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A Review Study on the Thermo Physical Properties and Storage Applications of Phase Change Materials

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ABSTRACT

Phase change material as a thermal energy storage medium has been widely incorporated in various technologies like solar air/water heating, buildings, and desalination for efficient use and management of fluctuating solar energy. Temperature and thermal energy requirements dictate the selection of an appropriate phase change material for its application in various engineering systems. This paper reviews some of the thermo physical properties of the phase change materials and their applications for thermal storage. Thermal cycles up to 300 are performed to investigate melting and solidification reversibility as well as degradation over time. Paraffin waxes have reversible phase change with no degradation of thermo physical properties over time. Melting/solidification temperature and thermal energy storage capabilities make them suitable for their application as a thermal energy storage medium, in high temperature vapour compression, multi-stage flash and multi-effect distillation processes of non-membrane based indirect desalination systems.

Keywords: convection; heat conversion; longitudinal fins; phase change materials (PCMs); thermo physical

1. INTRODUCTION

Phase change materials (PCM) are a group of materials which exchange large amount of heat as latent heat within a narrow temperature range of phase transformation. Since the first

application of PCM by NASA in aerospace field, thousands of single materials and mixtures of two or more materials have been investigated for their use as PCM in areas like solar energy/waste heat storage, load shifting, and power saving, textiles, passive-cooled shelters, energy-efficient buildings, cooling technology for electronics, transport containers for food and medicines, human comfort, and energy conservation through energy storage, etc. [1].

The current interest in PCM technology on solar heat storage for space heating can probably be traced back to its earlier respective work just after World War II. More volume efficient alternate approaches than sensible heat storage, invariably led to the advent of latent heat systems. The early studies of latent heat storage focused primarily on the fusion-solidification of low cost, easily available salt hydrates [2]. After the phase change, they have a tendency to super-cool, and the components do not melt congruently. So, these phenomenon of super-cooling and phase separation often determined the thermal behavior of these materials and caused random variation or progressive drifting of the transition zone over repeated phase change cycles. Although certain significant advances were made till then, but major hurdles still remain towards the development of reliable and practical based storage systems using salt hydrates and similar inorganic substances [3,4]. The inherent problems in inorganic PCMs have led to an interest in a new class of materials: low volatility, anhydrous organic substances such as polyethylene glycol, fatty acids and their derivatives and paraffins. These materials weren't accepted earlier because they happen to be more expensive than common salt hydrates, and they have comparatively lower heat storage capacity per unit volume. It has been realized by now that these materials have certain advantages like physical and chemical stability, good thermal behavior, and adjustable transition zone.

Not all PCMs can be used for latent heat storage. An ideal PCM should have a high heat of fusion and thermal conductivity, high specific heat capacity, small volume change, non-corrosive, non-toxic and possess little or no decomposition or super-cooling. In building applications, only PCMs that have a phase transition close to human comfort temperature close to 20 °C can be used [5]. Table 1 shows the possible candidates from the many hydrated salts and organic PCMs for application in buildings.

Table 1. Hydrated salts and organic PCMs

PCM	Melting Point (°C)	Heat of Fusion (kJ/kg)
CaCl ₂ ·6H ₂ O Calcium chloride hexahydrate	29.7	171
45% CH ₃ (CH ₂) ₈ COOH 55% CH ₃ (CH ₂) ₁₀ COOH 45/55 Capric–lauric acid	17-21	143
CH ₃ (CH ₂) ₁₁ OH Dodecanol	17.5–23.3	188.8
KF·4H ₂ O Potassium fluoride tetrahydrate	18.5–19	231

$\text{CH}_3(\text{CH}_2)_{16}\text{COO}(\text{CH}_2)_3\text{CH}_3$ Butyl stearate	18–23	140
$\text{CH}_3(\text{CH}_2)_{16}\text{CH}_3$ Tech. grade octadecane	22.5–26.2	205.1
$\text{CH}_3(\text{CH}_2)_{12}\text{COOC}_3\text{H}_7$ Propyl palmitate	16–19	186

2. WORKING OF PHASE CHANGE MATERIALS

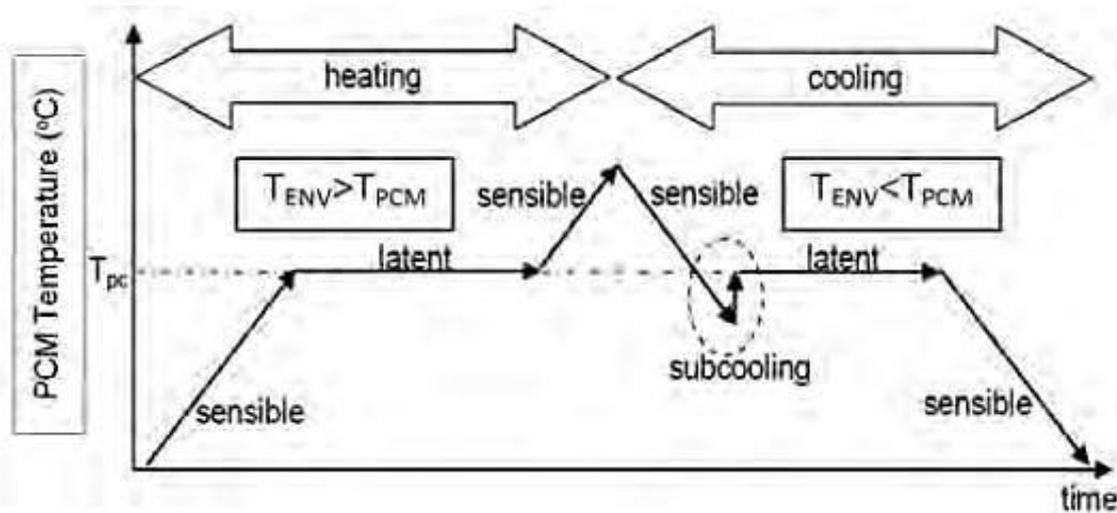


Figure 1. Schematic diagram, showing working of PCM

Working of PCM is schematically depicted in Figure 1. When heat is supplied to PCM from outside, it absorbs a large amount of latent heat at phase change temperature in breaking internal chemical bonds. In reverse cooling cycle, for phase reversal to start, temperature of PCM has to go down below phase change temperature (known as sub-cooling or under-cooling) to overcome the energy barrier required for nucleation of second phase. Once phase reversal starts, temperature of PCM rises (due to release of latent heat) and subsequent phase reversal takes place at phase change temperature by releasing back the latent heat to environment. Requirement of sub-cooling or under-cooling for phase reversal is an important property of PCM governing its applicability in particular applications. Latent heat of PCM is many orders higher than the specific heat of materials [6]. Therefore PCM can store 2-3 times more heat or cold per volume or per mass as can be stored as sensible heat in water in a temperature interval of 20 °C. As heat exchange takes place in narrow band of temperature the phenomenon can be used for temperature smoothening also. Important properties of PCMs other than latent heat, phase transformation temperature and sub-cooling requirement

are thermal conductivity, cyclic stability, congruent melting of PCM, and little volume change during phase change, low vapor pressure, chemical stability and compatibility of PCM with other materials like plastics. In addition to these technical requirements, safety concerns, low cost, easy availability, and good recyclability are important criteria in selecting a suitable PCM for a particular application. It is difficult to get a PCM ideal for a particular application. Certain amount of tradeoff between different properties is therefore essential [7]. Selection of a suitable PCM, determination of its quantity requirements based on heat-balance calculations, packaging of PCM, design of heat exchanging and heat distribution surfaces are other important steps in developing an effective PCM-based device or product.

3. PCMs EMBEDDED IN METAL FOAM

In many applications, thermal energy storage is required to receive, store and subsequently release heat. The major disadvantage of PCMs is their low thermal conductivities, which dramatically slows the phase change process and causes a wide temperature distribution within PCMs. Metal foams presents an order of magnitude higher thermal conductivity than PCMs. At the same time the random internal structure and high porosity of metal foam can enhance and accelerate the phase change process without significantly reducing PCMs' heat storage capacity [8]. The distribution of foam ligaments in PCMs makes the melting and solidification processes more uniform. There are many kinds of metal foams have been used in phase change materials, such as aluminum foam, copper foam and Nickel foam.

4. PCMs EMBEDDED IN ALUMINIUM FOAM

The development of a plate like structure thermal energy storage composite consisting of a central core of foamed aluminum foam packed with PCM to store heat during peak power operation of variable power dissipating devices has led to a certain breakthrough. Insertion of a matrix of continuously connected aluminum foam into phase change material (water) followed by its investigation shows the solidification heat transfer of the water. Results show inserting metal matrix into water provides a very effective way to enhance the solidification heat transfer. Preparation of shape-stabilized PCMs using bulk porous Al foams impregnated with organic PCMs (paraffin and stearic acid) shows great stability [9]. The thermal-/dynamic-mechanical properties of the shape-stabilized PCMs were studied. The filling fraction of PCMs was approximately more than 80%, the latent value of the paraffin/Al foam and stearic acid/Al foam composite is 72.9 kJ/kg and 66.7 kJ/kg.

5. PCMs EMBEDDED IN COPPER FOAM

Certain researchers made a new type of high efficiency energy storage devices consisted of copper foam and water. The cold charging process of the new type energy storage devices was approved to be faster and more adequate due to the embedding of copper foam. Preparation of a salt hydrate/metal foam composite phase change material by using barium showed much stability. Hydroxide octahydrate ($\text{Ba}(\text{OH})_2 \cdot 8\text{H}_2\text{O}$) as latent heat storage PCM and copper foams as a supporting matrix were also developed. Thermal cycling and heat transfer performance was studied. Results show that high porosity copper foam not only

enhance the heat transfer rate of $\text{Ba}(\text{OH})_2 \cdot 8\text{H}_2\text{O}$ but also effectively reduce the super cooling of the PCM. An investigation of the thermal performance of solid-liquid phase change thermal storage device with 98% pure Heneicosane ($\text{C}_{21}\text{H}_{44}$) filled in copper foam through a vacuuming procedure was being made. Experimental results show that the thermal conductivity and performance of the thermal storage device is obviously improved using copper foam as a heat transfer enhancement. Some researchers also prepared a composite PCMs using paraffin as phase change materials and copper foam as filled materials. The results show that copper foam can not only lead to a more uniform temperature distribution within the thermal energy storage unit, but also extensively shorten the charging time.

6. PCMs EMBEDDED WITH NICKEL FOAM

A study on the application of latent heat storage technology using a composite PCM (a copper or nickel foam saturated by PCM) was being made. The results indicate that composite PCM had increased effective thermal conductivity and could augment temperature change reduction of the heat transfer fluid. In order to improve the void distribution and thermal performance of phase change thermal storage devices, researchers designed and manufactured a thermal storage containers embedded with nickel foam cores. Embedding nickel foam into the PCMs enhanced both the void distribution and thermal performance of solid-liquid phase change process. Preparation of paraffin/nickel foam and paraffin/copper foam composite phase change materials (PCMs) using a vacuum impregnation method was being made. Results show that the thermal conductivity of the composite PCMs were drastically enhanced, e.g., the thermal conductivity of the paraffin/nickel foam composite was nearly three times larger than that of pure paraffin.

7. PROPERTIES OF PCMs EMBEDDED IN METAL FOAM

7. 1. Effective thermal conductivity

Due to the high thermal conductivity and porous structure, embedding metal foam into PCMs can enhance the heat transfer, thus improve the effective thermal conductivity of the composite PCMs. It is very difficult to predict the thermal conductivity of the PCMs embedded with metal foam for the complicated pore structure of the metal foam [10]. It was proposed that a new phase distribution model of metal foam matrix PCMs can be developed. Simplified heat transfer model with void sub model was established and the effective thermal conductivity formula was derived by the equivalent thermal resistance method. An investigation on the thermal parameters (Effective thermal conductivity, thermal diffusivity and thermal capacity) of copper-foam/paraffin with four different porosities using transient plane source (TPS) method was being made. The test results showed that the effective thermal conductivity is obviously improved by embedding the copper foam into paraffin and it reached 25 times compared to pure paraffin.

7. 2. Convection

Metal foam with high thermal conductivity is generally considered to have high potential to enhance the heat transfer performance for PCMs. In an attempt to enhance the convective thermal transport, metal foam can be used for making advanced compact heat exchangers because of the high surface area to volume ratio as well as enhanced flow mixing

due to the tortuosity of the pass ways. The natural convection in liquid region of the PCMs is suppressed by the metal foam. Buoyancy-driven velocities are too weak to produce dominant convection due to high viscosity and low thermal expansion ratio of the PCM and the large flow resistance of metal foam.

8. PHASE CHANGE MATERIALS COMPOSITES

To overcome problem of leakages of liquid PCM, an alternate route has been taken making its composite with high density polyethylene and by absorbing it in porous materials like ceramic granules, tiles and wood fiber board. In composite, PCM is finely distributed in polyethylene/ other porous matrix, which restricts flow of liquid PCM, and therefore can be cut in different shapes easily without any leakages [11]. Composite making is also being tried to solve the problem of poor thermal conductivity of PCM by mixing it with high conductivity materials like graphite. In another approach, a sufficient increase in thermal conductivity has been seen up to 6 W/mK by putting PCM inside metallic foam with per cent porosity.

8. 1. Commercial phase change materials

Many international companies like BASF, Climator, Cristopia, EPS Ltd., Mitsubishi Chemical Corporation, Rubitherm GmbH, TEAP, Witco., etc are marketing PCMs and PCM products. In India also, few companies are marketing their own or licensed PCM products. Currently more than 50 PCMs are commercially available. Most commercial PCMs are based on modified compositions of salt hydrates, paraffins and eutectic salt water solutions, with agents for nucleating, gelling, and thickening added in the base. Apart from micro- or macro-encapsulated PCMs, these companies are also marketing different PCM products like PCM wallboards, PCM-polymer or PCM-silica dry composite powders by Rubitherm. Recently Dupont Energain panels containing a copolymer and paraffin compound has been launched for building material. Garmisch Partenkirchen, Germany, has introduced pocket heaters for mountain-rescue teams. M/S Climator AB is marketing Cool vest to provide 28 °C temperatures around human body for 3 h. An American company Outlast Thermocules, is marketing fibre- and fabriccontaining microencapsulated PCMs. A different application of PCM is in ballistic vests produced by Outlast which protect people from gunshot. Other products for human comfort are underwears to reduce sweating, gloves, shoes, jackets, kidney belts, sleeping bags, etc. Various types of transportation containers to carry medicines and food items to field conditions are being marketed by various companies [12]. PCM products for heat therapy are being marketed by Lavatherm GmbH. A PCM jacket to protect battery from extreme climatic conditions is being marketed by TEAP together with Power Conversion Products and MJM Engineering.

8. 2. Phase change thermal storage for peak load shifting

When electricity supply and demand are out of phase, then one of the possible solutions to this problem can be the development of an energy storage system. Distributed thermal storage materials integrated with a building could shift most of the load coming from the residential air conditioners from peak to off peak time periods. As a result, capital investment in peak power generation equipment could be effectively reduced for power utilities and then

could be reflected in cheap service to customers. The building of integrated thermal storage would enable customers and consumers to take advantages of lower utility rates during off peak hours where power utilities are offering time of day rates. PCM wallboard can thus be used as a load management device for passive solar applications. A 120 m² house could save up to 4GJ a year (or 15% of the annual energy cost). The optimal diurnal heat storage occurs with a melt temperature 1-3 °C above average room temperature. Even certain claims determine that PCM wallboards can save up to 20% of house space conditioning cost.

8. 3. Fire retardiation of PCM-trated construction materials

The flammability requirements and stringent safety codes in the recent years ave been imposed on building materials to protect these from fire hazards. Following are some approaches that have been applied and measured successfully in laboratory tests to fire retard PCM imbibed plasterboard:

1. Addition of non-flammable surface to the plasterboard (e.g., aluminum foil and rigid polyvinyl chloride film)
2. Sequential treatment of plasterboard, first in PCM and then in an insoluble liquid fire retardant (e.g., Fyrol CEF). The insoluble fire retardant displaces some part of the PCM while some remains on the surface thereby imparting self-extinguishing characterization to the plaster board.
3. It is anticipated that brominated hexadecane and octadecene when combined with antimony oxide in plasterboard, the product would self extinguish itself.
4. Certain fire retardant surface coatings can be used to prevent effectively the wicking action of the plasterboard paper covers.

Flammability test evaluation of the burning characteristics of ordinary gypsum wallboard impregnated with approximately 24 % organic PCM shows that flames spread and smoke development classifications were determined in a Steiner tunnel, and heat and smoke release rates were determined by cone calorimeter¹³. When the comparison of the test results with similar data for other building materials are examined, it indicated the possibility of reduction in the flammability of energy storing wallboard by incorporation of a flame retardant.

8. 4. Organic PCMs

Organic compounds have very low thermal conductivity (from 0.1 to 0.7 W/mK), hence requiring certain processes and mechanisms to enhance heat transfer in order on achieve reasonable rates of heat output (W). Table 2 presents the thermo physical properties of the compounds identified for further analysis based on their relatively low price, quoted stability from the review and enthalpy of phase change. Some of the saturated fatty acids, sugar alcohols, carboxylic acids, amides ad alkanes appeared to be promising in this temperature range. Urea is not a promising compound in its pure state because of its instability in its molten state, but some of its eutectic mixtures have suitable properties for latent heat storage with details presented in Table 2.

Table 2. Latent heat storage details of eutectic mixtures

Compound	T_m (°C)	ΔH_m (kJ/kg)	C_{p_s} (kJ/kgK)	C_{p_l} (kJ/kgK)	λ_s (W/mK)	λ_l (W/mK)	ρ_s (kg/m ³)	V_{exp} (m ³ /m ³)	Edensity (kWh/m ³)
Paraffin Wax	0-90	150- 250	3.00	2.00	0.2	0.2	880- 950	12-14	50-70
Acetamide	82	260	2.00	3.00	0.40	0.25	1160	13.9	93
Lauric acid	44	212	2.02	2.15	0.22	0.15	1007	13.6	66
Formic Acid	8	277	1.00	1.17	0.30	0.27	1227	12.0	96
Palmitic acid	61	222	1.69	2.20	0.21	0.17	989	14.1	67
Stearic acid	54	157	1.76	2.27	0.29	0.17	940	9.9	49
Acetic Acid	17	192	1.33	2.04	0.26	0.19	1214	13.5	71
Oxalic acid	105	356	1.62	2.73			1900		211
Erythritol	117	340	2.25	2.61	0.73	0.33	1450	10.3	148
HDPE	130	255	2.60	2.15	0.48	0.44	952		80
Phthalic anhydride	131	160	1.85	2.20			1530		85
Maleic acid	141	385	1.17	2.08			1590		184
Urea	134	250	1.80	2.11	0.80	0.60	1320	16.7	97
d-Mannitol	165	300	1.31	2.36	0.19	0.11	1490		139
Adipic acid	152	275	1.87	2.72			1360	20.2	109
Hydroquinone	172	258	1.59	1.64			1300		105
2-Chlorobenzoic acid	142	164	1.30	1.60			1544		83

8. 5. Heat transfer enhancement method

Certain low thermal conductive PCMs can seriously affect the storage system charge and discharge rates. To deal with this limitation, extended metal surfaces, conductive powders or conductive matrices are proven to be effective in increasing the PCMs heat transport properties, leading to a more uniform temperature within the PCM and better charge and discharge effectiveness for the latent heat storage container/ system.

9. EXTENDED METAL SURFACES

One of the most widely used heat transfer enhancement techniques is to increase the heat transfer area by adding extended metal surfaces known as fins. Different studies have been made on modelling the phase change process with various fin geometries. A compact horizontal tube in tube container using erythritol as the PCM, using axial fins to enhance heat transfer. For this system, melting/solidifying properties that would provide a suitable heat source for driving an absorption cooling system illustrates two common fin geometries widely used in the literature.

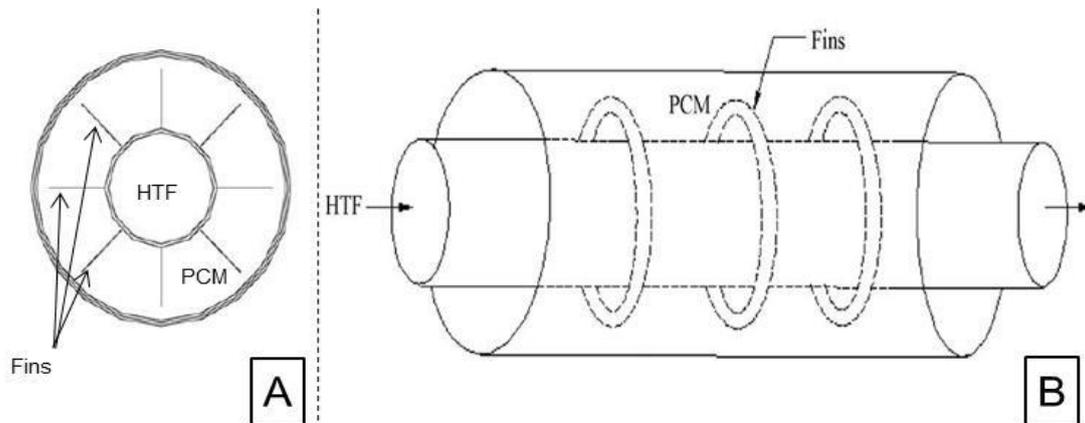


Figure 2. (A) Cross section of a tube in tube heat storage unit with longitudinal fins, an (B) a schematic representation of a tube in tube heat storage unit with annular fins

10. HEAT TRANSFER ENHANCEMENT USING CARBON

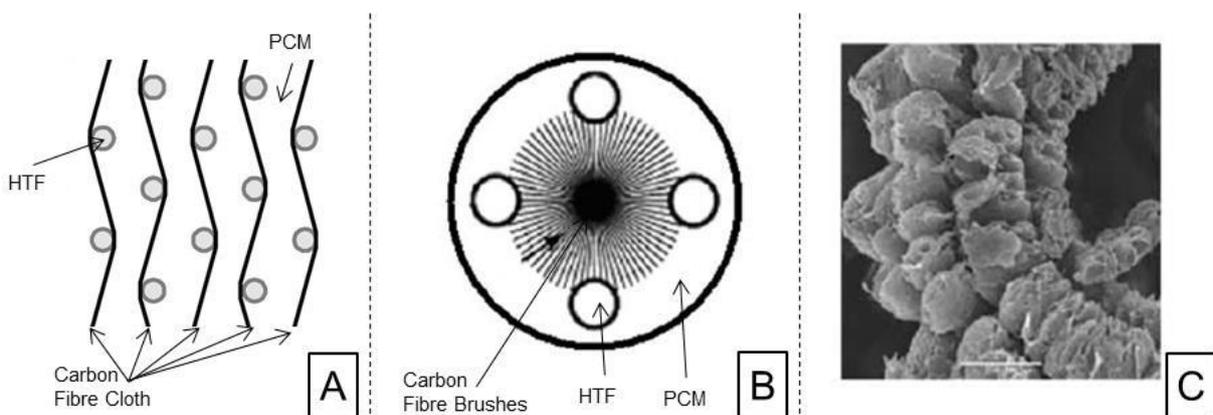


Figure 3. Cross-section diagram of a carbon fibre cloth (A), a carbon fibre brush (B) and (C) viewed with a SEM microscope

Exfoliated graphite, also known as expanded graphite (EG), with a thermal conductivity ranging from 24 to 470 W/m·K, has the potential to increase the global PCM thermal conductivity with low volume ratios (usually around 10-15%). EG is generally obtained from the oxidation of natural graphite with a mixture of nitric and sulphuric acid, followed by drying in an oven and rapid heating in a furnace at 800 to 900 °C to obtain rapid expansion. The PCM is impregnated into the EG under vacuum, this prevents the formation of air gaps within the EG/PCM composite. This technique is the most effective procedure currently used to enhance the PCM thermal conductivity. It also provides a shape-stabilized (SS) form to the PCM since the pore cavities can withstand the thermal expansion typical during phase change and prevent leakage of molten organic material.

The container studied was predicted to double its heat output (from 25 to 50 kWh) if a carbon fibre cloth of 0.8% v/v was incorporated into the system. The system would then provide a nearly constant heat output of 50 kW for around 10 hours and 20 minutes.

11. THERMAL CONDUCTIVITY ENHANCEMENT USING METAL MATRICES

Using sparse metal matrices is another significant way to increase thermal conductivity within a PCM container, structure that would also provide multiple nucleation points. Steel wool is a more feasible method to improve thermal conductivity of a PCM, compared to expandable graphite, but does not provide a shape stabilized solution; since it is not as compactable as graphite. Figure IV presents two approaches used to effectively enhance heat transfer within a PCM.

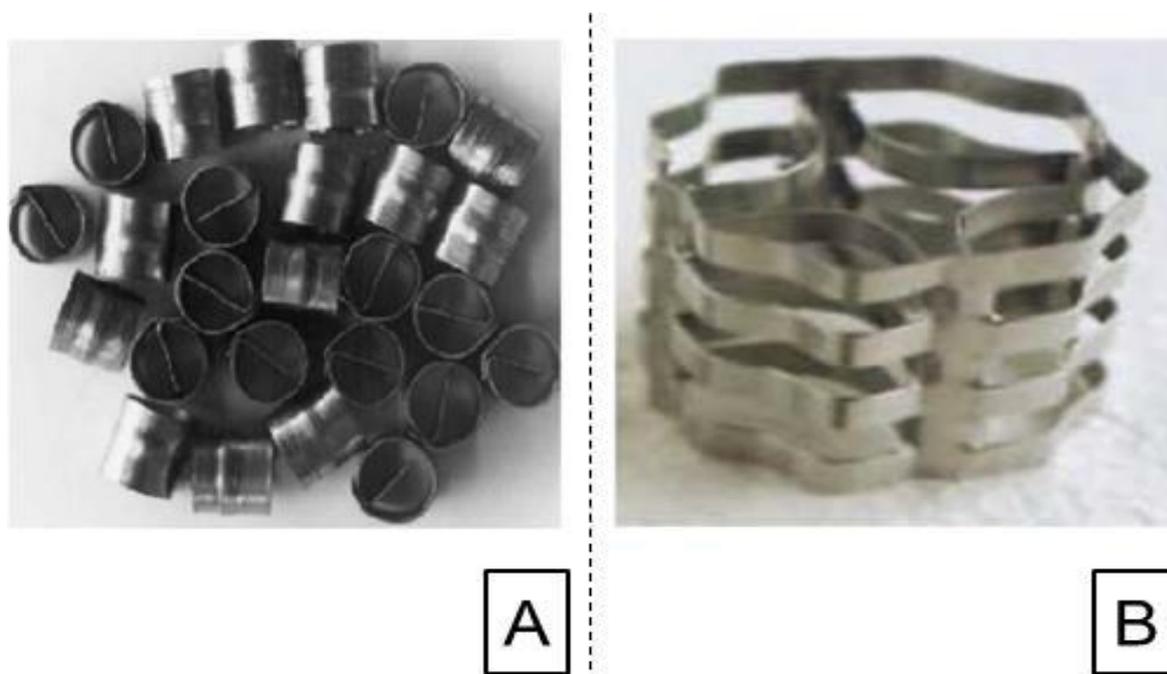


Figure 4. Stainless steel (A) and aluminium lessing rings (B)

12. USING CONDUCTIVE POWDERS

Including small percentages by volume of metallic particles (aluminium, copper, silver, nickel), or graphite can also increase thermal diffusion within low thermal conductivity PCMs. It would also have the added benefit of increasing the number of potential nucleation points, potentially enhancing crystallization within the PCM. However, the conductive material could lose its miscibility when the PCM is in the molten state (due to differences in density), separating from the storage material and sinking to the base of the container. This could be prevented by including gelling agents in the PCM with a consequent reduction in the PCM volume ratio.

13. DIRECT HEAT TRANSFER TECHNIQUES

Another technique to increase the heat transfer would be to provide direct contact between the PCM and the heat transfer fluid. This would provide an effective increase in heat transfer during the melting process since the convective nature of the heat transfer fluid would act directly on the solid PCM phase. The performance of a direct contact latent heat storage container using erythritol and an heat transfer oil concluded that at the beginning of the melting process the oil has a low flow rate due to the block of solid erythritol, the top surface of the PCM melts faster than the bottom due to the higher heat transfer rate and the melting time varies effectively with the oil flow rate.

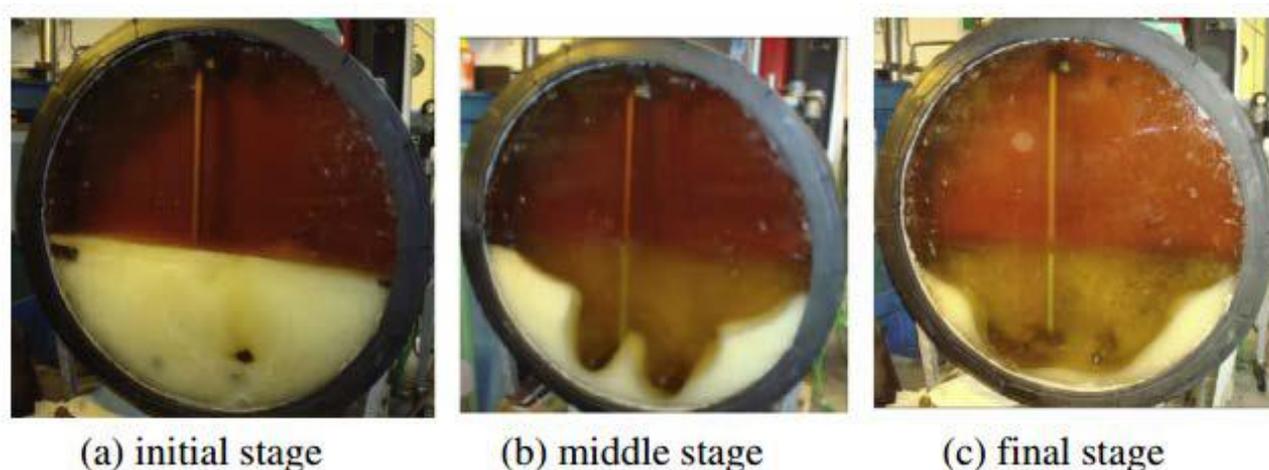


Figure 5. Temporal variation of the melting process in a direct contact heat transfer container using erythritol as the PCM and oil as the heat transfer fluid

To overcome the initial blocking of the fluid flow path when the PCM is in the solid state, the insertion of electric heaters is an effective way [14], Figure VI, and concluded that the overall energy spent on melting the initial flow pathways was 5% of the total thermal energy stored [15].

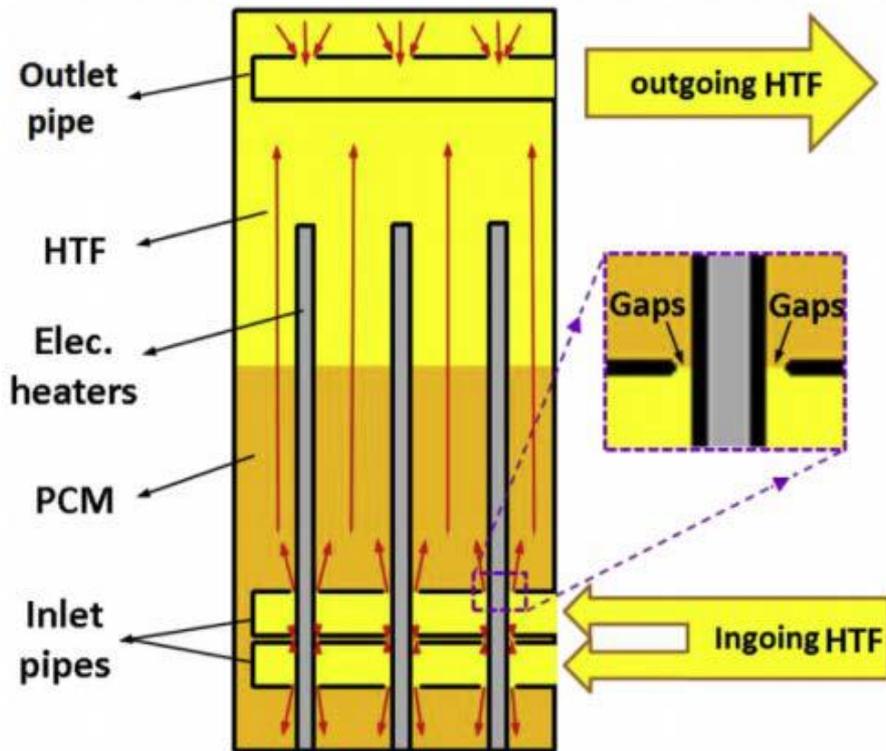


Figure 6. Schematic cross section showing the locations of the electric heaters in the inlet pipes.

14. CONCLUSIONS

Limited experimental studies of PCM wallboard determine few general rules relating to the dynamics of PCM wallboard. The significant conclusion is that a PCM wall is capable of capturing a large proportion of solar radiation incident on the walls or roof of a building. They are also capable of minimizing the effect of large fluctuations in the ambient temperature on the inside temperature of the building because of the high thermal mass of the PCM walls. For shifting the heating and cooling loads to off peak electricity periods, they can be very effective. During new construction and rehabilitation of a building gypsum wallboard impregnated with PCM could be installed in place or ordinary wallboard. Thermal storage will be provided by it that is distributed throughout the building enabling passive solar design and off peak cooling with frame construction. For installation of PCM wallboard in place of ordinary wallboard, little or no additional cost will be incurred.

Phase change materials have the potential to store large amounts of energy within a smaller temperature range when compared to common sensible heat storage materials. Due to the low thermal conductivities of many PCMs, poor rates of thermal diffusion within the PCM can seriously affect the storage system charge and discharge rates that can be achieved.

A comprehensive review of PCMs melting between 0 and 250 °C has been made and the thermo-physical properties of the materials having the most appropriate properties

presented. Below 100 °C, organic compounds and salt hydrates are the most interesting materials. Eutectic mixtures with Urea seem promising around 100 °C, and in the range from 130 °C up to 1250 °C eutectic mixtures of inorganic salts appear the most promising PCMs. A mixture of sodium and potassium formates melting around 170 °C appears attractive due to its relatively low price and moderate latent heat of fusion.

A review of potential indirect latent heat storage containers and systems suitable for integration with various process heating and cooling networks is also reported. Due to its geometrical versatility, encapsulated systems seem more feasible since they can be integrated to any existing system without major technical constraints, although they have lower PCM volume ratios. Compact systems offer larger isothermal stages due to their higher PCM volume ratios; however, heat transfer enhancement among the PCM is imperative to achieve reasonable thermal power output rates, since PCMs thermal conductivity can be a major issue.

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