:

- Low pressure drop in a single pipe heat exchanger;
- Easy placement of temperature probes to allow the evaluation of radial and axial temperature profiles inside the PCM. The thermocouples positions inside the PCM heat exchanger are shown in Fig. 3. Thermocouples 1 to 3 are close to the external surface of the thermal oil pipe. Thermocouples 4 to 6 are 2 mm below the external shell of the heat exchanger. Thermocouples 7 to 10 are midway between the oil pipe and the exchanger external shell. When jointly evaluating (T1, T2, T3) with (T4, T5, T6) and (T7, T8, T9, T10) it is possible to characterize the axial evolution of the temperature inside the PCM for different radii. When comparing (T1, T7, T4) with (T5, T9, T8, T2) and (T3, T10, T6), it is possible to evaluate the radial evolution of the PCM temperature at different levels. Finally, thermocouples T8 and T9 validate the thermal symmetry.

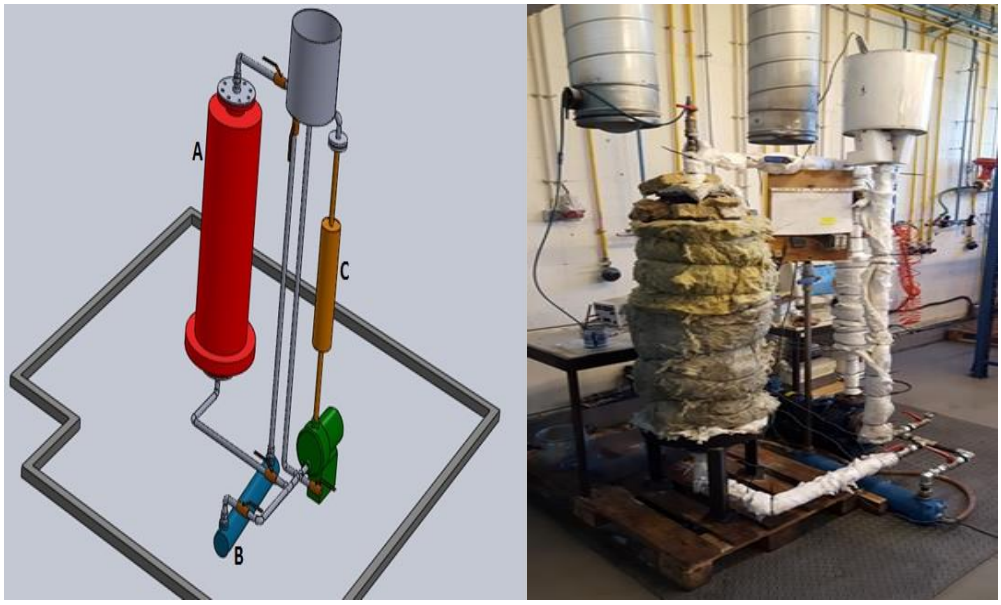

Figure 1 - 3D scheme and photo of the experimental set up. A - Thermal fluid heater; B – Thermal fluid cooling heat exchanger; C – PCM heat exchanger.

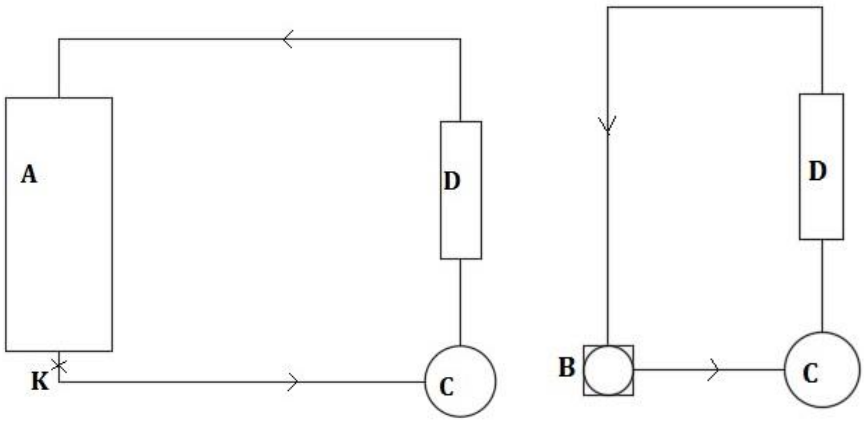

Figure 2 – Left side, scheme of the PCM heating process; A-Thermal fluid heater; C-Centrifugal pump; D-PCM heat exchanger. Right side, scheme of the PCM cooling process; B-Cooling heat exchanger; C- Centrifugal pump; D- PCM heat exchanger.

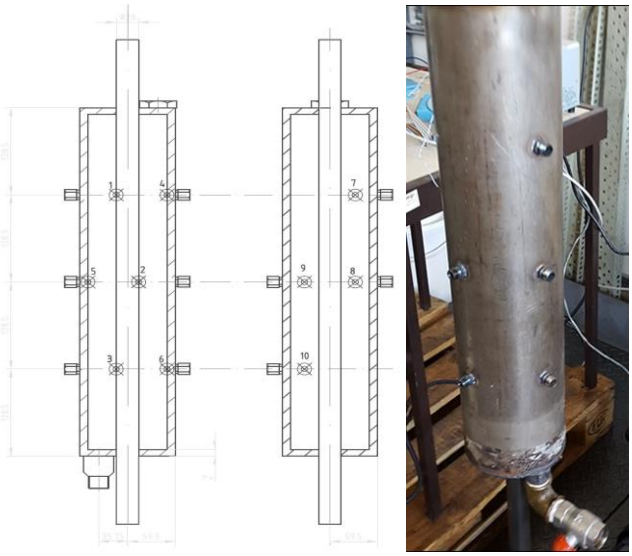

Figure 3 – Test heat exchanger and thermocouples location.

All these thermocouples are type T thermocouples, because they are more suitable for the range of temperatures under analysis and have an uncertainty of ±0.75 % above 90 °C (Lipták, 2003; Coleman and Steele, 2009).

3. THE PHASE CHANGE MATERIALS

The main thermal properties of the two tested PCM's are presented in Table 1. The H105 PCM is characterized by its maker as a high temperature material and is a granulated product. It is an inorganic material, common in high temperature latent heat storage, the salt KNO_3. As far as the material A82 is concerned it is an organic product, paraffin, although the manufacturer has not given any clear information about its nature. It was supplied as a package of small spherical particles, Fig. 4.

Table 1 – Thermal properties of the tested PCM's.

PCM designation	**Phase change temperature**	**Density**	**Latent heat per unit mass**	**Latent heat per unit volume**	**Specific heat**	**Thermal conductivity**
	[ºC]	[kg/m^3]	[kJ/kg]	[MJ/m^3]	[J/(kg K)]	[W/(m K)]
H105	104	1700	125	213	1500	0.500
A82	82	850	155	132	2210	0.220

As can be seen in Tab. 1 the density of H105 is twice that of A82. Thus for the same volume being tested the corresponding mass was double for H105. From measurements of the solid-liquid thermal expansion of both materials it was found that H105 had a thermal expansion of 21 % whereas A82 had a thermal expansion of 11 %. These values were taken into account during the PCM exchanger filling process.

Figure 4 – H105 and A82 phase change materials.

Table 2 presents the designation of the experiments that were carried according to the corresponding set point temperature and the centrifugal pump operating frequency.

Table 2 - Set-point temperature and pump operating frequency.

	H105	**A82**
t1	140 ºC, 50 Hz	140 ºC, 50 Hz
t2	140 ºC, 38 Hz	140 ºC; 38 Hz
t3	–	110 ºC; 38 Hz

The mass and volume flow rates of thermal oil used during the experiments were function of the centrifugal pump operating frequency, and are shown in Tab. 3 according to the adopted test conditions.

Table 3 – Mass and volume flow rate values of the heat transfer fluid for the adopted testing conditions.

		$\dot{m}$ [kg/s]	$\dot{V}$ [m^3/s]
t1	Fusion	0.825	3.20
	Solidification	0.675	2.45
t2	Fusion	0.700	2.70
	Solidification	0.550	2.00
t3	Fusion	0.675	2.65
	Solidification	0.550	2.00

4. EXPERIMENTAL RESULTS

The time evolution of the PCM's temperature for several points inside the two tested materials is now presented. Through the analysis of the time temperature curves it is possible to validate the different heat transfer regimes taking place during the fusion and solidification processes. The curves shown for the H105 concern results obtained for the t2 testing conditions while for the A82 the plotted curves concern the t1 experiments. The choice for the presentation of these results is connected with the better perception that can be achieved through the analysis of these experimental curves.

4.1 Characterization of the axial and radial evolution of temperature during the fusion process

During the fusion process the hot thermal oil supplies thermal energy to the PCM contained in the test heat exchanger, while the temperature evolution in several spots inside the PCM is measured. Accordingly it was possible to evaluate axial temperature evolution at different radii as well as the radial temperature evolution for different heights. Fig. 6 presents the axial temperature evolution while Fig 7 presents the radial temperature evolution for the same fusion experiment.

The PCM fusion starts in the lower region, close to the external wall of the inner pipe, plots i and iv in Fig. 6, obtained with thermocouple T3. In the following time instants the PCM moves up to the top of the container due to natural convection currents occurring because of the strong density differences between the solid and the liquid phase. Because of this sudden rise of the fused PCM there is an anticipated liquefaction of both the upper and lower regions, plots i and iv in Figs. 6 and 7, resulting on the formation of a conic geometry in the solid-liquid interface. Because of this conic evolution of the material fusion, the readings from thermocouple T6, placed at a lower external level in the test heat exchanger, present a longer phase change delay, plots iii and vi from Figs. 6 and 7. During this fusion process liquid and solid co-exist in close contact. The conduction is the unique heat transfer mechanism in the solid phase and the thermal conductivity is low in the PCM's. Such is clearly detectable in the radial temperature evolutions, plots ii to iv in Fig. 7, where, as far as the thermocouples are from the heat transfer surface, the external surface of the inner pipe, the slower is the event of fusion. It is also quite clear, by looking at the thermocouple data, the evidence of the conical geometry, i.e., the solid-liquid interface. However as soon as the liquid phase appears, the solid phase starts receiving convection heat transfer from the liquid phase remaining in the upper layers of the shell side. Concerning the liquid phase, the density differences, induced by the temperature variations, increased the heat transfer rate, and obviously the PCM circulation This explains the higher temperature values at its uppermost surface, Fig. 7 plot iv.

However when looking at the initial results obtained with the thermocouples T1, T4 and T7 for the H105 PCM, Fig. 7 plot i, it is necessary to remember that this material has a thermal expansion coefficient superior to that of A82, and because of that the volume occupied by the H105 in the solid phase is smaller and the initial temperature values measured by those thermocouples refer to the air remaining above the solid material. Afterwards the sudden temperature jump detected by those thermocouples refers to the corresponding liquid temperature reading, resulting

from the solid fusion and expansion, Fig. 7 plot i. This does not happen with A82, where the thermocouples T1, T4 and T7 are initially reading the solid phase temperature of A82.

Finally, the closeness of data collected by the thermocouples T8 and T9, plots ii and v from Figs. 6 and 7, clearly demonstrates that there is geometrical symmetry, symmetry of boundary conditions and a uniform liquid mass distribution inside the testing PCM exchanger.

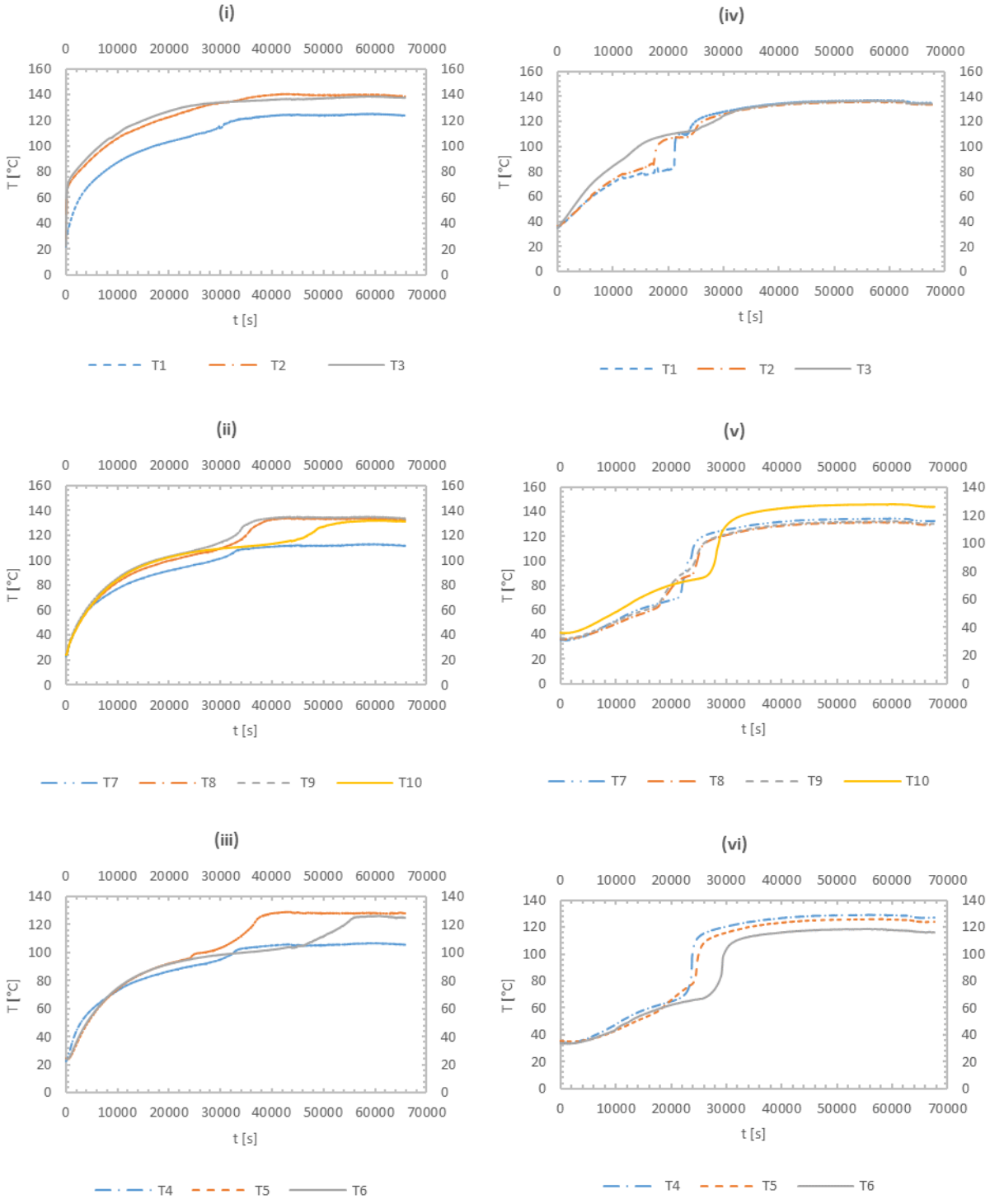

Figure 6 – Axial evolution of phase change materials temperature during fusion. Plots i to iii - H105; Plots iv to vi – A82.

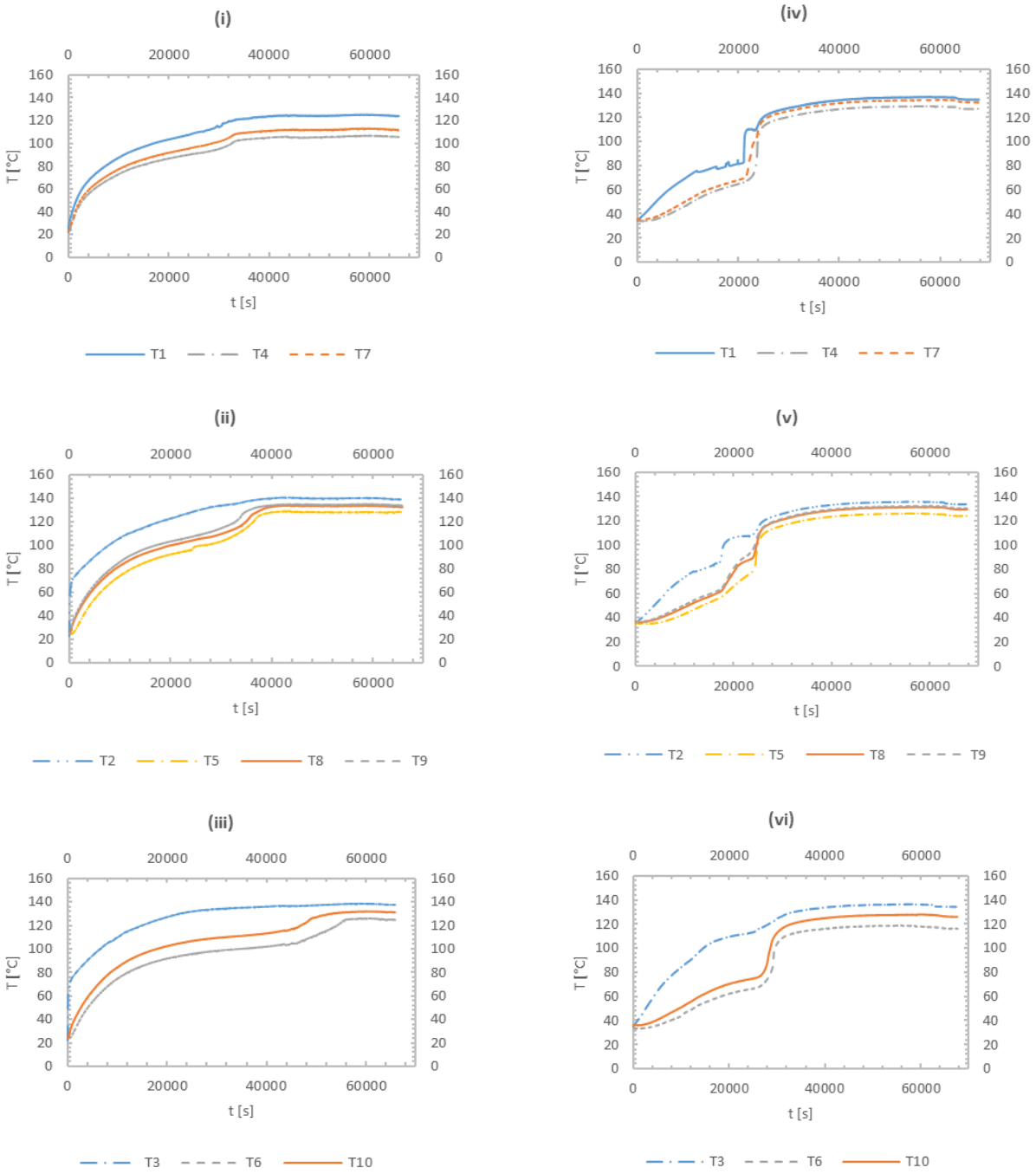

Figure 7 - Radial evolution of phase change materials temperature during fusion. Plots i to iii - H105; Plots iv to vi – A82.

4.2 Characterization of the axial and radial evolution of temperature during the solidification process

Immediately after the fusion step the temperature and circulation process of the heat transfer oil was adjusted to start the heat extraction from the liquid PCM. Figure 8 presents the axial temperature evolution during the solidification step, whereas Fig. 9 presents the corresponding radial temperature evolution. In the first time instants of the cooling process it is detected an abrupt temperature drop, followed by a smaller temperature gradient, during which the phase change takes place. If the material was a pure one, a constant temperature time evolution would be found, but as it is a mixture there is a temperature interval concerning this change of phase. Finally at the end of the process, temperature changes with time became slower.

These initial abrupt temperature changes due to the large thermal differences between the heat transfer oil and the PCM, are demonstrated by the readings from the thermocouples placed close to the external surface of the inner tube

crossing the testing heat exchanger, Fig. 8 plots i and iv, where the temperature readings closely follow the oil temperature evolution. The almost constant temperature periods refer to the change of phase, the most beneficial characteristic of these PCM, Fig. 8 plots ii to iv.

During the solidification the H105 presents a narrower temperature range, plots i to iii in Figs. 8 and 9, while A82 has a wider solidification temperature range, plots iv to vi in Figs. 8 and 9. This means that A82 is a mixture of several substances while H105 is closer to a pure substance.

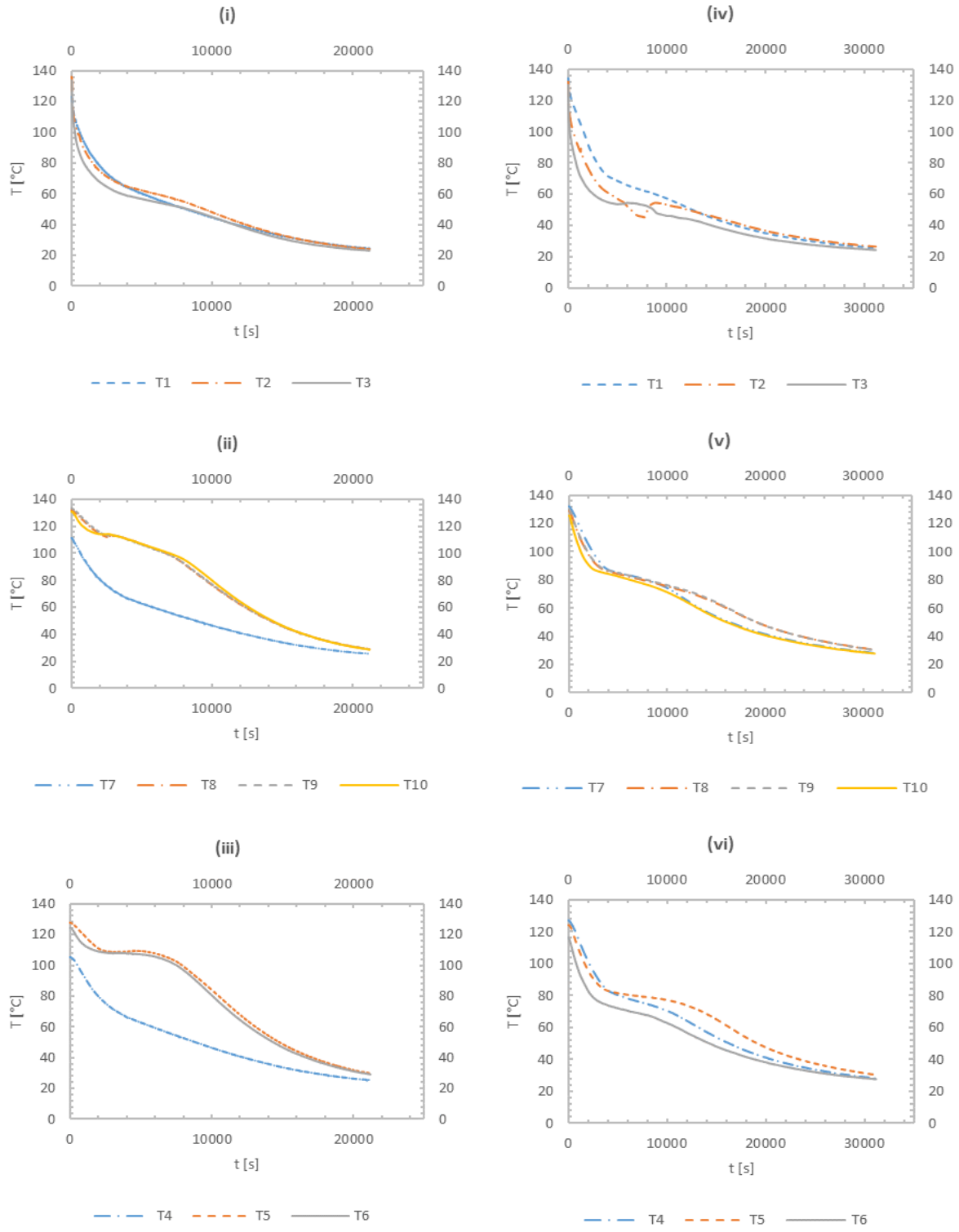

Figure 8 - Axial evolution of phase change materials temperature during solidification. Plots i to iii - H105; Plots iv to vi – A82.

At the end of the solidification process the temperature evolution is slower because the temperature difference between the PCM and the heat transfer oil is smaller. When the PCM solidifies the heat transfer process becomes very difficult as the only heat transfer mechanism is solid phase conduction, which is, as repeatedly referred, very small for the PCM's.

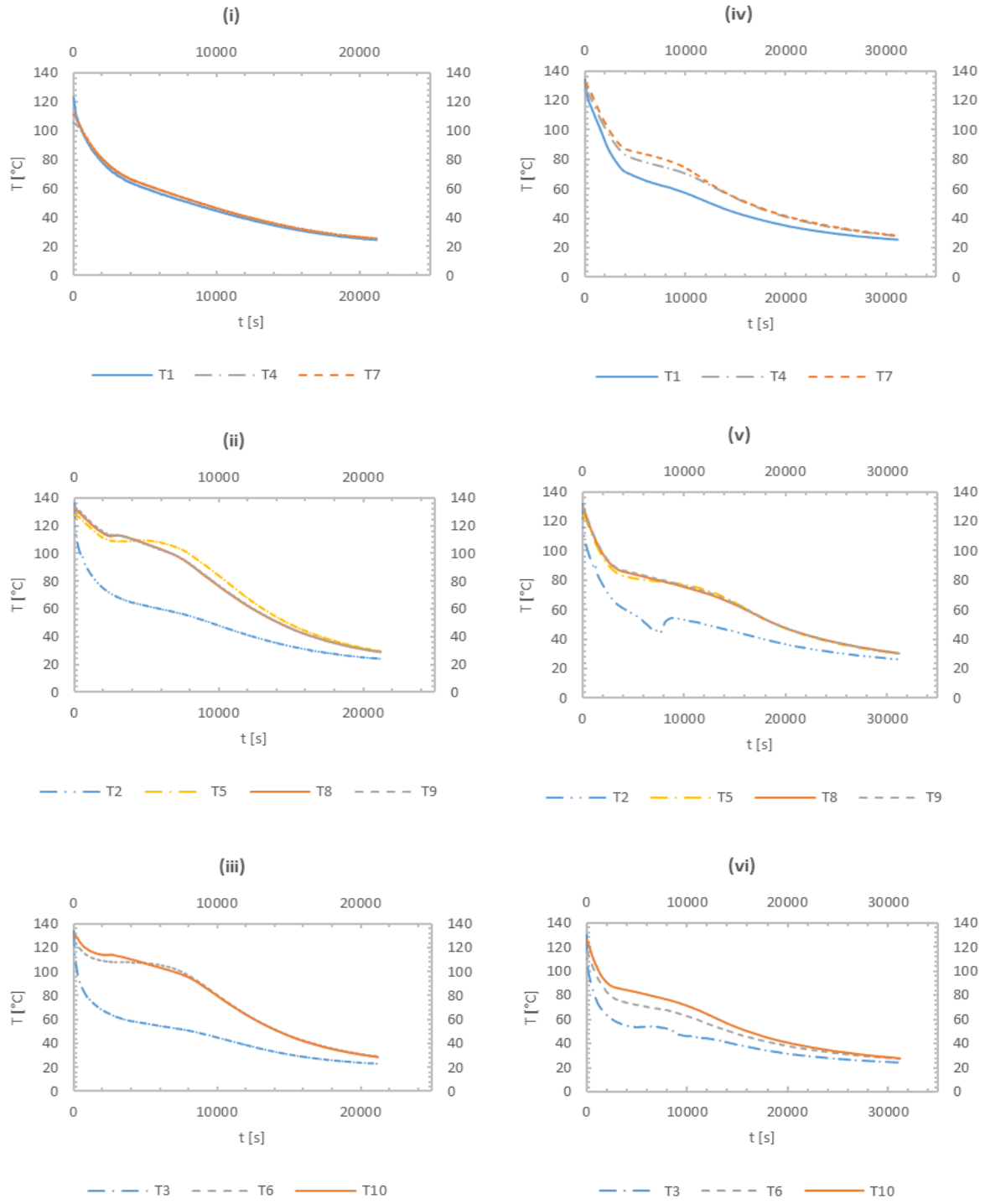

Figure 9 - Radial evolution of phase change materials temperature during solidification. Plots i to iii - H105; Plots iv to vi – A82.

As the heat transfer oil flows in the vertical ascending direction, now it is not possible, contrarily to the fusion situation, to take advantage of the natural convection currents that exist inside the liquid PCM. Now the PCM starts its

solidification at the bottom of the testing heat exchanger, Fig. 9 plots iii and iv, region where the temperature differential between the heat transfer oil and the PCM is higher, but on the other end the liquid phase is less dense than the solid phase and while occupying the upper part of the exchanger it counteracts any free convection phenomenon that could enhance the heat transfer process. Of course, had the heat transfer oil flow been inverted for the heat extraction process, the natural convection process could enhance the thermal energy transfer.

Again, the thermocouples T1, T4 and T7 continue to show abnormalities because of the more intense contraction of H105 when compared to A82.

5. CONCLUSIONS

The analysis of the fusion and solidification process of two commercially available PCM's was presented. To carry out such task a small laboratory scale installation was built. The heat was supplied and : retrieved to the PCM being tested in a single pipe shell and tube heat exchanger and the evolution of the temperature in several points inside the material was analyzed and commented. This work is the first part of a study where heat transfer coefficients, during the PCM fusion and solidification were also measured.

6. ACKNOWLEDGEMENTS

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