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editor@ijeast.com



A REVIEW ON ORGANIC PHASE CHANGE MATERIALS AND THEIR APPLICATIONS

Lorina Dash

Researcher, Masters in technology in Surface coating and Technology,
Department of Polymer and Surface Engineering,

Prakash Anna Mahanwar\

Institute of Chemical Technology, Mumbai, India
Professor, Department of Polymer and Surface Engineering,
Institute of Chemical Technology, Mumbai, India

Abstract- Phase Change Materials (PCM) can absorb energy while heating as it undergoes a change in phase and emits the absorbed energy to the environment in a reverse cooling process. Organic PCMs have been extensively used for thermal energy storage in building applications due to its phase transition temperature within the human comfort temperature zone. It has a tendency to leak which limits its application. Encapsulation of PCM in a core-shell pattern have efficiently resolved the issue. This paper is focused on Bio-based PCMs and the enhancement of thermal conductivity, encapsulation techniques of Organic PCMs. It also highlights its versatile application in food, textile, solar energy systems, buildings and paints.

Keywords- Phase Change Materials, Organic Phase Change Materials, Microencapsulation, Bio-based products, Thermal energy storage system

I. INTRODUCTION

Phase Change Materials (PCM) due to its potential applications in Thermal energy storage (TES) have been considered as the main topic of research from the last 30 years. Due to the fast economic growth, limited conventional fossil energy worldwide, and its harmful emission of gases, there is a need to move towards a sustainable and efficient energy system. Though thermal energy is abundant in nature, but what happens when the sun sets. Therefore, TES systems is a huge alternative to maintain the imbalance between energy demands and energy supply.

From the overall energy consumed worldwide, 28% of a large share is consumed by the building sector along with the industrial and domestic building[1]. During summer there is a greater need for energy consumption as the energy demand is very high and supply is less. To cope up with this energy gap, TES systems are fabricated. Three methods of TES: Sensible heat storage systems and latent heat storage systems and thermochemical energy storage systems are used to store and release energy when needed [2].

Therefore, PCM acts as a heat storing material that stores a huge amount of heat and releases it when needed. It has about 3-4 times more capability of storing heat per volume than is stored in sensible heat storing materials in the temperature increment of 20°C[3]. Organic phase change material(OPCM) is a very suitable material for TES due to its unique properties like congruent

melting, narrow melting or freezing temperature range. Hence in this review applications of OPCMs has been taken into consideration for discussion.

There are several review articles such as zalba et al[4], Oró *et al* [5], Raj et al [6] worked in PCM used in cold storage applications. Li *et al* [7] worked on microencapsulated PCM(MPCMs) incorporated in wallboards for yearlong for TES where MPCMs with melting temperatures of 12, 18, 29 °C for winter, mid-seasons and summer days were incorporated in gypsum and new PCM wallboards were fabricated. The incorporation of MPCMs into gypsum was done in two modes. Mode 1: three types of PCMs were mixed and combined in the gypsum matrix to form a single PCM wallboard. Mode 2: three different types of PCMs were combined to form three individual PCM wallboards. Results showed that Mode 2 PCM wallboard showed delayed heat transfer from indoor to outdoor and was suitable for use in building applications.

Jaguemont *et al*[8] reviewed PCMs used as Battery Thermal Management Systems for automotive applications. Sariera and Onder [9] studied OPCMs and their textile applications. PCM has also showed its potential to be used in hot food storage[10][11] and packaging industry[12][13]. J.Sarathkumar Sebastin[14] studied the concept of applying PCM into hot food storage application. He placed hot food on a plate inside a thermal casing to prevent the heat loss by PCM. Paraffin with melting temperature 46-48°C, heat of combustion is 42 kJ/kg was used in the experiment. When the hot food was stored inside the casing with PCM, the PCM absorbed the heat from the food and while storing the energy converted from solid to liquid. When the food temperature drops below the PCMs, PCMs starts solidifying, again releasing the stored heat and hence heating the food. This hot food storage system is cheap and effective and can be used where there is a need to keep the food warm like hotels, home.

There are also some applications where PCMs with low melting temperature have been used to improve the storage conditions of frozen foods in a refrigerator[15][16]. James Johnstan[17] engineered a composite with Nanostructured Calcium Silicate (NCS) matrix and hydrocarbon based Alkane-PCM with 8 °C melting temperature and 160-190 J/g latent heat of enthalpy, for the temporary storage and transport of chilled food items. A paperboard container bubble wrapped with this NCS-PCM composite with 400g of 300wt% PCM loading (3 kg PCM +1 kg NCS) was able to maintain the internal temperature at 10°C for about 5 hours and thermal buffering capacity of 101 J/g to provide temporary storage of chilled perishable food items from the

supplier to the market.

PCMs has also incorporated into paints for TES[18][19][20]. Very few research is done in this field though paints are commonly used to coat the buildings. PCM-paints can also be used to control the increase in temperature of the mobile phones, laptops. n-octadecane as a core PCM has been encapsulated with resorcinol modified urea-melamine-formaldehyde(UMF), incorporated into white primer paint as a LHS system by Tumolva and Sabarillo[21]. They have also studied the effects of homogenization speed and dropping rates of pre-polymer on the microcapsule size, latent heat and encapsulation efficiency of PCMs. Results have showed that increasing the homogenization speed and decreasing the dropping rate of pre-polymer, results into smaller microcapsules resulting in more PCMs per unit meter, implies more latent heat. The values of latent heat of the samples lie in the range 118.2 J/g to 159.6 J/g with a melting range 30.43°C to 33.64°C. They have also emphasized that increasing the MPCM concentration upto 10%, increases the total latent heat of the system. Therefore, PCM-paints can be potentially used as a good LHS system when applied to refrigerators, or anywhere where there is a need to control the temperature of the substrate. Information about the OPCMs has been studied by many researchers and is published in a scattered manner. Also reviews on bio-based PCMs are less covered. Therefore, this review paper includes PCM classification into OPCM and its types, its thermal conductivity enhancement, its encapsulation techniques and its applications in different sectors. Applications of Bio-based PCM have been highlighted here. It has also emphasized the addition of nucleating agents to reduce the supercooling effects in inorganic PCMs. Also, this paper has shortly discussed the supercooling effects of Inorganic PCMs.

II. PHASE CHANGE MATERIAL

2.1 Mechanism of PCMs

PCMs are used to store and release a huge amount of thermal energy. The mechanism of PCM includes when there is a rise in the temperature of the environment and reaches the transition or melting temperature of the PCM, the solid PCM absorbs latent heat at a constant temperature and becomes liquid, when the surrounding temperature of the melted PCM falls, the absorbed latent heat is released and it becomes solid which is explained in

Fig 1. There are three types of the transition of PCM is possible solid-solid, solid-liquid, liquid-solid. But TES consisting of PCM of solid-gas and liquid-gas phase transition are not practical due

to large volumetric changes during the phase transition.

2.2 Properties and Selection criteria of PCMs

The selection criteria of PCMs depend upon their physical, chemical, thermal and kinetic characteristics. Also, their cost, availability, safety, and adaptability becomes an important selection criterion. The end-use application of the PCM acts as the game-changer of selection criteria. PCM has properties like latent heat of fusion, melting/freezing temperature, thermal conductivity, specific heat capacity, and energy density. PCM also shows properties like supercooling, corrosion, low thermal conductivity, hence these characteristics are also to be considered while selecting a PCM for an application [22] along with the phase transition temperature which decides PCM selection.

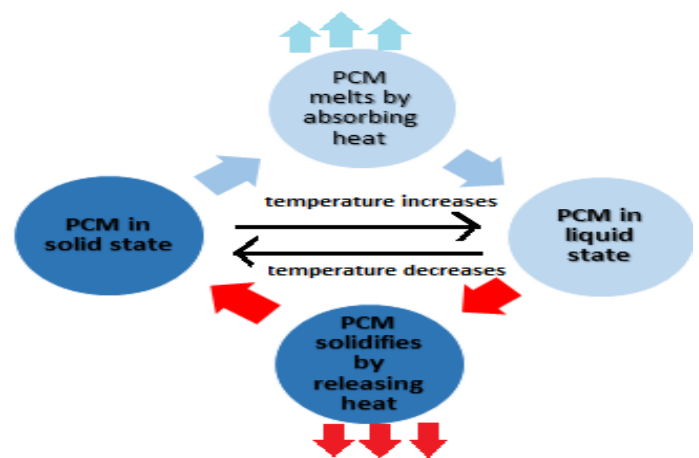


Fig 1: Working of PCM

2.3 Classification of PCMs

TES is classified into physical storage and chemical storage. Chemical storage includes Thermochemical energy storage which stores energy by endothermic/exothermic reaction. Though it is the most efficient method of storing energy, but has not been developed properly and not yet used in building applications. Physical storage is classified into sensible heat and latent heat storage. Sensible heat storage system stores energy by increasing the temperature of the storage media which can be water, soil, brick, brine whereas latent heat storage systems store energy by changing its phase at a constant temperature called melting point as shown in **Fig 2.**

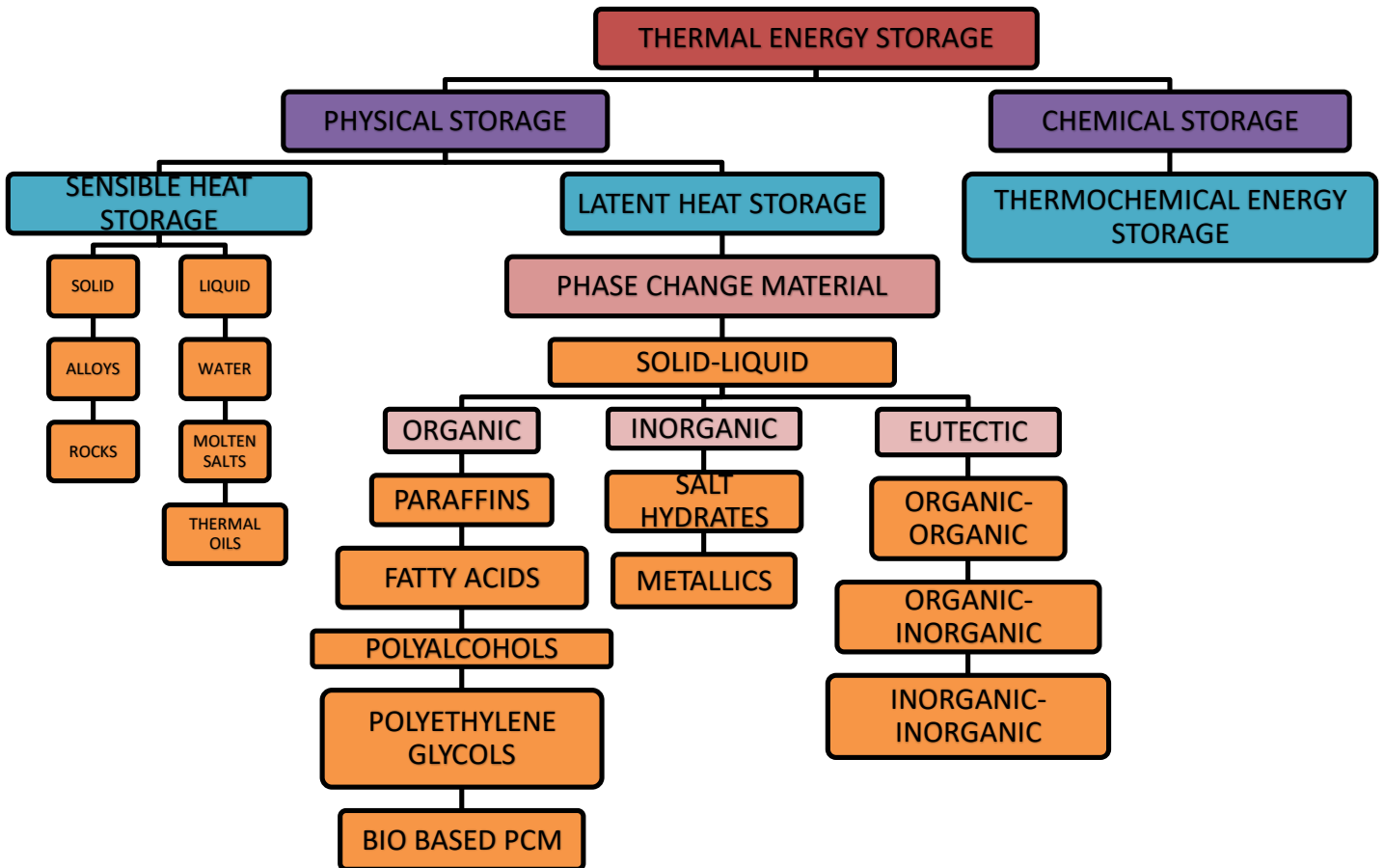


Fig 2: Mechanism of PCM

Latent heat energy is stored in PCM. PCMs can be classified into Organic PCMs, Inorganic PCMs and Eutectic PCMs[23]. These categories can further be divided into subgroups as shown in Fig 3.

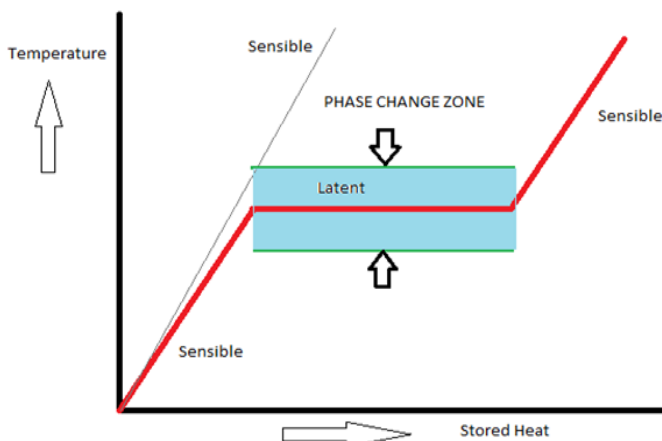


Fig 3: Classification of TES

2.3.1 Inorganic phase change material

Inorganic phase change materials consist of salts, salt hydrates, metal, and metallic alloys. Salt hydrates have been most extensively studied for building purposes[24] because they have

high thermal conductivity ($\sim 0.5 \text{ W/ (m K)}$) and high volumetric storage density ($\sim 350 \text{ MJ/m}^3$) compared to OPCMs. Tatsidjoudoug, Le Pierrès and Luo [25] have discussed potential organic and inorganic PCMs that can be used in heating and cooling purposes for residential applications.

They have a high melting temperature and hence they are used in high-end applications. But inorganic PCMs especially salt hydrates melt incongruently and undergo subcooling. The corrosion of salt on the metal container is also a problem in inorganic PCM[26]. Nucleating agents such as borax and thickening agents such as bentonite and carboxyl methylcellulose are added to reduce the phase separation and supercooling [27]. Liu, Chen and Yu[28] encapsulated sodium phosphate dodecahydrate with Tetraethoxysilane and (3- Aminopropyl triethoxysilane as silica precursors to form the microcapsule shell for TES. The DSP@SiO₂ microcapsules were prepared by water-in-oil inverse emulsion interfacial polymerization with different core/shell ratio. The thermoregulating performance of the microcapsules were tested by adding them to a gypsum board coated with epoxy mixed with 1-3wt% encapsulated microcapsules to the roof of the building which was compared to gypsum board with pure epoxy coating. Real-time temperatures difference of its surface under heating and cooling conditions were measured by J-type thermocouple thermometer which was 13.7°C. This successfully encapsulated microcapsule with 4:1



core/shell mass ratio showed the largest melting enthalpy with the highest encapsulation efficiency of 177.0 J/g and 75.3% respectively and 0.5004 W/mK thermal conductivity.

2.3.2 Organic phase change material

The most familiar OPCM is water which is used for cold storage due to its high latent heat capacity of 334kJ/kg. But a high degree of supercooling limits its applications. Hence nucleation has to be induced to decrease the supercooling effect. Eutectic mixtures of salt hydrates which initiates water crystallization have been used to lower the freezing temperature of water and tends to change phase above 0°C. But its corrosion effect, incongruent melting, sedimentation inside the container causes serious problems in its application. Therefore, a bio-based soya oil ester is chosen as a nucleating agent. An increase in addition of soya oil which contains unsaturated fatty acids, increases nucleation and induces crystallization and decreases the degree of supercooling [29].

OPCM is classified into paraffin and non-paraffins. Paraffinic group includes a family of straight and saturated hydrocarbon chain with similar properties. The melting temperature of these PCMs increases with the increasing carbon chain. Non-paraffinic family includes the largest group of OPCM with fatty acids, esters, alcohols, and polyethylene glycols. OPCM and inorganic PCMs were tested for thermal cycling for thermal energy storage. It was found that OPCM was found to be more stable than inorganic PCM even after 1000 cycles of thermal cycling and showed a gradual change in melting behavior and latent heat of fusion[30]. There are some factors which decide the effective use of OPCM. For instance, high enthalpy of latent heat, suitable phase transition temperature, high thermal conductivity, low volume changes during phase transition, no super cooling effect. Chemically the PCM should possess good chemical stability capable to undergo continuous melting/freezing cycles, no degradation, good compatibility with shell polymers, non-toxic, non-corrosive and without explosives compounds. Generally, OPCMs are initially expensive than inorganic PCMs but the installed cost is competitive.

Demerits of OPCM includes flammability and release of harmful fumes during combustion. Seepage of solid-liquid PCM during melting, large volume changes during transition and low thermal conductivity (0.1510 W m⁻¹ K⁻¹) decreases its heat storing capacity and hence can limit its application. To overcome this, form-stable PCMs where liquid PCM is incorporated into porous polymer matrix, encapsulation, have been further discussed [31].

III. ORGANIC PCM

3.1 Paraffin Wax

Paraffin wax is the most extensively studied PCM due to its cheap, safe, chemically inert, non-corrosive, and stable under 500°C but possess flammability and low thermal conductivity (0.1-0.3 W/mK). They have congruent melting, no phase separation and a wide range of melting points and no supercooling. These are potential materials for energy storage in building applications with a heat capacity of 2.14-2.4 kJ/kg K and latent heat of 200-220 kJ/kg. **Table 1** enlists some common straight chain paraffins with their number of carbon atoms and melting point.

Table 1: Common straight chain paraffins

SL No	Compound	No. of Carbon atoms	Melting Point (°C)
1	n-tridecane	13	-5.5
2	n-tetradecane	14	5.9
3	n-pentadecane	15	10.0
4	n-hexadecane	16	18.2
5	n-heptadecane	17	22.0
6	n-octadecane	18	28.2
7	n-nonadecane	19	32.1
8	n-eicosane	20	36.8
9	n-heneicosane	21	40.5
10	n-docosane	22	44.4
11	n-Tricosane	23	47.6
12	n-Tetracosane	24	50.9
13	n-Pentacosane	25	53.7
14	n-Hexacosane	26	56.4
15	n-Heptacosane	27	59.0
16	n-Octacosane	28	61.4

Paraffin shows excellent stability even after 1500 cycles of thermal stability [32]. Also, it has been reported that paraffin shows slow oxidation when it reacts with oxygen, therefore it needs a leak proof container to avoid leakage. It was found out that Expanded Graphene suppressed leakage of paraffin when encapsulated in polyethylene matrix. Polyethylene was found to be the most compatible polymer for paraffin incorporation due to structural and chemical similarities and does not allow leakage of paraffin due to incompatibility between the two components [33]. But when incorporated in epoxy resin, some amount of paraffin always leaked out because there was no significant interaction between the epoxy resin and paraffin wax and the wax started decomposing at a temperature much lower than the epoxy resin[34]. Paraffins have a low thermal conductivity which makes a reduction in their heat absorbing capacity and hence their heat storage capacity[35].

The thermal conductivity is increased by using following methods[36]:

- Adaptation of extended surfaces or fins
- Multiple PCMs
- Encapsulation of PCM
- Using nano encapsulated PCM

Voelker, Kornadt and Ostry [37] incorporated salt hydrates such as calcium chloride hexahydrate and paraffins in building materials to reduce the temperature of the room. Experimental results showed that the room without PCM heated up to 44°C whereas the room with PCM-gypsum plaster heated up to 40°C. Therefore, it was proved that the PCM-conditioned room can have a temperature reduction of up to 4K.

3.2 Fatty acids

Fatty acids are represented by the chemical formula CH₃(CH₂)_{2n}COOH and are called as non-paraffins. Unlike paraffin wax, these do not show similar properties. The individual category of fatty acids behaves differently and have their own properties. Fatty acids are divided into 6 groups: caprylic, capric,



lauric, myristic, palmitic and stearic[38]. The fatty acids (capric, lauric, palmitic and stearic) have a melting range of 30°C to 65°C approximately and their heat of transition was observed to have a range of 153 to 182 J/g[39]. Yuan *et al* have done a detailed review of fatty acids, eutectic mixtures of fatty acids and fatty acid esters, as well as the preparation and characteristics of fatty acid composites as PCM [40]. Fatty acids have many advantageous properties, including high heat capacity, proper melting temperature range and small volume change. Little or no supercooling during phase transition, non-toxic, low cost, non-flammability, non-corrosive to metal containers, good thermal and chemical stability.

Kant, Shukla and Sharma [41] have prepared 43 ternary mixtures of fatty acids such as capric acid-lauric acid-stearic acid and capric acid-palmitic acid-stearic acid (CA-LA-SA and CA-PA-SA) and found that only 13 mixtures showed single peaks in Differential scanning calorimetry(DSC) curve which showed their use in low temperature TES applications. The rest of the mixtures developed two peaks which could have been due to incongruent melting or phase separation due to which they cannot be used for low temperature TES applications. These mixtures have a melting temperature range between 14- 21°C with an adequate amount of latent heat for TES systems.

Applications of fatty acids have also been studied for solar energy storage by Kahwaji [42] at moderate temperatures ranging from 30-70°C. Decanoic acid (capric acid, $[\text{CH}_3(\text{CH}_2)_8\text{COOH}]$), Dodecanoic acid (lauric acid, $[\text{CH}_3(\text{CH}_2)_{10}\text{COOH}]$), Tetradecanoic acid (myristic acid, $[\text{CH}_3(\text{CH}_2)_{12}\text{COOH}]$), Hexadecanoic acid (palmitic acid, $[\text{CH}_3(\text{CH}_2)_{14}\text{COOH}]$), Octadecanoic acid (stearic acid, $[\text{CH}_3(\text{CH}_2)_{16}\text{COOH}]$) and 1-octadecanol ($[\text{CH}_3(\text{CH}_2)_{17}\text{OH}]$) showed melting temperature range from 30-70 °C with 145 J/g enthalpy change. These organic PCMs were thermally stable upto 3000 cycles.

3.3 Polyalcohols

Polyalcohols have phase change temperatures in the range of -15 to 245 °C, and phase change enthalpies of 100–413 kJ/kg which are also called sugar alcohols [43]. Alcohols such as xylitol, erythritol have much higher latent heat than others in this family. Polyols are non-toxic and safe to use and some are common sugar substitutes like erythritol, xylitol, lactitol. Saman nimali gunasekara [43] discussed the two different melting temperatures of polyols. Erythritol has melting and crystallization temperature in the range of 118.5-120°C and 106-108°C respectively and an average supercooling of 18.5°C. Glass transition temperature of xylitol is 0-113°C probably. Many polyols showed supercooling, volume expansion of 10-15% during melting and decompose under excessive temperature.

Talja and Roos [44] have discussed the phase and state transition effects on dielectric, mechanical and thermal properties of polyols. It is found that they have complex glass transition temperature and melting of the crystalline polyols occurred at their specific melting temperature.

Aran Sole [45] reviewed the thermal stability of sugar alcohols for medium TES applications. Three Polyols, D-mannitol, Galactitol/Dulcitol, Myo-inositol which have high phase change enthalpy and high melting temperatures. The thermal stability was carried in DSC at 10 k min⁻¹ and it was found that D-mannitol and dulcitol showed low thermal stability. Myo-inositol has better thermal stability in the heating process, but in the solidification

process, a decrease of 11% of temperature and 20% in enthalpy values were observed. Ali Memon *et al* [46] developed a form stable composite incorporating lauryl alcohol 24% into kaolin through vacuum impregnation. This composite has a melting temperature in the range of human comfort that is 19.14°C which is less than the melting temperature of pure lauryl alcohol that is 21.69 and considerable latent heat storage capacity which is 48.08 J/g. It was concluded that this composite has a potential application in TES in buildings.

3.4 Polyethyleneglycols

PEG or polyethylene glycols are the family of phase change materials that are extensively studied due to its wide molecular weight, wide thermal transition temperature, high thermal stability and ease of modification. They are non-corrosive and non-toxic. PEG has been incorporated into many PCMs to increase the thermal properties of composite and increase their performance for thermal energy storage. Kou *et al* [47] have studied on heat capacities, thermal stability and phase change properties of PEG sample of molar mass 1000,1500,2000, 4000,6000, 8000,10000,12000,20000. Heat capacities of PEG increased with their increasing molar mass. Thermal decomposition of PEG started from temperature of (523-623) K and ended above 723K. PEG of higher molar masses decomposed at a higher temperature than lower molar mass samples. Hence it was concluded that PEG was thermally stable below 400K. Thermal conductivity of PEG increased with increasing molar masses from 1000 to 6000 then started decreasing continuously upto molar mass of 20000. The highest thermal conductivity was 0.34 W.m⁻¹K⁻¹ for PEG6000. The thermodynamic property such as heat capacity is an important parameter to investigate the thermal properties of PEGs for thermal energy storage.

The main drawback of PEG is some leakage in its liquid phase above the melting temperature. Form stable PCMs can be used to overcome this leakage which remains in the solid state while the working material changes its phase from solid to liquid. EG [48], graphene oxide [49], silica [50], carbon nanotubes [51] have been used as form stable matrix. Many researchers have tried to form solid-solid PCM using PEG. Meng and Hu [52] have tried to synthesize a shape memory polyurethane(PU) as a smart PCM by using PEG 3400 as its soft segment and isophorone diisocyanate and 1,4-butanediol as its hard segment via bulk polymerization. This composite PEGPU has hydrogen bonding in its hard segment underwent a phase of “physical crosslinking” which restricted the molecular mobility of soft segments even after the transition temperature of PEG which was 150°C and made PEGPU still solid. This composite showed an excellent solid-solid phase transition by the transition of PEG segments from amorphous to crystalline states and latent heat storage capacity is 100 J/g.

A novel form stable phase change material(FSPCM) has been synthesized by Chen *et al*[53] by using PEG/the synthesized polymeric solid-solid phase change material(SSPCM). They have used Glucose as the supporting material and molecular skeleton, PEG as a phase change functional chain in the polymeric SSPCM and methylene diisocyanate (MDI) as a crosslinking agent. The form stable characteristics of the prepared composite of Glucose/PEG/MDI have been tested by leakage test which showed that the maximum PEG content in the SSPCM is 70 wt% without any PEG leakage. Phase change properties of the prepared SSPCM has been tested by DSC which showed no



endothermic and exothermic curves for Glucose below 100°C. But the prepared SSPCM showed both endothermic and exothermic curves between 25-80°C. PEG has also been used as a supporting material by Mu *et al* [54]. They synthesized a novel FSPCM of cross-linked poly(acrylonitrile-co-itaconate)/PEG which showed melting temperature of 23-53°C and phase change enthalpy of 118.5 J/g. Slight thermal degradation is observed when the temperature rises above 249°C. This composite has excellent thermal stability, environment friendly and therefore has promising applications in solar energy storage and buildings.

3.5 Bio-based PCM

Due to the constant threat of the exhaustion of petroleum products, researchers have tried to find an alternative as bio based PCM. Also many PCMs are paraffin based products which are highly flammable which urged the researchers to use more of Bio-PCMs which are less flammable and eco-friendly. Mona Nazari [55] did a detailed comparative study between paraffin based PCM and bio-based PCM on TES performance for construction applications. He chose three paraffin based PCMs and one bio-based PCM derived from natural oil with T_m of 24,25,26 °C. He performed Dynamic and Step-wise DSC to compare their latent heat enthalpy and thermal conductivity and found them to be very similar (almost negligible). Bio-PCM has proved to be a promising PCM for thermal energy storage [56][57]. Bio-PCM has also showed its energy saving potential in building applications [58][59].

Leakage in solid-liquid PCM is a problem which needs attention. Different substrates, encapsulation technique or matrix has been chosen to avoid this leakage. Bio-composites were synthesized by Jeon *et al* [60] by vacuum impregnating fatty acid as the bio based phase change material into biochar for thermal energy storage in buildings. A bio-based composite was synthesized by using fatty acid as bio based PCM and biochar produced from rice hulk which was used as leakage proof container. The composite formed consists of oil based PCM with T_m 20-26°C namely coconut oil and palm oil and wax based PCM with T_m 45-50°C namely soy wax and palm wax. Results showed that the biocomposite synthesized by vacuum impregnation method when incorporated in building materials such as cement and gypsum can reduce annual power consumption by 531.31 kWh per year. Trials of bio-PCM composites using wood as a substrate for TES have been tried by Damien Mathis [61]. The decorative wood panels contained pouches of bio-based PCM with an enthalpy of 57.1 J/g and an onset melting temperature of 23°C. Engineered wood flooring with these Bio-PCM impregnated boards can be used to store solar energy and increase buildings energy efficiency. Different wt% of fatty acids as bio based PCM were integrated into concrete as a substrate for TES in buildings by Cellat *et al* [62]. The binary mixtures of CA, MA, LA, as CA-LA and CA-MA has experimented for TES which was found to be in the range of 109.0–155.4 J/g with a differential scanning calorimetry. Better results for compressive strengths were shown when 1wt% PCM was blended into concrete which reduced when 2wt% was blended. DSC results showed that it is mechanically and thermally stable but Thermogravimetric analysis (TGA) showed its degradation starts at 120°C. UV-curing of the PCMs have been used by Emre Basturk [63] to prevent the leakage. The photo crosslinking of the polymer with the PCM prevents the interior PCM from leakage. The problem is minimized by coating the bio-

based PCM with a photo-crosslinked polymer which acts as a matrix. These UV-cured Bio-based PCM has good thermal stability for TES applications.

Lisa Boussaba *et al* [64] elaborated on the use of bio based PCM derived from coconut oil recovered from underused feedstock into incorporated into a composite matrix prepared from cellulose fibers and natural clay via direct immersion method for TES in buildings. Lisa Boussaba *et al*[65] impregnated Hydrogenated Palm Kernel vegetable fat, a bio-PCM with T_m 26.53°C and 74.35 J/g latent heat of fusion. The bio based PCM was incorporated into the matrix which was prepared from natural clay, cellulose fibers and graphite, using direct impregnation method without vacuum treatment. Graphite enhanced thermal conductivity by up to 430%. DSC results showed the composite-PCM has temperature of melting as 27.33°C and 40.27 J/g as heat of fusion. TGA showed that the thermal degradation of the prepared composite-PCM starts above 200°C due to the thermal degradation of fatty acids contained in the bio based PCM. Therefore, this novel PCM-composite has the potential to be used in latent heat TES of buildings. Neil *et al* [66] synthesized a 'Green PCM', alkenones which are extracted from Isochrysis algal is seen as a potential PCM. Alkenones are unique class of algal lipids known as polyunsaturated long-chain alkenones. Alkenones have melting temperatures ~65°C and possess higher latent heat of fusion ~275J/g comparable to synthetic fatty acid PCMs. Results from DSC and thermogravimetric analysis showed its higher thermal stability, hence it is a promising PCM for thermal energy storage.

Reviews have been made to improve thermal conductivity of Bio-PCMs. Jeong *et al* [67] incorporated exfoliated graphite nanoparticles(xGnP) to increase the thermal conductivity bio-based PCM. Bio-based PCM composite was made by vacuum impregnation method. In the thermal conductivity analysis, the thermal conductivity of Bio-based PCM shows 0.154W/mK whereas thermally enhanced Bio-based PCM shows 0.557 W/mK, with an increase of 375%. Graphite has been used by Lisa Boussaba *et al*[65] to increase the thermal conductivity by 430%. They impregnated Hydrogenated Palm Kernel vegetable fat, a bio-PCM with melting point 26.53°C and 74.35 J/g latent heat of fusion. The bio based PCM was incorporated into the matrix which was prepared from natural clay, cellulose fibers and graphite, using direct impregnation method without vacuum treatment. DSC results showed the composite-PCM has temperature of melting as 27.33°C and 40.27 J/g as heat of fusion. TGA showed that the thermal degradation of the prepared composite-PCM starts above 200°C due to the thermal degradation of fatty acids contained in the bio based PCM. Therefore, this novel PCM-composite has the potential to be used in latent heat TES of buildings.

Applications of Bio-based PCM has reached the fields of biomedical thermal insulation due to its bio-compatibility and non-toxicity. Lu Liu[68] prepared a composite with PEG in-situ loaded with calcium ions crosslinked with sodium alginate (SA). 93% of PEG 6000 loaded in the PEG/SA composite provides excellent leakage proof due to the crosslinked structure and shape stability. High latent enthalpy of the composite, 156.8J/g and outstanding thermal stability at 150°C proved its potential applications in waste heat utilization and medical thermal insulation. Hu and Yu [69] encapsulated natural soy wax as Bio-based PCM with PU as the shell via the electrospinning technique. PU



was varied in concentration from 7 wt% to 15 wt%. Nanofibers with PU less than 10 wt % showed beads on strings morphology and PU above 12wt% concentration was too viscous to form steady jets during electrospinning. Optimal morphology without beads and steady jets were formed between 10-12 wt% PU concentrations. DSC results showed that by varying the wax concentration from 10% to 60%, T_m increases, which indicates that the wax increases the thermal stability and crystallinity of PU/wax composites. This novel composite can be used in thermal storage and thermal shielding applications. X. Lu et al [70] synthesized a novel SSPCM with blends of poly(lactic acid)(PLA) as the bio-based material and high density polyethylene(HDPE) as the PCM. PLA chosen as the supporting matrix was melt blended with HDPE in different PLA/HDPE ratios (100/0, 90/10, 80/20, 70/30, 60/40, 50/50, 40/60, 30/70, 20/80, 10/90 and 0/100 w/w). 50PLA/50HDPE blend showed latent heat for melting and freezing process as 100.1 J/g and 97.6 J/g, and the relative enthalpy reached 104.2% which showed thermal stability up to 10 thermal cycles and then the blend showed constant thermal parameters. It also showed no thermal degradation during the phase change temperature 100-140°C. Hence 50PLA/50HDPE blend showed excellent thermal reliability, reusability and shape stability, proved to be potential composite for TES in solar energy plants and waste heat recovery plants.

IV. SUPERCOOLING OF PHASE CHANGE MATERIAL

When a PCM exists in its liquid form below its phase change temperature or freezing temperature or crystallization temperature, without fully freezing is due to supercooling. It is the difference between the temperature at which actual solidification begins and the temperature of the freezing point derived from the equilibrium phase diagram is called supercooling temperature, which is a negative quantity. Supercooling is the difference between the melting temperature(T_m) and freezing temperature(T_f), denoted by the equation (A):

$$T_o = T_m - T_f$$

Salt hydrates experience a high rate of supercooling. They have poor nucleating property hence undergo supercooling before crystallization which creates undesirable practical applications. For instance, if inorganic-PCM is incorporated in building wallboards, the phenomenon of supercooling would delay the crystallization or solidification process of the salt hydrates even after the surrounding temperature of the PCM has reached its freezing point. Hence nucleation has to be triggered to reduce the effect of supercooling [71]. Different mechanisms to trigger nucleation [72]:

- Adding nucleating agents: derivatives of n alkanes are effective nucleating agents to reduce supercooling effects
- Adding nucleating agent or metal additives to the PCM prior to encapsulation.
- Optimizing the composition and structure of the capsule shells [73]
- Using nanofluid PCM to eliminate supercooling
- Additives such as nanoparticles (such as nanoadditives).

Generally organic PCM does not undergo supercooling whereas encapsulated PCM shows supercooling due to reduced space for

nuclei formation [74]. Supercooling mitigation techniques by thickening agents (such as carboxy methyl cellulose), nucleating agents (such as carbon nanotubes, fine salt particles and nano additives) and macroporous structures have been discussed by various researchers [75][76][77].

Experimental results have shown that mean PCM supercooling degree, particle diameter, and cooling time of phase change emulsion affected the freezing of PCM particles. Supercooling characteristics of phase change emulsion have been studied by Morimoto et al [78]. It is reported that the PCM used as a surfactant (such as polyvinyl alcohol) played an important role in triggering the nucleation of PCM. Nanoparticles also enhanced nucleation suppressing supercooling. Use of polyvinyl alcohol as a surfactant is used to suppress supercooling from 12K down to 2K of paraffin wax(n-octadecane) is demonstrated by Georg Hagelstein [79].

Yuan, Ye and Xu [80] fabricated erythritol/expanded graphite(EG) composite via impregnation and sintering method where erythritol was chosen as PCM and EG as a supercooling improver. Experimental results showed that the degree of supercooling (T_{e_0}) of pure erythritol is 75°C whereas T_{e_0} of 15 wt % erythritol/EG composite is 50°C. The extent of supercooling degree decreased by 15 °C. Therefore, erythritol/ EG with a reduced supercooling effect has potential TES application.

Nano encapsulated PCM (NePCM) shows low thermal conductivity and supercooling. Y. Zhu et al [81] found a novel method to eliminate supercooling in NePCM. They encapsulated n-octadecane with SiO₂/graphene composite shell with different compositions of SiO₂/graphene as 0.25%,0.75%,1.5%,2.5%,3.75% and 5% which showed a flower like morphology. With increasing dosage of graphene, the outer surface of capsules became rougher. The average melting temperature and ΔH of the composite was found to be 27.74°C and 107.8 J/g. This NePCM suppressed supercooling and also enhanced thermal conductivity by 132.9% due to graphene content. Wang, Fang and Zhang [82] synthesized a high performance phase change material emulsion with mixed polymeric emulsifier consisting of PEG 600 and polyvinyl alcohol(PVA) with paraffin wax of melting temperature range 62-64°C. Phase change material emulsion contribute little supercooling effect as mixed polymeric emulsifier acts as a nucleating agent. Emulsifier to paraffin ratio was 1:5 whereas PEG600: PVA was 50:50. This showed good stability and reduced supercooling to be used as a thermal energy storage system.

V. THERMAL CONDUCTIVITY ENHANCEMENT

PCM suffers from the defect of low thermal conductivity. Organic PCM generally shows this property whereas inorganic PCM is known for its good thermal conductivity. Thermal conductivity should be enhanced to increase the charging and discharging rates of PCM [83].

Generally thermal conductivities of PCM can be increased by adding additives:

1. Metal based additives such as Cu or Ni or Al foam
2. Carbon based additives such as EG, carbon fiber, graphene, carbon nanotube [84]
3. Encapsulation of PCM

Thermal conductivity is calculated by the following equation (B)



[85][86]:
 $\lambda = q / 4\pi(T_2 - T_1) \ln t_2/t_1$

Many researches have reviewed the enhancement of thermal conductivity of organic PCM [87]. S. Drissi [88] discussed that different techniques to enhance thermal conductivity have resulted in reduced paraffinic content in the Paraffinic-PCM

system, hence lowering the thermal efficiency. Therefore, it is suggested that the paraffinic content should be ~50% or above should be incorporated in Paraffin Phase Change Material for better performances.

Table 3: Thermal Conductivity of PCM materials

<i>PCM</i>	<i>Thermally Conductive materials</i>	<i>'k' of the composite (W m⁻¹ K⁻¹)</i>	<i>References</i>
Tetradecanol (TD)/expanded perlite	Cu powder (CuP) and carbon fiber	CuPTD: 1.9	[89]
Erythritol	aluminum	30	[90]
paraffin	diamond foam	13.88	[91]
Paraffin	carbon black	3.49	[92]
paraffin wax	carbon black nano powder	~135%	[93]
Sodium acetate trihydrate	xanthan gum/copper foam	2.10 ± 0.17	[94]
dodecanol core and melamine- formaldehyde resin shell	graphene oxide and carbon nanotube hybrid filler	~195 %	[95]
paraffin wax	Cotton derived carbon sponge consist of hollow carbon fibers	0.434	[96]
Octanoic acid(OA) and Myristic acid (MA)	EG and Hydroxylated multi-walled carbon nanotubes(MWCNT)	OA-MA/EG:0.99 W/(m·K) OA-MA/MWCNT: 0.36(W/m·K)	[97]
paraffin wax	Manganese dioxide nanowires and nanotubes	377.16%	[98]
n-octadecane	silver coated	1.346	[99]
paraffin wax	Graphene on MgO	51%	[100]
Palmitic acid	mullite/graphite powder	86%	[101]
Paraffin Wax	hexagonal boron nitride	0.85	[102]

VI. ENCAPSULATION TECHNIQUES IN PCMS

Encapsulation is done to increase the surface area of PCMs for the heat transfer and therefore increases the thermal conductivity of the system. It also protects it from environmental stress, increases the compatibility of the PCM with the storage materials, reduces corrosion and also tries to control volume changes during transition[103]. Encapsulation of organic PCM using organic or inorganic polymer matrix ensures no leakage and enhanced thermal conductivity. Many researchers have studied the encapsulation techniques of organic PCM which includes both chemical and physical methods [104][105][106]. A critical study on the use of Nano-PCMs in buildings have also been studied [107][108]. Classification of encapsulation is based on the size of the capsules. Diameters of Macro, micro and nano encapsulation varies from 1mm to greater than 1cm, 1µm to 1mm, and less than 1µm respectively. Macro encapsulation has also been studied along with the criteria for the selection of PCM [109]. Many researchers have also emphasized on the microencapsulation techniques and the thermal energy storage of PCM in building purposes [110][111]. Encapsulation techniques involves the physical and chemical process. Physical methods include centrifugal, spray drying, and fluidized bed processes.

Encapsulation ratio (R%) is calculated using the following equation (C):

$$R\% = \frac{\Delta H_m(MPCM)}{\Delta H_m(PCM)} * 100wt\%$$

where ΔH_m (MPCM) is melting enthalpy in J/g of MPCM; ΔH_m (PCM) is the melting enthalpy in J/g of bulk PCM.

The Encapsulation Efficiency (E%) describes effective encapsulation of core material into the microcapsules which is calculated using the following equation (D):

$$E\% = \frac{\Delta H_m(MPCM) + \Delta H_f(MPCM)}{\Delta H_m(PCM) + \Delta H_f(PCM)} * 100\%$$

where ΔH_f (MPCM) is freezing enthalpy in J/g of Microencapsulated PCM; ΔH_f (PCM) is the freezing enthalpy in J/g of bulk PCM.

Chemical methods include:

- Interfacial polymerization
- Suspension polymerization
- Emulsion polymerization
- Miniemulsion polymerization

Physio-chemical methods include Coacervation and Sol-gel process as shown in **Fig 4**.

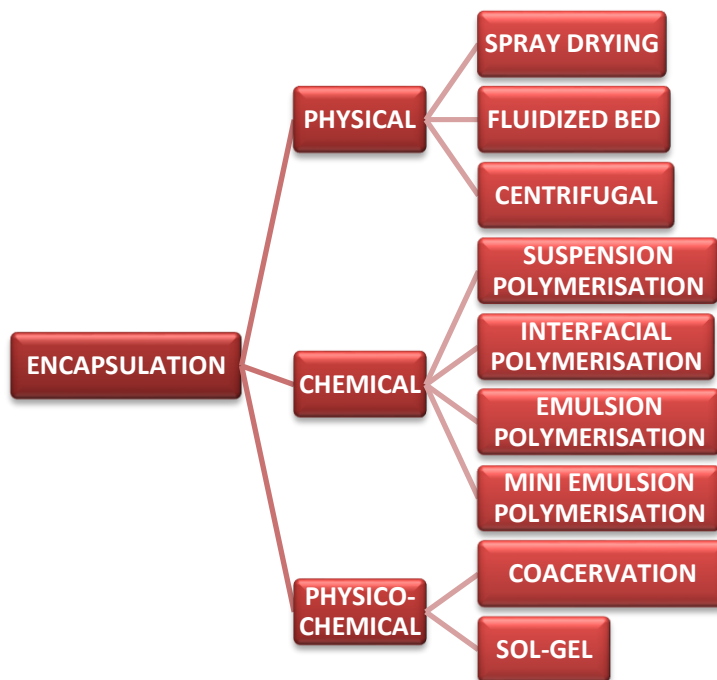


Fig 4: Types of Encapsulation methods of PCM

Table 4: Studies on encapsulation of PCM

Shell	PCM	Technique	Application	Reference
Starch	n-heptadecane	Multi stage adsorption of starch	Textile and coating	[112]
Silicone oil	n-tetradecane and n-hexadecane	Microfluidic emulsification	High temperature	[113]
Polyuria	Paraffin	Interfacial polycondensation	Anti-osmosis	[114]



triethoxyvinylsilane	Methyl methacrylate	mini-suspension polymerization.	Batteries(thermally conductive ,electrically insulative)	[115]
elastic silicone	n-hexadecyl bromide	Microfluidic approach	Thermally stable	[116]
Polyuria	octadecane	In-situ polycondensation	flash memory and data storage	[117]
Polyurethane with pentaerythritol(crosslinked)	butyl stearate	interfacial polymerization	-	[118]
poly(glycidyl methacrylate-ethylene dimethacrylate) beads + silica shell	paraffin and alcohol	cetyl suspension radical polymerization	shape-stable environment	[119]
natural chitosan	poly(octadecyl acrylate)	Coacervation	medical treatment	[120]
calcium carbonate	binary cores	Emulsion	Batteries thermal management	[121]

VII. APPLICATION OF PHASE CHANGE MATERIALS

There is a gap between energy supply and demand and PCMs can be used to fill this gap. PCM can store huge amounts of energy at a constant temperature during the transition phase making them widely applicable in various fields such as buildings, solar power energy, textiles, food, cooling storage, industrial heat recovery, paints. Many research papers have been published on the use of PCM to reduce the cooling loads during the peak hours in different climatic conditions [122][123][124].

7.1 PCMs in textiles

S Mondal[125] have discussed the use of microencapsulated PCMs in smart textiles. He has further discussed various ways of incorporating PCMs in textiles by methods such as fiber technology, coatings and lamination which has been used in sportswear, space, shoes and accessories. The use of PCMs in textiles has been reviewed by many researchers[126].

Y. Lu *et al.*[127] synthesized a novel and smart textile consisting of paraffin wax as the core layer and polyacrylonitrile as the shell and incorporated into textiles via coaxial electrospinning to minimize the leakage problem of paraffin wax. Hexagonal cesium tungsten bronze ($Cs_{0.32}WO_3$), the excellent near infrared region absorbent was incorporated to increase the solar power utilization. This smart textile showed encapsulation efficiency of 54.3% and latent heat of 60.31 J/g with excellent stability over 500 heating and cooling cycles, hence showed its potential application in textiles.

J. Zhang *et al* [128] developed hollow nanofibrous electrospun mats of SiO₂ with paraffin oil phase change material. DSC results showed its melting and crystallization enthalpy as 123.80°C /120.50J/g. This composite showed excellent stability even after 30 cycles of melting and freezing hence serving as a potential thermal energy storage.

7.2 PCM in food

Recently PCMs are also used as refrigeration in food industry [129][16] but there are not many research papers in this area. Nyoman Suamir [130] developed corn-oil ester as a phase change material for frozen food refrigeration applications. Here corn oil acts as a nucleating agent in reducing supercooling of water. This corn-oil mixes with water with a phase transition temperature lower than water from 0°C to -3.5°C to -28°C. DSC results showed that the latent heat of melting and freezing were 297.4 J/g and 102.4 J/g respectively, hence had a great application in refrigeration.

Alzuwaid *et al* [131] incorporated PCM (water gel) with silver iodide as the nucleating agent into open display cabinets. This PCM installation in the cabinet showed up to 5% energy savings and lower cabinet temperatures were achieved.

7.3 PCMs in solar thermal energy storage

The PCMs chosen for solar power plants has high energy storage capacity, high phase transition temperature and high enthalpy. The novel and most recent developments of PCMs in solar thermal energy systems, such as, solar thermal power plants [132],solar water heater [133] and solar cookers have been duly covered. Many researchers have tried to bridge the gap of use of PCMs in solar power plants through simulation and numerical investigation [134] [135][136]. Molten salt is the most efficient PCM to store solar thermal energy. Molten salt is a cheaper PCM with high operating temperature. hence can be used in solar water heaters [137]. PCM can also be used in Photovoltaic(PV) solar systems to minimize the surface temperature and increase its efficiency from 66% to 9.7%. The amount of PCM for cooling the PV panels are optimized. The melting temperature of PCM, ambient temperature, wind velocity and direction would decide the depth of PCM container for cooling the PV panels [138]. Hasan *et al* [139] incorporated PCM of the melting range 38-43°C in the back of



PV panels.

The PV-PCM is found to be consistent most of the year and enhanced the PV annual electrical yield by 5.9% in hot climate. Teamah, Lightstone and Cotton [140] investigated the use of a hybrid PCM/water thermal energy storage in a water tank. water here acted as heat transfer fluid. Simulation and numerical investigation showed that this hybrid system enhanced solar fraction at undersized tank volumes relative to the demand.

Eutectic $\text{Na}_2\text{SO}_4\text{-NaCl}$ salt of melting point 626.0°C and 263.3 J/g as heat of fusion was investigated by Jiang *et al* [141] as a novel high temperature phase change material for solar power plants. Results show that the eutectic mixture of $\text{Na}_2\text{SO}_4\text{-NaCl/}\alpha\text{-alumina}$ as a nucleating agent at 1:1 wt % showed excellent thermal stability up to 100 thermal cycles without cracking.

7.4 PCMs in buildings

The building sector itself consumes one third of global energy consumption leading to the need for energy storing materials such as Phase change materials. The use of PCMs in buildings have been extensively studied [142][143][144].

Navarro *et al* [145] incorporated PCM in concrete matrix. He designed a new precast concrete panel (enhanced using PCM) for radiant walls. He took bulk PCMs, micro-encapsulated PCMs and macro-encapsulated PCMs from the market based on the thermal comfort temperature of buildings between 21°C to 26°C . DSC, Thermal Conductivity and Thermal Cycling stability up to 10000 melting/solidification cycles results showed that the bulk PCM has better thermal properties, higher enthalpy, high specific heat capacity and higher thermal conductivities than micro and macro encapsulated PCMs.

Zhang *et al* [146] synthesized a composite of building materials such as cement with PCM for thermal energy storage. The PEG-1000 was selected as PCM and the expanded perlite, the Granular Activated Carbon, the Powdered Activated Carbon and EG were selected as porous adsorption materials which were mixed with the cement. After analysis results showed that the composite which consisted of PCM and EG was excellent with light weight, high thermal conductivity, high heat storage capacity, high stability and compression resistance and hence can be used as a building mortar material for thermal energy storage.

D.W Hawes [147] incorporated PCM in concrete for thermal energy storage in Canada which shows diurnal temperature swing. He chose butyl stearate, dodecanol and PEG 600 as the PCM materials with a latent heat of 130,140 and 200 J/s respectively. The alkaline nature of concretes makes them unsuitable for incorporation of PCMs especially fatty acids and fatty esters. Hence concretes are autoclaved under high pressure steam to reduce the alkalinity and increase the compatibility of PCM. Experimental results showed that the 3.5 wt% dodecanol was the most compatible PCM with autoclaved concrete with 133% increase in the thermal energy storage.

Esam M. Alawadhi [148] has experimented with the effects of PCM in cylindrical holes in building bricks for thermal analysis to be used in hot climate. Three types of PCM were chosen such as n-octadecane, n-eicosane, and P116 with a melting temperature of 27°C , 37°C , 47°C which is within the operating temperature of PCM-brick system. These PCMs were incorporated into bricks with one, two and three cylindrical holes located in the center of the bricks. Total heat transferred at the indoor space reduced by 11.5%, 17.9%, 24.2% with one, two and three cylindrical holes.

Different design parameters like location of the cylinders, amount of PCM and its type were taken into consideration. Results showed n-eicosane is the best PCM among the chosen PCMs with reduced heat and also the effect increased positively when the quantity of PCM was increased.

7.5 PCM incorporated into paints

PCMs due to its heat storage ability have been incorporated in paints and are further applied to roofs and tiles to reduce room temperature fluctuations. Jeong *et al* [149] incorporated MPCM of melting temperature $21\text{-}31^\circ\text{C}$ and latent heat capacity of 80 kJ/kg into hydrophilic paints. They found out that the MPCMs were more compatible with the hydrophilic paints than the hydrophobic ones. The PCMs with melamine shell were incorporated into an acrylic emulsion resin and a urethane waterproof agent as hydrophilic and hydrophobic paints. The compatible PCM/hydrophilic system was fabricated into heat storage tiles. Tests conducted confirmed that the tiles with more amount of MPCM loading showed lower peak temperature than the tiles without PMCs. Hence this helped in peak temperature reduction of rooms.

A TES-paint with PCM [150] have been developed by Xu Han [150] to study the effects of EPCM on the indoor temperature and energy savings of the building. He encapsulated an organic composite PCM as core with PMMA shell and mixed it with an ordinary paint in 1:1 ratio. This EPCM-paint was applied with 2-3mm thick to two rooms with similar settings and dimensions and was monitored for its energy saving capability. Results from a comparative study of rooms with and without EPCM showed that the EPCM with a diameter of 2-3 μm , 26.2°C melting temperature which is suitable for human comfort and with 139.68J/g of enthalpy showed savings in energy consumption upto 26%. PCM-paint can also be used to decrease the surface temperature of the heated objects. Charles W [151] incorporated PEG of molecular weight 400 and a latent heat more than 30 cal/g into polyvinyl acetate latex paint. This TES paint can be coated in tiles and walls in construction applications. Fitch and Hambrun [152] also used a layer of paint loaded with encapsulated PCMs of different melting temperatures which absorbed the heat generated by the electronic components of the laptop.

Colvin *et al* [153] invented an insulated coating to protect substrates like aircraft skin, bridges, concrete, roadway surfaces from repeated thermal impulses. They dispersed paraffinic MPCMs such as eicosane, heptadecane, nonadecane into binders such as polyvinyl alcohol, polyurethane, acrylic, silicone. MPCM incorporated paint acts as a dual wall, the first is the wall of the microcapsule and the second is of the matrix or polymer itself, hence protecting its leakage from coating during its phase change. Diameter of the microcapsules can range from 0.50 microns to 1000 microns and the coating thickness can vary from 0.010 inches to 0.100 inches. In the same field, to protect spacecrafts from aerothermal heating during launch, Raj K Kaul [154] invented a thermal insulating coating by using PCM. Temperature the most important parameter for the selection of PCMs, shells and binders in paints. So, they chose paraffinic hydrocarbons of ten to thirty carbon atoms containing PCMs, aliphatic alcohols, esters, glycols and inorganic PCMs such as sodium sulphate decahydrate, calcium chloride hexahydrate were used with ceramic or polymeric shells which was incorporated into polyurethane, epoxies or silicone binders due to their desirable



heat stability from 70°F to 150 °F. The percent loading of MPCMs with a transition temperature of 111°F to 175°F was in the range of 46 to 58 percent. The substrate was spray coated and was allowed to heat cure at 130°F.

A flame resistant-PCM has been invented by Hart et al[155] by incorporating flame retardants such as decabromodiphenyl oxide, octabromodiphenyl oxide, antimony oxide into paraffinic hydrocarbons of 13 to 28 carbons atoms, esters and fatty alcohols may be used as PCMs. Microencapsulation can be done by polyurea, polyurethane, urea-formaldehyde and melamine formaldehyde shells. The size of the microcapsules ranged from 2- 10 microns depending upon the applications. These microcapsules can be incorporated into fibers or resins depending upon the end use applications.

VIII. CONCLUSION

This review has enlisted the classification and applications of OPCMs. Bio-PCMs and its applications are also reviewed. Phase Change Materials were studied years ago, but now has gained acceleration due to its inherent property like latent heat storage. Its capacity to store latent energy and reuse it makes it an area of research interest. Both organic and inorganic PCMs due to their different phase transition temperatures are used in different fields of application. But its low thermal conductivity and supercooling has limited its applications to various fields. Therefore, the solutions to these problems have also been discussed in this paper. It can be concluded from the previous research that the OPCM has limited its applications to low and medium temperatures. Paraffins and fatty acids have been researched and used in the areas where very low temperature needs to be maintained. Polyethylenes have been incorporated into various polymer matrix as composites due to its high compatibility and cross linking ability with amines and isocyanates. These PEG based composites increases the application temperature, stability and also prevents leakage of the PCM. Bio-based PCM has recently been the area of research due to its nature-friendly properties like low flammability, renewability and reusability. Bio based PCMs has comparatively low enthalpy of fusion but still it has great future potential to be researched, applied to various fields and commercialized.

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Data Statement:

All the above cited references are accessible and available.

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