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Investigation of a Thermal Storage System based on Phase Change Material: Heat Transfer and Performance Characterisation

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Abstract

The integration of latent heat storage solutions into modern domestic heating systems has the potential to enhance the overall system performance compared to standard hot-water systems (radiators and tanks) due to augmentation of the stored heat by the latent heat of a suitable material. This paper presents computational prediction and experimental validation of the dynamic behaviour and performance of an active thermal storage system for domestic applications, based on the use of a hydrated salt PCM. The thermal extraction and heating rates for the PCM tank are compared to a water filled tank. Flow and temperature fields are analysed in a customised storage tank design for heat transfer and performance characterisation. Experimental findings show excellent agreement with the 3D CFD simulation results. The heat removal performance has been identified as being the limiting factor when compared to a water-based system. It is also confirmed that the PCM solution has the capability to store a large amount of heat effectively but design improvements are required to eliminate the cooling limited heat transfer process in the current apparatus.

Keywords: thermal energy storage; latent heat; salt hydrate; phase change material; domestic hot water

1 Introduction

1.1 Thermal energy storage systems

The depletion of finite natural fossil fuel resources together with concerns, both health-related and environmental, relating to the release of product gases from fossil fuel combustion (COx, NOx, SOx, particulates, etc.) to the atmosphere are acting as major drivers towards a higher efficiency and more sustainable energy future. Essential components of such a future are harnessing and utilisation of renewable alternative energy sources; recovery and re-use of waste and low-grade heat. The availability of most alternative energy sources (e.g. solar, geothermal) exhibits appreciable fluctuations (caused by day-night cycles, weather, etc.). This intermittence in supply behaviour is also true for low-grade waste heat that is abundantly available in the industrial sector, and also in the domestic environment. The desire to efficiently utilise energy from such resources has led to an enhanced interest in thermal energy storage (TES) systems. TES plays a vital role in energy conservation as it helps in reduction of the discordance (in time or rate) between the production or availability of energy and its consumption [1]. Shifting the heating/cooling loads to night-time hours, when the cost of electricity is lower, can help cut down the energy costs if electricity is stored as thermal energy. The electricity grid peak loads can thus be lowered which in effect will help slashing the overall carbon dioxide emissions as the peak power loads are generally covered by fossil fuel thermal plants [2, 3]. In light of the energy supply security, whilst saving fuel (and cost) and minimising wastage of energy, TES has emerged as a strong candidate for improving the global energetic scene and is being employed to increase the efficiency and performance of different applications.

In a domestic setting, hot water tanks are often used as active energy storage systems to provide hot water for sanitary and cooking purposes [4]. Such hot water tanks are an important source of energy consumption in the domestic environment and have traditionally used water as the heat storage medium. Thermal energy is stored as sensible heat and is proportional to storage mass and temperature rise of water. In a situation where the demand for hot water increases, a simultaneous increase in volume of the water tank will be required in such TES systems which will result in more bulky structures and higher space requirements. Consequently, for a given amount of thermal energy, it is not favourable to store large amount of hot water but it is desirable to have a higher energy storage density.

1.2 PCMs in TES

Latent heat thermal energy storage (LHTES) is an attractive TES method that uses phase change materials (PCMs) to achieve a high density of heat storage. The PCM stores heat as sensible heat before reaching its phase change temperature. Beyond the phase change temperature the PCM undergoes a phase transition from solid to liquid phase (i.e. melts) and the heat energy storage goes on – now as latent heat at a constant temperature i.e. isothermally. Most popular PCMs are either inorganic compounds (crystalline salt) or organic compounds (paraffin) that have certain properties, such as congruent melting, formation of initial crystals nucleate, unchanged thermal properties during phase change, compatibility, availability, low price, high thermal conductivity, and high relative latent heat, to be suitable for use as a heat storage material [5].

PCMs can be employed as storage material in the hot water tanks to increase the heat storing capacity and density (since the latent heat is much larger than the sensible heat). This would result in smaller physical volumes of tank compared to tanks using water as the heat storage medium. In addition to this, uncontrolled rise in water temperature (occurs when water is not withdrawn from the tank while heat input is maintained) can be eluded by carefully selecting a PCM that changes its phase at a temperature close to the desired usage temperature of hot water. The excess heat, instead of being utilised to increase the temperature of water, is absorbed and stored by the PCM, thereby restricting the water temperature to a set point and facilitating small temperature variations from storage to extraction [6].

The use of PCMs for TES has been of significant research interest with particular importance given to material selection, heat transfer characterisation and phase change problem formulation for design of LHTES systems [7]. Tay, Belusko & Bruno [8] experimentally investigated the thermal performance of a tube (vertical) in tank PCM system and formulated an equation that can be used for design purposes (also [9]). It was suggested that the resistance to heat transfer offered by the PCM was relatively small compared to those caused by increasing the heat transfer fluid flow rate or surface area of heat exchange. Gracia et al. [4] used experimental data from a domestic electrical hot water cylinder that incorporated encapsulated PCM in vertical tubes to validate a developed code for numerical simulation using finite volume method. Based on a numerical parametric analysis it was concluded that the timing and quantity of hot water demand are pivotal in defining the distribution of the PCM inside the tank. An optimisation of the heat transfer area and the associated heat release rate was essential to meet a set hot water demand.

Solar assisted cylindrical TES tanks using PCM have been studied both experimentally [10] and theoretically [1, 11]. A common finding from these studies was that the selection of PCM, cylinder radius, heat transfer fluid (HTF) mass flow rate and initial temperature of the HTF are controlling factors for optimum performance of the tank.

Thermal stratification is a common feature of solar water heaters where higher temperatures occur near the outflow level (top) of water while lower temperatures are observed at the inflow level (bottom). The existence of thermal layers within the tank is favourable as it helps concentrating the thermal energy at the hot water exit to meet the demand temperature requirements. Using PCMs in the upper part of the water tank augments the beneficial effect stratification of thermal energy and improves the performance of the system [12]. Heat transfer by thermal convection must be avoided (through design modifications) as it degrades the thermal stratification [5].

Farid et al. [13] performed measurements of heat transfer and used an effective thermal conductivity in a theoretical model based on one dimensional unsteady heat conduction equation in both phases to address the natural convection during melting and solidification of PCM in a vertical cylinder. The rates of heat charged and discharged were well predicted by the model as confirmed by experiments at different conditions. The authors recommended the use of physical properties of the melt PCM to characterise the effect of natural convection successfully. Experimental investigations by López-Navarro et al. [2] in an ice-water system showed that due to significant density difference, convection was the predominant heat transfer mechanism during phase change process. Natural convection heat transfer in microencapsulated PCM has also been experimentally evaluated by Zhang et al. [14].

Computational fluid dynamics (CFD) is a powerful tool for fluid dynamics and thermal design of PCM systems [15, 16]. In a modelling effort, Tay, Bruno & Belusko [15] developed a three-dimensional CFD model to analyse the transient heat transfer during PCM phase change in an arrangement where the HTF flowed through tubes in a tank of PCM. Natural convection was ignored in the CFD model and a disagreement was discovered between the behaviour of the model and the results from experiments. While natural convection caused the melting process to expedite, the overall performance of the system during PCM freezing was unaffected owing to dominance of heat transfer process by conduction.

According to Tay, Bruno & Belusko [17], as the phase change process (e.g. freezing) advances, the solid/ liquid boundary of the PCM moves away from the heat transfer surface and the phase changed PCM acts as

insulation reducing the heat transfer to the HTF. To decrease the thermal resistance, design modifications have been suggested such as extending heat transfer surface through use of fins/pins and impregnation of a metal matrix into the PCM [6]. Microencapsulation of the PCM is another popular technique.

In this paper, research work was carried with an ultimate aim of proposing an innovative solution that consumes low grade/inexpensive thermal and electrical energy (over-capacity) and provides dynamical storage until further use thereby replacing existing water tank systems and thus promoting the uptake of heat pumps and solar thermal solutions. A non-toxic hydrated salt based PCM has been inspected with a phase change temperature of 58°C. The PCM is a blend of inorganic salts (chiefly magnesium nitrate), nucleators, and stabilisers. The thermo physical properties of this PCM are given in Table 1.

Table 1: Thermo physical properties of the solid state PCM

Property	Value
Phase Change Temperature (solid-liquid)	58°C
Density	1505 kg/m^3
Latent Heat Capacity	145 kJ/kg
Volumetric Heat Capacity	218 MJ/m^3
Specific Heat Capacity	2.55 kJ/kg K
Thermal Conductivity	0.69 W/m K

The PCM selected had a high heat storage ratio (PCM to water), phase changing temperature close to typical domestic hot water supply temperature (approximately 50-60°C) and high thermal conductivity.

The overriding aim of this paper is to present the findings from a computational-experimental effort to improve our understanding of the heat transfer process and subsequently to the performance of an active LHTES-PCM based modern domestic heating system. The framework established in this investigation was utilised to capture the time-varying interplay between fluid flow and heat transfer.

2 Methods

2.1 Experimental apparatus

A 0.555 m internal diameter and 0.976 m high commercially sourced (Flamco Supastor stainless steel cylinder) domestic water heating tank with a holding capacity of 120 L was retrofitted to accommodate the PCM. A schematic representation of the experimental setup is shown in Figure 1.

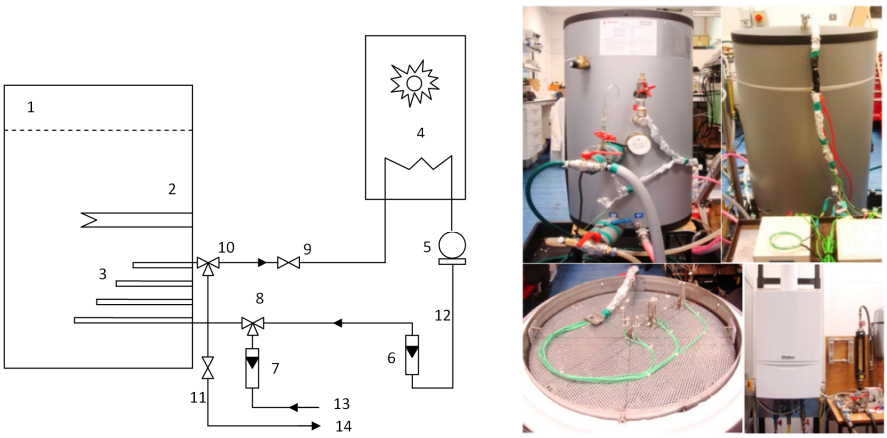


Figure 1: Schematic of the experimental apparatus

The tank (1) was completely filled with the PCM and was heated: (i) from the bottom with a 3 kW electric thermostat-controlled immersion heater (2) and (ii) hot water supplied in a 0.025 m internal diameter and 5.76 m long heating coil (3) from a 7-24 kW domestic condensing combination boiler (4) (Vaillant ecoTEC 824). The apparatus allowed flexible variation of the input variables: power input to the immersion heater, flow-rate and temperature of water through the heating coil. Water was chosen as the HTF. A 0.025 m thick polystyrene insulation was provided all around the tank to minimise heat losses to the surroundings.

The boiler was adjusted to generate heat between 7 and 24 kW. The hot water at 60-70°C was pumped out of the boiler by an electric circulator pump (5) supplied with the boiler and was circulated (in a closed

loop (12)) through the heating coil of the PCM tank to melt the PCM. To extract heat from the hot PCM, cold water (from laboratory main water supply (13)) was allowed to circulate (in an open circuit – the returning water was drained (14)) through the coil. Three way valves at the inlet (8) and outlet (10) of the coil were installed to switch between heating (tank-boiler) and cooling (tank-water mains) circuits. A globe valve (9,11) and a rotameter (6,7) were used in these circuits to control and record the water flow rate through the coil. The indicated flow rates read from the rotameter were corrected for density variations due to temperature using the standard mutilplier correction term $[\rho_0(\rho_s - \rho)/\rho(\rho_s - \rho_0)]^{1/2}$; where ρ_0 and ρ are the water densities at the calibrated (20°C) and the actual measuring conditions respectively, while ρ_s is the density of the stainless steel float. The maximum applied correction was 1.2% at 70°C. A variable resistor/rheostat was used to control the power input to the electric immersion heater. A plug-in mains power and energy monitor was used in series with the rheostat to record the power input value.

Nine 0.50 mm diameter mineral insulated K-type thermocouples were used to record temperatures at key locations in the tank: two thermocouples recorded the inlet and outlet temperature of water in the heating coil, and seven thermocouples were used to record the PCM temperature at different radial (0.08 m and 0.18 m from the centre of the tank) and axial (0.2 m, 0.4 m, 0.5 m, 0.6 m, 0.65 m from the top of the tank) positions within the tank. A 16-bit National Instruments USB-6259 data acquisition device connected to a National Instruments SCB-68 connector block (equipped with on-board low noise filtering system and a temperature sensor for cold junction compensation) was used to record the outputs from thermocouples. A dedicated personal computer was available for logging, saving and processing the thermocouple readings.

2.2 Numerical modelling and simulations

A numerical model of the PCM was developed based on the work by Rosler & Bruggemann [16]. This model applies a Boussinesq approximation for the PCM's density change with temperature and solves the resulting mass, momentum and energy conservation equations:

$$\nabla.(u) = 0 \tag{1}$$

$$\frac{D(\rho u)}{Dt} = \nabla.(\mu\,\nabla u) - \nabla^2 P + S_d + S_b \tag{2}$$

$$\frac{D(\rho h)}{Dt} = \nabla.(k\,\nabla T) \tag{3}$$

Here u is the velocity, ρ is the density, μ is the dynamic viscosity, P is the pressure, k is the thermal conductivity and h is the specific enthalpy of the fluid which is dependent on the liquid volume fraction (γ) of the PCM and the temperature (T). For numerical reasons Rosler & Bruggemann [16] modelled the phase change of the material as occurring in a fixed but narrow temperature range. That is, the coupling between the liquid volume fraction and energy conservation would require an iterative solution to update the volume fraction at each time step. However by applying a liquid volume fraction which is a continuous function of temperature allows its direct substitution into the energy conservation equation (Equation 3). The solidification of the PCM material was modelled with the addition of a Darcy type source term, S_d, in the momentum equation (Equation 2), such that:

$$S_d = \left[\frac{-C\,(1-\gamma)^2}{(\gamma^3+b)}\right] u \tag{4}$$

where C is a large constant and b is a small numerical constant to avoid division by zero. In this paper a value of 1.6e06 was chosen for C and 1×10^{-3} for b as it seemed to give stable results.

This computational model was implemented by Rosler & Bruggemann [16] in the open source CFD toolbox OpenFOAM® as an application titled meltFOAM. The application meltFOAM uses the PISO-SIMPLE [18] algorithm to calculate a solution to the pressure--velocity coupled momentum and continuity equations. In addition to the work by Rosler & Bruggemann [16], the meltFOAM application was modified to use a dynamic meshing library allowing mesh refinement in the vicinity of the melting front (see Figure 2(b)). This was undertaken partly because the authors were unsatisfied with the speed of the original meltFOAM application and secondly to try to tackle various numerical mesh artefacts seen in the solution.

Firstly, the speed of the original meltFOAM application was hampered by the need for a fine mesh resolution throughout the domain to capture the propagation of the melting front whereas for the majority of the simulation there is not actually much fluid activity in the solidified region, thus wasting valuable CPU time. Secondly, at the melting front a non-realistic fluid "jetting" effect was observed every time the melting front progressed by a row of cells. This was probably due to a large imbalance between the Boussinesq, S_b, and the Darcy, S_d, source terms as the Darcy force receded with the melting front. One way to tackle this was to make the cell size small in the vicinity of the melting front.

The hexahedral mesh was constructed using the OpenFOAM blockMesh and snappyHexMesh utilities. First a cylindrical blockMesh was created to model the inner volume of the hot water tank,

then the immersion heater and internal water heat exchange coils were added using CAD generated STL (STereoLithography) geometries and the snappyHexMesh utility (Figure 2(a)).

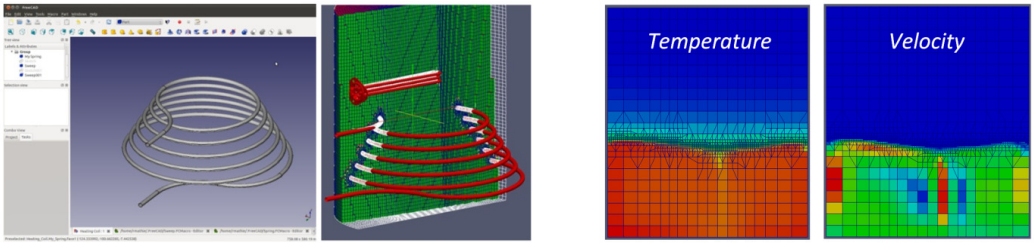


Figure 2: (a) Coiled heat exchanger geometry, and meshed PCM vessel, immersion heater and flow coil. **(b)** Mesh refinement showing propagation of the melting front in a cylinder with a heated base

The walls of the tank were assumed adiabatic and a no-slip boundary condition was applied to all surfaces, except the upper "surface" of the tank that was set as slip to approximate the free-surface of the PCM in the tank. Isothermal or isoflux boundary conditions were then set on the heating element and heat exchange coil depending on weather heat addition or removal was being modelled. The simulation was run on a 16-core Intel Xenon node of the Imperial College HPC cluster facility. Originally a mesh with 1.7M cells was tried however the simulation time of a week to achieve one hour of real time was considered impractical. A scaled down mesh of 170K cells, found to have a more reasonable simulation time (1 day to simulate 1 hour of real time), was finally employed.

2.3 Model validation

Experimental tests were carried out for charging and discharging conditions. During charging or heating tests, heated water (HTF) at approximately 70°C and a volumetric flow rate of 5 L/min was supplied from the boiler to the coil of the tank. A constant power input of 2.6 kW was given to the electric immersion heater. Following this, the temperature data from the thermocouples was monitored and the charging phase was sustained until a constant temperature was registered from all thermocouples. Discharging or cooling tests were performed by directing cold water from the laboratory water mains through the tank coil at a constant flow rate of 5 L/min. The electric immersion heater was switched off during these test runs. The completion of a discharging test was marked by equality in inlet and outlet temperatures of the HTF through the coil. Both charging and discharging experiments were performed for the tank filled with (i) water and (ii) PCM. This was done in order to conduct a comparative analysis of the nature of heat transfer and system performance. The results from this study are discussed in the next section.

3 Results and Discussion

The results generated by the CFD simulations were crucial in providing insight into the flow and temperature fields in the PCM within the heat storage tank. An initial CFD simulation in a simple geometry – a cylinder with a heated base – illustrated the melting behaviour of the PCM. The propagation of the melting front along with the associated temperature and velocity profiles can be seen in Figure 2(b). Stratification of the temperature profile and discontinuity in the thermal profile at the fluid-solid interface can be observed. A single convection cell is also evident from the velocity profile within the PCM melt.

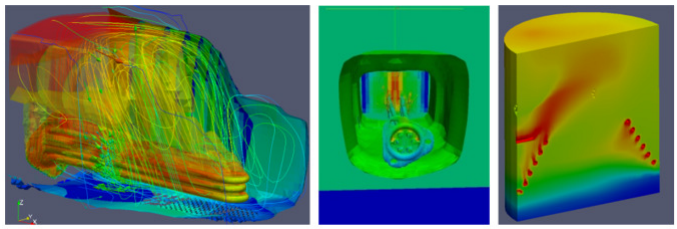


Figure 3: From left-to-right: 3-D image of melting front and velocity field around the immersion heater; melting front and temperature distribution around the heater; temperature profile when heating the coil

The melting rate of the PCM was investigated when either heat was applied with the hot water coil or when the immersion heater was switched on. The 3D CFD images (Figure 3) generated with the electric immersion and heating coil in place gives a qualitative description of the resulting flow phenomena, and propagation of the melting front. Ostensibly the flow pattern is similar to that in Figure 2(b) with the main convection cell being clearly visible in Figure 3(b). However the convection cell becomes highly skewed

with a large radial component. The melt front also proceeds faster at the extremity of the tank (close to the tank wall). Again with the coil, we see significant stratification of the temperature profile with the heat flux (or power (in Watts, W) as heat transfer area is constant), and hence the ability to input heat and melt the PCM, dropping once the melt front is below the coil. From Figure 4 it is evident that unlike water the rate of heat addition to PCM can be sustained and is relatively constant with charge time. This is because in the case of water the available temperature difference decreases with as the amount of heat in the tank increases, reducing the heat transfer rate while with the PCM the temperature difference between the heating element and the melting front is constant due to the phase change process of the PCM.

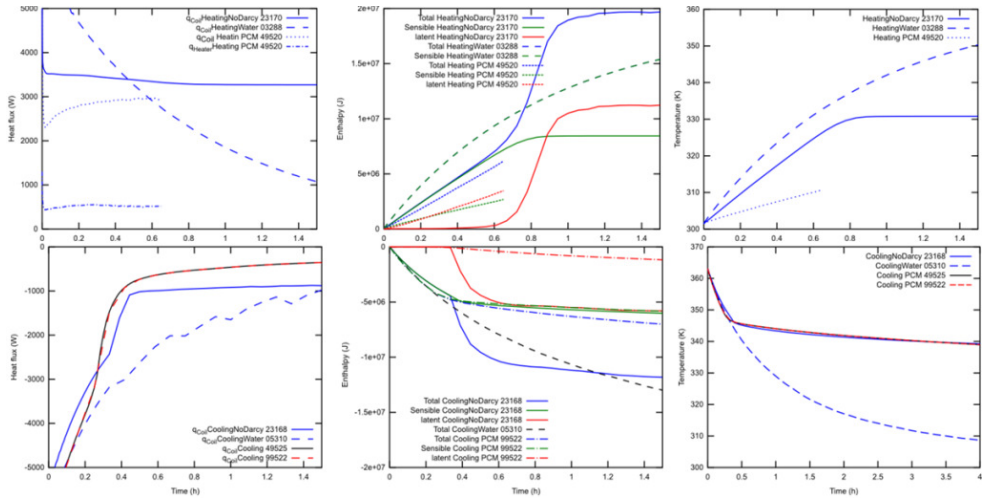


Figure 4: Heat addition (top row) and heat removal (bottom row) to/from the PCM tank**.** In the runs labelled "No Darcy" the solid phase was mobile showing the maximum possible heat transfer for PCM

As heat was supplied to the tank, the phase change started at the heater transfer surface of the electric immersion and heating coil. The melting front propagated away from the heat transfer surfaces. Initially, when the PCM was completely solid, heat transfer through the PCM was purely by conduction but with the growth of PCM melt volume, natural convection assisted the heat transfer process. Circulation of the PCM melt was observed around the heat transfer area and the PCM liquid fraction developed at a faster rate. Only melting of the PCM above the electric immersion and heating coil was observed in the tested time frame of the CFD simulation. The PCM below remained in the solid state. This could be because this portion of the tank received heat solely through conduction while the top PCM experienced a conjugate effect of conduction and convection. To summarise, the results from simulations of PCM melting show that natural convection enhances the speed of thermal energy storage.

Temperature curves were obtained for water and the PCM as a function of time from both simulations and experiments. From the charging simulations it was predicted that the temperature of the water should increase monotonically while the PCM temperature must only increase until the phase change temperature as following this phase change will occur at a constant temperature (Figure 4). Based on the temperature curves and thermo physical properties of the PCM, predictions regarding the heat flux and the total enthalpy of the tank were also made. During heating of water in the tank, it was expected that the heat flux from the electric immersion heater would remain constant (due to a constant input of electric power) while the heat flux from the HTF carrying heating coil would decay with time as a consequence of the decreasing temperature difference between the tube side and the tank side water. A similar behaviour was also expected for the PCM. Corresponding to the temperature and heat flux behaviour, the enthalpy content of the tank content was also predicted to increase with time (Figure 4).

An interesting observation from the charging simulations results was that the magnitudes of heat flux and enthalpy for water tank charging were considerably higher than the PCM tank charging. Also, for a given time, the heat flux to the water tank decreased while the heat flux in PCM tank remained constant. To validate these observations the experimental temperature distribution data within the tank was analysed - Figure 5(a). The temperature distribution in water tank clearly suggested existence of two thermal layers – one above the heat transfer surfaces and the other below them. The temperatures of both these thermal layers increased with time. On the other hand, thermal stratification was also observed in the PCM tank, but there the temperature distribution suggested existence of multiple thermal layers or regions (since flow visualisation was not possible therefore the exact shape of the solid-liquid interface cannot be determined with confidence but the temperature readings from spatially distributed thermocouples can be matched with the CFD simulation results for an approximate identification of the interface shape). The PCM above and in the immediate vicinity of the electric immersion heater and the heating coil melted while pockets of heated (but not to the point of melting) PCM existed in middle and base of the tank.

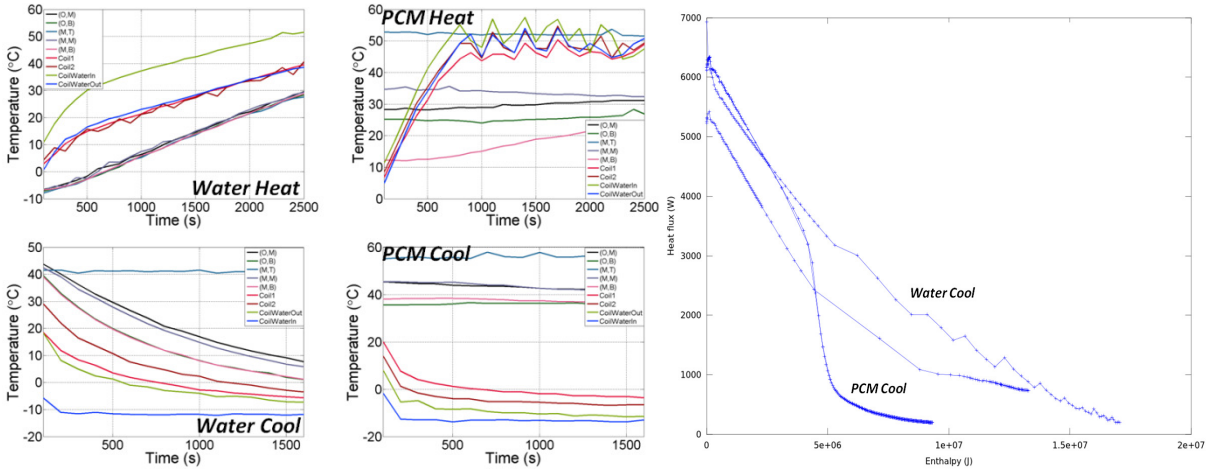


Figure 5: (a) Temperature (relative to 18℃ ambient) data from experiments. **(b)** Heat flux versus enthalpy removed from the tank

This distinct behaviour of the water and PCM tank charging may be attributed to the nature of material itself. Water is less viscous compared to the PCM (both in solid and molten state) and therefore natural convection caused a larger heat transfer augmentation in water carrying the heated water (even less viscous and dense than cold water) to the upper part of the tank and thus forming the two distinguishable thermal strata. This also resulted in higher heat fluxes (as heat transfer surface was constantly refreshed with cold water brought by the convection currents) and heat absorption by a larger volume of water. The constant and lower magnitude heat flux in the PCM tank was due to the slower conductive heat transfer that melted the PCM (majority of heat being stored as latent heat) leading to smaller temperature gradients as there was no physical movement of the PCM. Thus natural convection played a very small role in overall heat transfer process.

Similarly the discharging simulations (Figure 4) and experiments (Figure 5(a)) were analysed. From the simulation results, it was observed that the initial heat flux extracted for the PCM is comparable to the water tank, but declined subsequently due to the high fluid viscosity of the PCM melt and ice formation on the heat exchanger surfaces (Figure 6), which resulted in a significant temperature drop. Spatial temperature distributions from discharging experiments supported this observation. During discharging in water tank again two distinct thermal layers were observed. The upper portion of the water tank remained hot. The cooling water in the coil was only able to effectively cool the water in the lower portion of the tank. Natural convection would have played a role in the heat transfer process as the temperature decay was fairly uniform over a large spatial spread within the tank. On the contrary, cooling in the PCM tank was very ineffective (Figure 5(b)). During the entire duration of the discharging experiments, the temperatures were nearly constant suggesting quick ice formation i.e. PCM solidification on the tank coil when cooling water was introduced in it. The solid PCM around the coil offered a very high thermal resistance and nearly stalled further heat extraction from the tank.

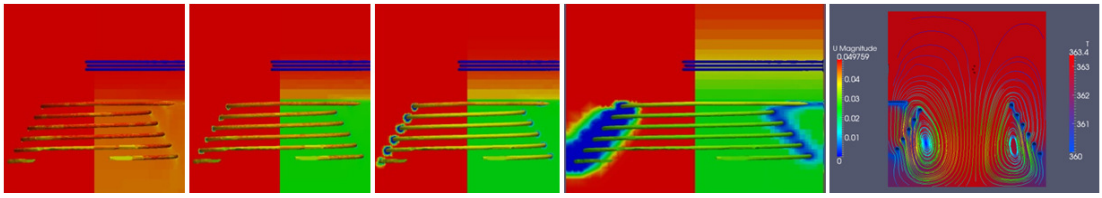


Figure 6: Image sequence (1-4) showing thermal stratification and ice formation on heat exchange coil. The left-hand side of each image shows the phase fraction (red-liquid/blue-solid phase), and the right-hand side the temperature distribution through the tank (red-hotter/blue-cooler regions)**.** Velocity contours around the coil as heat is extracted (Image 5)

Although the experimental tests show excellent agreement with the CFD results, the heat removal performance emerges as the limiting factor when compared to a water-based system. The PCM solution has the capability to store a large amount of heat effectively, but to achieve full utilisation of this advantageous feature the current apparatus (specifically the tank) design needs modification. Thus heat transfer enhancement between PCM and HTF is a key design concern and some previous works [17] have addressed this concern. In such studies, although higher heat transfer rates were achieved but the energy storage effectiveness and capacity was significantly lower compared to the current system as only a small fraction of the tank contained the PCM. Undoubtedly, there exists a competition between the

thermal storage capacity and the heat transfer rates which is controlled by the design of the system. This work was an attempt to achieve the maximum thermal storage capacity for a new type of PCM. Future work in such a system must introduce design improvements to the heat transfer arrangement (through use of fins, changing coil locations, and employing the PCM as emulsions/encapsulated spheres) while ensuring that the PCM is contained in the maximum possible volume of the tank.

4 Conclusion

This paper is concerned with computational prediction and experimental validation of the dynamic behaviour and performance of a thermal storage system for domestic applications, based on the use of a hydrated salt PCM. A high-fidelity computational prediction tool (OpenFOAM) was implemented to analyse the melting front of the PCM. The CFD results were validated through experiments in a retrofitted commercial heating tank. The thermal extraction and heating rates for the PCM tank were compared to a water filled tank. It was observed that the initial heat flux extracted for the PCM was comparable to the water tank, but declined subsequently due to the high fluid viscosity of the PCM melt and ice formation on the heat exchanger surfaces, which resulted in a significant temperature drop. During melting the heat flux into the PCM tank was constant whereas the heat flux into the water tank decreased significantly as the tank water heated up. The tests showed excellent agreement with the CFD results with the heat removal performance being the limiting factor when compared to a water-based system. It was confirmed that the PCM solution has the capability to store a large amount of heat effectively. Possible design improvements to the heat transfer arrangement are suggested through use of fins, changing coil locations, and employing the PCM as emulsions/encapsulated spheres.

Acknowledgements

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