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## A Review of Integrated Phase Change Materials for Evacuated Tube Solar Collector System

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### Abstract

This study presents a short review of research outcomes of low and medium temperature solid-liquid Phase Change Materials (PCMs) that are used for latent heat storage. The aim is to determine the appropriate materials for integration with the Evacuated Tube Solar Collector system (ETSC) in order to stabilize the intermittent temperature fluctuations and extend the operating hours. The study provides an insight into recent efforts to integrate PCMs with solar thermal systems, focusing on providing solutions to three of the disadvantages of PCMs, namely thermal stability, thermal conductivity and supercooling. The study uses the 'Papadimitratos' approach of integrating PCMs into the ETSC to provide an acceptable degree of validation in the selection of materials.

### Keywords:

Thermal Energy Storage; Phase Change Materials; Solar Thermal; Evacuated Tube Solar Collectors

### Introduction

The development of solar, wind and other renewable energy technologies for sustainable power generation can play a vital role in a time of growing heat and electricity demand and amidst the climate change debate. However, these resources are intermittent and result in major challenges due to the daily and seasonal variations in heat and electricity demand. Thus, future energy systems require the integration of renewable sources with short and long-term storage in order to balance the daily and seasonal mismatch and to reduce the reliance on conventional energy sources. This will result in huge potential to increase the utilisation of variable resources, improving the reliability and performance of energy systems (Fleischer, 2015; Garg, et al., 1985).

Solar energy is one of the promising renewable energy sources around the world. Integrating with energy storage means allowing the storage of excess energy and providing it whenever solar energy is not available. This would improve the utilisation of collected solar energy (Papadimitratos et al., 2016).



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## (I) Evacuated Tube Solar Collector

One of the solar systems that has been widely developed is 'Solar Thermal', in particular, the Solar Water Heater (SWH) application. A solar collector, which is an important part of solar thermal systems, is a special kind of heat exchanger that converts the solar radiant energy into heat. Two collectors, namely: Flat Plate Collectors (FPCs) and Evacuated Tube Solar Collectors (ETSCs), are used in Domestic Hot-Water (DHW), cooling absorption chillers and space heating applications (Kalogirou, 2003; Ricci et al., 2015). The FPCs system is low cost, has a longer life expectancy and gives better year-round performance compared to the ETSCs whereas the ETSCs system is more efficient at lower working temperatures and provides higher fluid outlet temperatures. Furthermore, the ETSCs are less sensitive to sun angle and orientation compared to FPCs. Despite the fact that the design of ETSCs is complex due to the vacuum between the glass tubes, it has excellent thermal insulation as the convection losses are reduced compared to the FPCs (Kim and Seo, 2007).

The ETSCs system is a time-dependent energy source which can be maximised through its supply with a Thermal Energy Storage (TES) system. Among the sensible and latent heat storage methods of TES that are associated with solar thermal systems, the Latent Heat Storage (LHS) system, in particular, the solid-liquid phase change materials (PCMs), has attracted interest for a long time (Abhat, 1983; Prakash, 1985; Boy et al., 1987; Rabin et al., 1995; Zalba et al., 2003; Halawa et al., 2005; Mettawee and Assassa, 2006; El-Qarnia, 2009; Kenfack and Bauer, 2014; Pielichowska and Pielichowski, 2014; Sharma et al., 2015; Englmaier et al., 2016; Tri et al., 2017; Khan et al., 2018). This is due to the higher energy density compared to Sensible Heat Storage (SHS) resulting in less material-mass required and therefore, smaller tank volume. Another feature is that the solid-liquid transition results in only small density changes compared to the liquid-vapour phase change process with a smaller temperature difference between storing and releasing heat compared to the SHS method (Jong Choi, 1995; Sharma et al., 2009).

The integration of PCM with solar thermal systems means either by developing TES in the collector, in the water tank or in a separate container. The integrated TES-ETSC system was achieved by developing the storage in the header (manifold) [studied by Naghavi et al., (2015); Mehla and Yadav, (2015); Mehla and Yadav, (2016)] or achieved inside the inner cavity (in-tube) [studied by Papadimitratos et al., (2016); Xue, (2016); Abokersh et al., (2017)], refer to Figure 1. Interestingly, Papadimitratos and others (Papadimitratos et al., 2016) incorporated organic materials: Trtriacontane with a 72°C melting temperature and Erythritol with a 118°C melting temperature, into the ETSC system in order to address the evening peak demand. They claimed an efficiency improvement of 26% compared with a

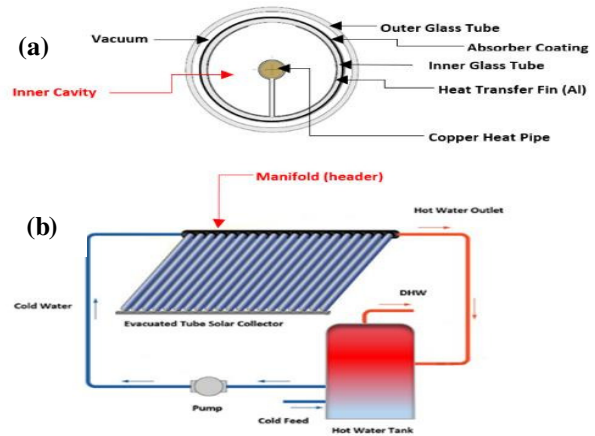


Figure 1. Cross-sectional and Schematic Configuration of the ETSC (a) Inner Cavity (IC) Storage (b) Manifold (header) Storage



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standard system in the normal operation (with draw-off water) and 66% in the stagnation mode. They selected these materials based on their chemical and physical properties, low cost and availability. However, the temperature of the Inner Cavity (IC) storage in the ETSC system may reach more than 150°C because of the vacuum layer. This makes the material stability a real challenge for the heat storage functionality inside the IC. As a result, overheating should be avoided to protect the PCM from thermal degradation.

## (II) Challenges for Implementation of PCMs

A large melting enthalpy and a suitable phase change temperature are the two essential requirements of a PCM. However, the thermophysical properties of the material must be considered for most applications in order to select the appropriate material for the TES system. Table 1 shows the thermophysical properties required which have been grouped into physical, technical and economic requirements (Abhat, 1983; Sharma et al., 2009).

While there is great potential in using PCMs with many different applications, by identifying and overcoming the major issues, it will prove beneficial to combine TES with other applications e.g solar thermal system. several researchers have reviewed the disadvantages of PCMs for TES with different applications (Rathod and Banerjee, 2013; Safari et al., 2017; Lin et al., 2018). However, among these applications, there are no critically analysed articles dealing with these issues of IC storage within the ETSC application.

This paper reviews three of the problems of solid-liquid PCMs for TES applications in order to determine the potential role of integrating PCMs within the ETSC system. This will be achieved through:

- Phase Change Materials
- Long Term Stability
- Thermal Conductivity
- Supercooling

In addition, it also reviews the simulation studies to identify the behaviour of these issues through their useful life and thermal cycles.

### Phase Change Materials

PCMs are materials that undergo the solid-liquid, liquid-vapour or solid-solid phase transformation during the process of absorbing or releasing heat from surroundings while remaining at nearly a constant temperature (Agyenim et al., 2010;

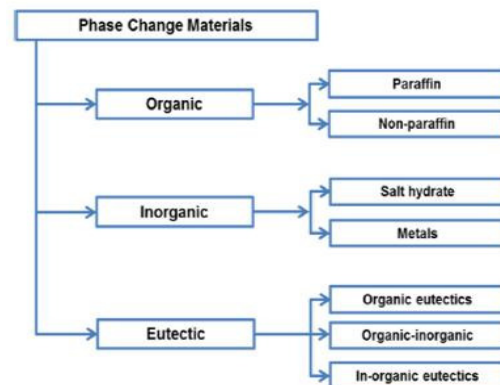


Figure 2. Classifications of Phase Change Materials (Mohamed et al., 2017)

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Physical	Technical	Economic
Large phase change enthalpy	Low vapor pressure	Low cost
Suitable phase change temperature	Small volume change	Good recyclability
Cycling stability	Chemical stability of the PCM	
Little subcooling	Compatibility of the PCM with other materials	
Good thermal conductivity	Safety constraints	

Table 1. Thermophysical Properties (Requirements) of the PCMs (Abhat, 1983)



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Fleischer, 2015). In this paper, the focus will be on solid-liquid PCMs only. The transformation process is known as melting-solidification, where the energy absorbed or released during this process is known as the latent heat of fusion. The storage capacity ( $Q$ ) of using solid-liquid PCM as a storage medium can be given by:

$$Q = \int_{T_i}^{T_m} mC_p dT + ma_m \Delta h_m + \int_{T_m}^{T_f} mC_p dT$$

Where  $T_i$ ,  $T_m$ ,  $T_f$  are the initial, melting and fluid temperatures (K);  $m$  is the material mass (kg);  $C_p$  is the material specific heat capacity (kJ/Kg.K),  $a_m$  is the material melted fraction, and  $\Delta h_m$  is the latent heat of fusion (kJ/Kg). Notice that the first and last expression correspond to the sensible heat stored which results in a change in temperature within the material, while  $[ma_m \Delta h_m]$  is the latent heat stored or released at a constant temperature. The latent heat would not be exploited without the solid-liquid transition which means the energy stored would be on a sensible heat only. Furthermore, the latent heat energy is governed by the enthalpy of fusion (kJ/kg). Thus, the process is mass-based which means the melting process of the material depends on the mass of the material in the design of the storage application whereas, the rate of the melting is governed by the operating condition of the system (Fleischer, 2015; Mohamed et al., 2017). Figure 2 (on page 381) illustrates the classification of the PCMs which are divided into organic, inorganic and eutectic materials. Table 2 shows the main different features between them and a detailed comparison between organic and inorganic materials is presented by Zalba et al., (2003); Farid et al., (2004). Solar water heating based on PCMs has been the subject of several investigations. Boy et al., (1987) made one of the earliest studies on integrated PCMs with solar collectors. They used salt hydrate (inorganic), claiming that the improvement of the thermal efficiency occurred by incorporating the appropriate PCM-device.

Cabeza, (2006) added encapsulated PCMs-graphite on the top of the water tank with stratification in order to test the PCM behaviour in real conditions. Subsequently, (Mazman et al., 2009) developed a short-term LHS on the top of cylindrical water tank (0.176m diameter and 0.316m height) using a mixture of 80% Paraffin and 20% stearic acid. They claimed less heat loss from the top of the water tank and water temperature on the top increased by 3-4°C in 15minutes. Mettawee and Assassa, (2006) used paraffin-wax (organic) when investigating a new compact design of PCM with the FPC. They tested a 1.3m length collector fixed at 45°C with a single water pipe at a specified, central location to maximise heat intake from the surroundings, filled with the paraffin wax. They found the heat transfer increased with increased layers of thickness during the melting process. Riffat et al., (2006) also used paraffin wax in varying proportions with water, when conducting an experimental study in order to investigate the best performance of TES combined with the ETSC system. Several other applications of

	Organics	Inorganics
Advantages	Non-corrosive Low or none under-cooling Chemical and thermal stability	Greater phase change enthalpy
Disadvantages	Lower phase change enthalpy Low thermal conductivity Inflammability	Undercooling Corrosion Phase separation Phase segregation, lack of thermal stability

Table 2. Main Differences Between Organic and Inorganic Materials (Zalba et al., 2003)





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PCM-TES combined with domestic, solar thermal systems are reviewed by Kenisarin and Mahkamov (2007); Sharma et al., (2015); Mohamed et al., (2017) who reviewed the organic and inorganic solid-liquid-PCMs for TES and Salunkhe and Jaya Krishna, (2017) reviewed the PCMs used for low-temperature TES (25-80°C). To the best of the author's knowledge, the integration of PCM with the ETSC system was not effectively developed nor recorded in the literature compared to the FPC. Therefore, this review includes both FPC and ETSC systems integrated with LHS, but the focus will be on the development of ETSC system. The requirements of thermal energy storage and the state-of-the-art thermal energy system has obvious potential with ETSC technology.

## Long-term Characteristics

Among the several criteria that should be considered when selecting the PCM for TES, the material should exhibit stability, both chemically and physically. This should be during repeated thermal cycling with repeatable and consistent melting and freezing. Also, more attention should be considered when designing TES using the solid-liquid PCMs with the liquid phase leakage. In addition, some of these materials tend to be corrosive or chemically incompatible with the material of the storage container.

### (I) Thermal Stability

Materials with high melting temperatures tend to have higher costs and poor stability during thermal cycling. Less attention was paid to studying the thermal stability of the PCMs during the operating life-time. Kenisarin, (2010) reviewed high-temperature PCMs for TES. The study explained how each application supported the material life-time. Rathod and Banerjee, (2013) reviewed the thermal stability for some PCMs including common methods used by researchers to establish the thermal properties of PCMs; for example, the Differential Scanning Calorimetry (DSC) technique which is used for measuring melting temperature (PCM temperature range) and latent heat of fusion. They also concluded that paraffins have good thermal and chemical stability after a number of thermal cycles. This was approved by Shukla et al., (2008) who conducted an experimental study testing the thermal stability of some promising organic and inorganic materials for TES systems such as paraffin-wax with different melting temperatures. For solar thermal systems, there is one cycle a day known as normal cycle which can be tested by using an oven in the laboratory. Thus, in their study, they used an oven to conduct 1000 test cycles. Erythritol was one of the organic materials tested which has a higher enthalpy of 339kJ/kg. They found it degraded after the 500th cycle where the melting and freezing temperatures decreased by 9-10°C and about 35-40kJ/kg in the capacity. Figure 3 shows the DSC curve for 1000th for Erythritol. This material was used by Papadimitratos et al., (2016) for the in-tube TES integrated with the ETSC system.

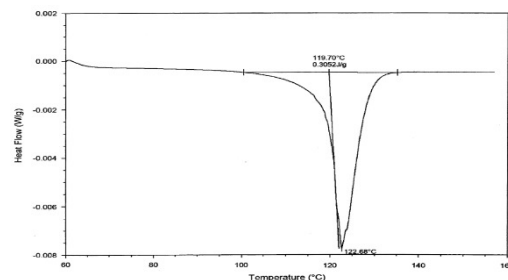


Figure 3. DSC Curve of Erythritol for 1000th Thermal Cycle (Shukla et al., 2008)



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## (II) Liquid Phase Leakage and Incompatibility

The transition to a liquid state creates specific design challenges, therefore, encapsulation of PCMs is usually essential. Encapsulation technique controls the volume change during the solid-liquid phase transition as PCMs exhibit around 15-20% volume change (Fleischer, 2015). Also, it reduces corrosion, prevents possible interaction with the environment and has the benefit of increasing the compatibility of PCMs with storage materials. Furthermore, it helps to increase the heat transfer surface, thus, improving the thermal conductivity (Amin et al., 2014). Figure 4 shows the techniques of PCM encapsulations; the classification depends on the size of the encapsulating. For example, Microencapsulation is encapsulated PCMs with diameters (1 $\mu$ m to >1 $\mu$ m) which is considered for solar thermal energy storage systems (Jacob and Bruno, 2015; Mohamed et al., 2017). Jamekhorshid et al., (2014) reviewed the techniques used for Microencapsulation of PCMs. Su et al., (2017) formulated microencapsulation-PCM (MPCM) for solar hot water storage systems using 5 samples of paraffin wax (shell-material) and melamine-formaldehyde resins (core-material). They performed a theoretical evaluation for the thermal properties with the highest enthalpy of MPCM of 126 kJ/Kg achieved. The results showed Solar Water Heater (SWH) with MPCM has less volume and large density. However, it has less thermal conductivity compared to the standard SWH system with water tank storage. Also, Jacob and Bruno, (2015) reviewed encapsulation for shell materials with high thermal stability under higher temperatures. In addition, a detailed review by Salunkhe and Shembekar, (2012) investigated the thermal performance of LHS with encapsulated PCM. They concluded that lower shell and higher conductivity results in rapid melting of the encapsulated-PCM, where conduction and then natural convection are dominant during charging and discharging. Furthermore, an experimental study done by Fang et al., (2017) used Microencapsulation-PCM (MPCM) as a heat transfer medium in order to investigate the ETSC efficiency. Regarding the IC storage, Papadimitratos et al., (2016) observed a 13% volume expansion for the Erythritol during solid-liquid transition inside ETSC. They introduced a new approach by placing the PCM in aluminium bags after considering the volume change.

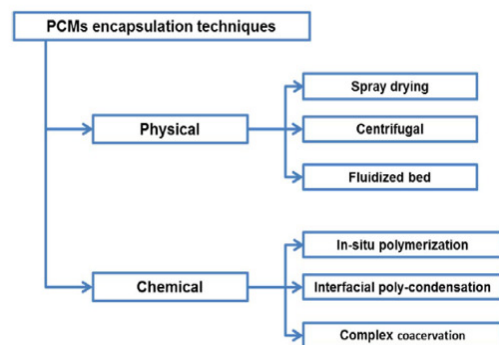


Figure 4. Microencapsulation Techniques of PCMs (Mohamed et al., 2017)

## Thermal Conductivity

Generally, PCMs have high viscosity and low thermal conductivity where it is desirable for the storage material to have high thermal conductivity in order to prevent thermal bottlenecks and degrading in the system performance (Lin et al., 2018). As mentioned earlier, encapsulated PCMs is one of the techniques used to enhance the thermal transport. The second technique is by adding other materials with high thermal conductivity. This is



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divided into three methods used to improve the thermal conductivity and diffusivity in PCMs, illustrated in Figure 5 (Fleischer, 2015). For SWH system, Narayanan et al., (2017) used 0.5 wt% nanographite (NG) with a mixture of two organic materials (Eutectic gel PCMs) with two different applications, including an indirect solar water heating system in order to investigate the effect of the NG additive in the melting rate and latent heat of storage materials. They claimed a reduction of 3 minutes in melting time and 93% overall improvement in the melting rate. Thus, using the composite for SWH system results in an overall efficiency improvement with less charging time and a low discharging rate. Al-Jandal and Sayigh, (1994) conducted experimental and analytic studies to investigate the performance of solar tube collector integrated with TES (PCM-Stearic acid). They used aluminium fin which reduced the melting time. Several researchers (Shabgard et al., 2010; Nithyanandam and Pitchumani, 2011) used heat pipes to enhance thermal transport into the mass of the PCMs for TES during charging and discharging. Also, Khalifa et al., (2013) conducted an experimental study using six copper pipes in series (80mm diameter each) inside a paraffin-wax container for SWH-TES system in order to investigate the solar collector system performance. The results showed an improvement of the integrated system compared to the standard SWH system. Papadimitratos et al., (2016), used microencapsulated paraffin-PCMs within the ETSC, in conjunction with silicon oil, as an excellent medium for paraffin in order to enhance the convection heat transfer. However, there is limited investigation into this in the literature (Papadimitratos et al., 2016).

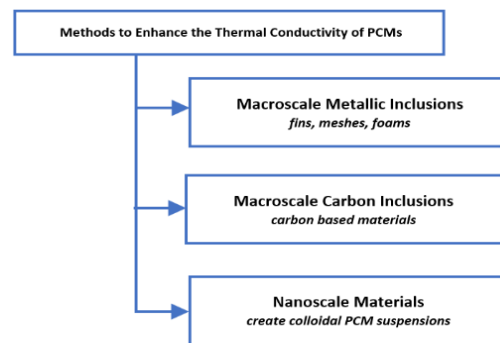


Figure 5. Methods of Enhancement of Thermal Conductivity of PCMs

## Supercooling

Supercooling, or subcooling, is when a liquid-PCM is cooled below its melting temperature while still remaining in liquid phase (metastable state). Therefore, the freezing process starts a few degrees lower than the actual solidification temperature point. This means there is a sensible heat release for a prolonged period until the actual freezing starts (Mehling and Cabeza, 2008). A low degree of supercooling is always desirable for short-term LHS. This represents the temperature difference between melting and freezing points (crystallization). A large degree of supercooling means that less heat will be stored during the actual solidification as more heat is released during the initial supercooling (Zhang et al., 2010). For example, salt hydrates are inorganic PCMs with high thermal conductivity and an energy density of around 350 MJ/m<sup>3</sup> which makes them competitive to use for LHS. However, these materials have a

No. of cycles	Measured by data logger		Measured by DSC	
	Melting temperature (°C)	Freezing temperature (°C)	Melting temperature (°C)	Latent heat (kJ/kg)
0	117	112	117	339
100	120	115	122	340
300	115	105	115	339
500	110	106	106	312
1000	107	105	119	305

Table 3. Melting-Freezing Temperatures of Erythritol (Shukla et al., 2008)





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tendency to melt incongruently which, as a result, causes phase segregation. This affects the storage thermal performance and reduces its lifespan in terms of a number of thermal cycles. A study by Sandnes and Rekstad, (2006) investigated supercooling behaviour on four salt hydrates products: disodium hydrogen phosphate dodecahydrate, sodium acetate trihydrate and a commercial product STL-47, by obtaining enthalpy-temperature curves. The results showed that with the large degree of supercooling, the maximum temperature reached was reduced as well as the enthalpy difference (heat capacity in solid phase is less than in the liquid phase). Reducing the degree of supercooling is not clarified in the literature, however, there are a few methods used in order to alleviate the supercooling for PCMs. Dannemand et al., (2016) used two methods on sodium acetate trihydrate (SAT) to investigate phase separation. The first method was by using 9% excess water with 199.5 Kg of SAT to avoid the phase separation. The second method was by adding a thickening agent of 1% carboxymethyl cellulose (CMC) to the 220 Kg of SAT (nucleating agents such as copper and titanium oxide). They claimed that due to the high viscosity of the SAT and CMC mixture, its charging was significantly low. Its energy discharge after solidification was nearly stable on 205 kJ/kg, where the energy discharge after solidification of supercooled SAT with extra water was gradually reduced from 194 kJ/kg in the first cycle to 188kJ/kg and 179 kJ/kg after 5 and 20 test cycles. In general, stable supercooling was not always achieved. Other methods were used to alleviate the supercooling including encapsulation of storage materials (Hong et al., 2011) and modification of the chemical composition of the storage materials (Xu et al., 2017). A detailed review of supercooling on salt hydrates, paraffin-wax and fatty-acids was conducted by Safari et al., (2017).

As mentioned earlier, Erythritol was used by Papadimitratos et al., (2016) for the integration of LHS within the ETSC, Shukla et al., (2008)

conducted tests investigating its thermal stability. They claimed the supercooling behaviour appeared in some of the thermal cycles of Erythritol with variations in the temperature range: see Table 3 (on page 385). The maximum supercooling of 14°C was observed in the 9<sup>th</sup> thermal cycle. Figure 6 shows the temperature vs time during the 1<sup>st</sup>, 9<sup>th</sup> and 1000<sup>th</sup> cycles. This recorded a sudden drop then increase in temperature during the freezing process before it decreased to the ambient temperature.

## Modelling

Rabin et al., (1995) developed a theoretical model to study the charging process of integrated salt-hydrates PCM in a layer of stationary heat transfer liquid. Their study claimed that two thirds of the incident solar radiation on the collector can be stored. Whereas, Halawa et al., (2005) studied the charging and discharging of several layers of PCM slab, they discovered the stored or released heat in the final melting or freezing period is not useful. Ghani et al.,

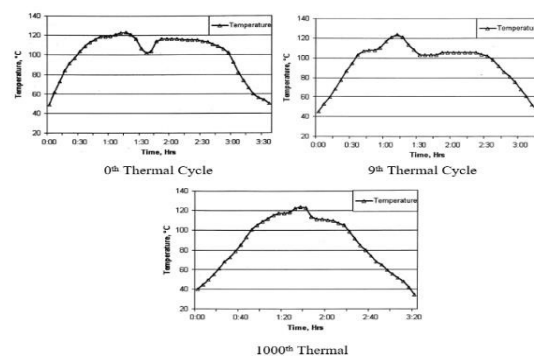


Figure 6. Temperature vs Time for Erythritol for 1<sup>st</sup>, 9<sup>th</sup> and 1000<sup>th</sup> Thermal Cycles (Shukla et al., 2008)



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(2018) developed a dynamic neutral network in order to model the non-linear operational characteristics of a LHS system.

For solar thermal systems, Khalifa et al., (2013) conducted a mathematical model to validate the experimental data of a separate storage container for the SWH system using paraffin-wax. Also, El-Qarnia, (2009) used the same approach to validate the experimental data of three storage medium-PCMs in order to predict the thermal behaviour and performance of a solar-LHS. Naghavi et al., (2015) and Nithyanandam and Pitchumani, (2011) developed a mathematical modelling integrated PCM into the manifold of the evacuated tube heat pipe collector system.

## Discussion and Conclusion

It is not easy to find a material with high enthalpy, specific heat and thermal conductivity. However, the melting point is the primary consideration when selecting PCM. It is also important to know that the applied heat flux and the temperature difference from the heat source to the melt point will govern this process of latent heat.

In literature, there is less compatibility data for the PCMs. However, the storage material used in TES should be chemically resistant. The ullage space must also be considered during the design as it creates an air gap in the container and can result in a degradation of thermal performance. The use of additive with PCMs to help the heat quickly diffuse into the PCMs is desirable. However, the composite mixture should not lead to decreasing their thermal stability. In most of the studies, PCMs are integrated with SWHs as a separate storage tank. So, due to the limited volume of the in-tube storage (2 kg) within the ETSC system, it is important to choose the right method for enhancing the thermal conductivity.

The degree of supercooling is important to determine the percentage of the supercooled liquid that would be crystallized, however, there is no uniform standard to select a specific additive or method for this. Therefore, there is potential to use numerical methods to investigate the heat transfer during phase change. In TES systems, reducing the degree of supercooling is the main objective for short-term LHS, but it is desirable for long-term storage as remaining phase transition energy can be stored for extended periods of time without further heat loss.

So, it can be concluded that successful utilization of the latent heat energy storage system depends on the thermal reliability and stability of the PCMs. There is a concern that needs to be addressed for these challenges to be overcome. Future investigation should include a modelling approach for IC-LHS storage within the ETSC in order to investigate the PCM charging and discharging process inside the tube as, currently, there is no study in the literature which covers this aspect. Also, the investigation should include the use of eutectic materials as a storage medium, as they have potential to be used for LHS system. However, the phase separation problem must be considered as a priority. Furthermore, the improvement of the ETSC systems' design will be based on geographical location, taking into consideration normal, stagnation and freezing temperatures, and how these affect the storage materials' long-term stability.



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