



Review A Comprehensive Review of Composite Phase Change Materials (cPCMs) for Thermal Management Applications, Including Manufacturing Processes, Performance, and Applications

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Abstract: To manage the imbalance between energy supply and demand in various energy systems such as energy storage and energy conversion, "phase change materials" are presented as promising options for these applications. To overcome the long-standing disadvantages of PCMs, for instance, small values of thermal conductivity, liquid leakage, separation of phase, and the problem of supercooling, advanced phase change composites (PCCs) manufactured by chemical modifications or the incorporation of functional additives are essential to overcome these disadvantages and promote the large-scale application of PCMs. Herein, we discuss a complete assessment of the recent development and crucial topics concerning PCCs, with a brief description of PCC preparation using different techniques, enhancing PCCs thermal conductivity approaches, and their applications. The various techniques of elaboration of PCCs used can be illustrated as polymerization, encapsulation, and hybrid confinement. Phase change materials (PCCs) are described as potential energy materials for thermal management and storage of thermal energy with the intention of fulfilling the gap between the source of energy and the need in different energy systems. Herein, we give a global presentation of PCCs subjects including PCC elaboration, enhancing thermal conductivity techniques, and applications.

Keywords: phase change composites; thermal conductivity enhancement; energy conversion; energy storage

1. Introduction

Faced with rising fuel sources use, the resultant CO_2 emissions and pollutants, and, moreover, a worldwide energy shortfall, the business community has adopted steps to shift energy consumption, moving in the direction of sustainable energy development [1,2]. In recent decades, there has been great interest in the extraction of renewable energy resources (solar, thermal energy, ... etc.) and their application [3,4]. Nonetheless, using these energies is subject to the natural limits of intermittency, unpredictability and volatility, resulting in a lag in time, space, and intensity between supply and demand [5].

Thermal energy storage technologies (TESs) have been developed and have become an important part of the renewable energy storage infrastructure since the previous century in order to increase the efficiency of use and achieve the required control performance of thermal energy [6]. There are three types of thermal energy systems divided into sensible heat, latent heat, and thermochemical (see Figure 1) [7,8]. For this reason, phase change materials (PCMs) are a possible answer to the energy problem. They have the ability to store and release thermal energy when changing from one phase to another. In addition,



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Copyright: © 2022 by the author. Licensee MDPI, Basel, Switzerland. This article is an open access article distributed under the terms and conditions of the Creative Commons Attribution (CC BY) license (https:// creativecommons.org/licenses/by/ 4.0/). latent heat storage utilizing phase change materials (PCMs) is one of the most effective and sought-after thermal energy storage systems due to its ease of operation and similar energy storage density [9,10].



Figure 1. Thermal storage methods and materials that are commonly used.

"Phase change material" refers to compounds and combinations with varying chemical characteristics [11]. Their common feature is a high latent heat of phase shift and an appropriate transition temperature. Figure 1 depicts the many forms of PCMs that exist. PCMs include four types, solid–liquid, solid–gas, solid–solid, and liquid–gas, according to their phase transition states. In turn, PCMs are classified into three major families according to their challenges and advantages [12]: organic PCMs, inorganic PCMs, and eutectic PCMs [13]. Each family is composed of several subfamilies. Figure 1 shows the different PCM families. To describe the differences between the three major families, Table 1 summarizes the benefits and drawbacks of the different phase change materials.

	Advantages	Disadvantages
Organic	 Accessible in a wide range of temperature range No phase segregation Chemical stability Supercooling is negligible Compatibility with the building material Environmentally friendly, 100% recyclable 	 Low latent heat Low thermal conductivity thermal conductivity High volume expansion Flammable High cost compared to hydrated salts
Inorganic	 High heat of fusion Availability High thermal conductivity high Low volume expansion Low cost Nonflammable 	 Segregation Loss of efficiency due to the cycles of fusion/solidification cycles Supercooling Corrosivity Dehydration due to thermal cycles
Eutectic	 Volumetric storage density slightly higher than organic compounds. They have a net melting point similar to a pure substance 	 Only limited data are available on the thermodynamic properties. The use of these materials is very recent for the application of thermal storage

Table 1. Benefits and drawbacks of the different PCMs.

PCMs are employed in a variety of technologies, including building construction, solar energy storage, electronic component cooling, air conditioning systems, and the textile industry [14,15]. However, their application is limited due to the complexity of the solid–liquid phase change phenomenon, which is related to the dynamics of heat and mass transfers, the spatiotemporal distribution of the evolutions, and the specific behavior of PCMs during the phase change, which introduces an important singularity within the heat transfer model. Understanding and analyzing these transfer and phase change events is thus critical for designing a latent thermal energy storage system. The low heat conductivity of PCMs further limits their use. For example, kerosene waxes have a thermal conductivity of 0.15-0.4 W.m⁻¹.K⁻¹ [16]. In order to improve the thermal conductivity, it is necessary to add conducting fillers to this material to elaborate phase change composite (PCC). One of the ways used to remedy this problem is to impregnate the PCMs in a porous medium of high thermal conductivity. Furthermore, conventional PCMs have been modified with functional additives to enable PCCs functionalization and to broaden their potential applications. Metallic foams (MFs) are a porous medium with a metallic matrix that offer unique properties such as high porosity (porosity ranging between 0.8 and 0.98) and high thermal conductivity [17]. This qualifies them as a good option for enhancing heat transmission during PCM melting and solidification. To enhance the thermal conductivity of the pure phase change materials and hence their performance in different applications, PCMs were encapsulated in shells or inserted with highly conductive fillers such as carbon, metals, and ceramic materials [18,19]. Despite significant progress in the preparation and application of high-performance PCCs, advanced multifunctional PCC use is still in its early stages and requires additional exploration and research.

The goal of this study is to give a complete understanding of the most recent advances in the research and development of composite phase change material (cPCM) including material elaboration techniques and enhancement techniques of the thermal conductivity. Furthermore, special emphasis is placed on advanced applications of cPCMs, such as energy storage, energy conversion, and thermal management (Figure 2).



Figure 2. Form-stable PCCs integrated diagram, encompassing preparation processes, enhancement of thermal conductivity techniques, and their applications.

2. Fabrication Methods of PCCs

2.1. Encapsulation

Depending on the architecture of the material, encapsulated PCMs are classed as core-shell materials (CS-PCMs) or shape-stabilized PCMs (SS-PCMs) (Figure 3) [20]. Core-shell materials are made up of PCM particles (core) covered with another material (shell) that encapsulates the PCMs, whereas SS-PCMs are made up of PCMs and other materials that hold the melted PCMs by capillary action. Encapsulation processes are categorized into four types: in situ polymerization, complex coacervation, sol–gel, and solvent extraction/evaporation [21,22].



Figure 3. Architectures of (**a**) a capsule of core-shell encapsulated PCM and (**b**) shape-stabilized PCMs are depicted.

2.1.1. Core-Shell Encapsulation Methods

The discovered core-shell encapsulating approach for inorganic PCMs is summarized in Figure 4. The core-shell encapsulation methods can be divided into three categories: chemical, physicochemical, and mechanical. In situ polymerization, emulsion (direct emulsion, reverse emulsion, and Pickering Inverse Emulsion (PIE)), electrodeposition,



and interfacial polymerization are among the chemical processes used, as depicted in Figure 4 [22].

Figure 4. Representation of different techniques of PCC fabrication methods.

2.1.2. Shape Stabilized Encapsulation Methods

Chemical Methods

There are many encapsulation methods by using chemical methods. However, the three most often used chemical techniques for the production of microcapsules are interfacial polymerization, emulsion, and in situ.

In the process of "in situ polymerization", all the aspects are dissolved (similar to interfacial polymerization), and the microcapsule wall is created from the emulsion's nonstop segment that used to be at the start surrounding the PCM cores [23]. The PCM is, at the beginning, emulsified in the water before the prepolymer is added to the nonstop section to encapsulate the PCM in a shell by way of a polymerization response [24]. Zhang et al. previously developed PCM microcapsules shell using melamine-formaldehyde (MF) utilizing "cellulose nanocrystal-stabilized Pickering emulsion in situ polymerization (CNC)" [25]. As a result, they obtained the flame-retardant characteristic of the MF shell, where the microcapsules PCM are self-extinguishing. It has been established that the use of PCM microcapsules in prospective applications such as temperature management is feasible. In another work by Kumar [26] on 1-dodecanol as the core PCM and melamine-formaldehyde as the shell material, the obtained material is considered a potential candidate for thermal energy storage at low temperatures.

For emulsion polymerization, it is a type of radical chain polymerization in which one or more monomers are spread in a continuous phase. A traditional recipe typically consists of one (or more) monomer(s) previously dispersed in the form of droplets in a continuous phase (generally water), a water-soluble initiator (or soluble in the monomer(s)), a surfactant or emulsifier, which may be ionic or nonionic and whose role is to stabilize the dispersion of the monomer droplets and polymer particles. The general steps of this process are: "(1) free radical chain length precipitation, (2) free radical penetration into the monomer-swollen emulsifier micelle, and (3) propagation triggered by free radical growth in the medium" [27]. Boya Zhang et al. [28] developed a hybrid shell via Pickering emulsion polymerization-based polystyrene/cellulose nanocrystal (CNC). The obtained PCMs are characterized by outstanding temperature-regulating capabilities and promise for thermal energy storage systems. Dong Xu et al. [29] used poly(methyl methacrylate) (PMMA) and PMMA copolymers as shells and paraffine as the core utilizing the emulsion polymerization technique.

In contrast to the previous method, the interfacial polymerization method happens at the oil–water interface under the action of an initiator when two reactive monomers are dissolved independently in the oil and water phases. The general steps of interfacial polymerization are as follows: (1) the formation of an O/W emulsion containing PCMs and a hydrophobic monomer; (2) the addition of the hydrophilic monomer to initiate the polymerization under appropriate conditions; and (3) the formation of microcapsules via filtering, washing, and drying. Jian Shi et al. [30] elaborated on this idea to create PCM-based paraffin@ polymethyl methacrylate (PCM@PMMA) nanocapsules, which are characterized as promising for energy storage applications. In another study, Xin Yang et al. [31] created a new lauric acid (LA) microcapsule with a silica (SiO_2) shell using interfacial polymerization. The obtained material is found to be a good candidate for energy storage applications. Liu et al. [32] produced a range of biobased, shape-stable composite phase change materials (SSCPCMs) consisting of chemically treated wood (CTW) as the porous supporting material and myristic acid (MA), paraffin (PW), and polyethylene glycol (PEG) as the phase change working component (Figure 5). The obtained SSCPCMs were found to be good candidates for thermal energy storage.



Figure 5. Diagram for the preparation of SSCPCMs [32].

Physico-Mechanical Procedures

Spray drying is the primary technique used to produce microencapsulated particles. Spray drying is a physical method that converts microcapsules from a liquid dispersion to a solid powder composition. It can be divided into two steps: (1) The wall and core elements are contained in an emulsion or suspension. (2) Spray drying using a nozzle atomization method for microencapsulation. Bin Fei et al. [33] developed microcapsules with a titania shell and an n-octadecane (C18) core using a quick aerosol method with a hydrothermal post-treatment. They were discovered to exhibit photocatalytic activity in the elimination of microorganisms and the reaction with mercaptans. They also give UV protection. They should increase the comfort and health of the user's skin when applied to garments through the coating. Carvalho et al. [34], using the spray drying method of green coffee oil microparticles utilizing emulsions stabilized by lecithin and chitosan by an electrostatic layer-by-layer deposition method. They were discovered to exhibit photocatalytic activity in the elimination of microorganisms and the reaction with mercaptans.

Physical-Chemical Methods

Encapsulation based physical-chemical methods includes two techniques, which are coacervation and sol–gel encapsulation [35]. Coacervation usually involves the use of two or more colloids [36]. The coacervation technique is based on the phase separation from the initial solutions and subsequently forms a new coacervate phase; then the material is

coated by coacervate to finally obtain core material surrounded by a shell [37]. Shanshan Tie et al. [38] developed a simple technique based on self-crosslinking between core and wall materials in the coacervation system for effective procyanidin encapsulation (PCs). The coacervates were created by interacting bioactive PCs, gelatin, and sodium alginate, then forming a sodium alginate-calcium ion cationic bridge to increase the PCs' stability. This indicates that PCs have a greater protective effect after being encased in coacervates. Another work by Bayés-García et al. [39] consists of using two coacervates agar-agar/acacia gum and sterilized gelatin/acacia gum as shell compositions in the manufacture of PW-based microcapsules using a sophisticated coacervation technique.

Another encapsulation method-based sol-gel used as a chemical-physical technique is used to prepare capsules at the micro or nano level. It is characterized by low processing costs and moderate processing conditions, which is the reason it should gain more attention from the scientific community. Generally, the sol-gel technique can be divided into three actions: (1) hydrolysis, (2) condensation, and (3) drying [27]. The sol–gel approach with an oil-in-water (O/W) emulsion has been used effectively to encapsulate SiO₂ and other inorganic minerals. For instance, Junping Ren et al. [40] prepared four different samples of phase change composites by sol-gel encapsulation (pure PCM, silica powder-adsorbed PCM, floating bead-adsorbed PCM, and activated carbon-adsorbed PCM). As a result, the activated carbon-adsorbed PCM show higher heat storage capacity compared to other PCMs. Maria Dolores Romero-Sanchez et al. [41] used sodium nitrate, NaNO₃, as a phase change material (PCM) and the microcapsulation process using the sol–gel technique, taking an inorganic compound (SiO_2) as a shell material. The obtained material is convenient as a potential for thermal energy storage applications such as solar plants or recovering of waste heat in industrial processes. In a study by Zhang et al. [42], the sol-gel procedure was used to create a series of C18 microencapsulated with SiO₂ shells at varied pH levels; the spherical microcapsules had a definite heart-shell configuration (Figure 6).



Figure 6. A sol-gel technique is used to produce C18/SiO₂ microcapsules [42].

A summary of some example encapsulated of PCCs is represented in Table 2.

Encapsulation Technique	PCMs Shell		Ref
	C18	TiO ₂	[33]
Conver densine	Green coffee oil	Lecithin/chitosan	[34]
Spray drying	PW	Gelatin/acacia	[43]
	Graphite/silicon	Porous carbon	[44]
	Comb-like poly (octadecyl acrylate)	Natural chitosan	[45]
Coacervation	1-hexadecanol	Crosslinked gum Arabic–gelatin modified with vinyl groups	[46]
	Hexadecane	Soy glycinin	[47]
	Caprylic	Acid PMUF	[48]
	Tris(hydroxymethyl) aminomethane	Silica-organosilica	[49]
	Palmitic acid	TiO ₂	[50]
	Paraffin	TiO ₂	[51]
Sol-gel	Myristic acid/palmitic acid	Organosilica	[52]
Sol ger	Palmitic acid	Silica	[53]
	SA	SiO_2	[54]
	C20	Fe ₃ O ₄ /SiO ₂	[55]
	SA	TiO ₂	[56]
	PA	Alooh	[50]
Emulsion polymerization	Hexadecanol	PS	[57]
Emuision porymerization	PW/butyl stearate	Acrylate-based polymer	[58]
	C18	PMMA	[59]
	C18	St/MMA copolymer	[60]
Miniemulsion	Tetradecane	PS	[61]
polymerization	PW	PS	[62]
porymenzation	Dodecanol	PMMA	[63]
	C18	PMMA	[64]
	C18	PMMA	[65]
Suspension polymerization	C18	PAMA	[66]
Suspension porymenzation	Methyl stearate	PU	[67]
	$Na_2 HPO_4 \bullet 7H_2O$	PMMA	[68]
	PW	Polyurea–Fe ₃ O ₄	[69]
Interfacial polymerization	PW	PMMA-SiO ₂	[70]
	Xylitol	Poly(urethane)	[71]
	PW	SiO _{2/} GO	[72]
In situ polymerization	Dodecanol	HDPE	[73]
in situ polymerization	Tetradecane	UF	[74]
	CA	PMUF	[48]

Table 2. Summary for encapsulated PCCs.

2.2. Hybrid Confinement

Hybrid confinement is one of the primary approaches in which the original molecular structure of the PCM is inserted [75]. Generally, hybrid containment technology restricts solid–liquid PCM in a carrier matrix's inner holes, reducing PCM leakage and thereby boosting dependability throughout the charge/heat discharge process [76]. The common techniques used are the melt impregnation and vacuum impregnation, as are described below.

2.2.1. Melt Impregnation Method

The melt impregnation technique is the simplest and most applicable method for fabricating of porous materials (PCCs) [77]. The process of the melt impregnation technique can be divided into three steps: (1) The porous substrate and PCM are mixed in ethanol solution at different mass percentages and (2) then stirred for 3 to 4 h at a constant temperature. Finally, (3) the mixture is placed in a vacuum oven set to 80 °C for an extended length of time (approximately 12 h) to allow the ethanol to evaporate [78]. This technique was used by Liu et al. [79] to array-encapsulate a carbon nanotube, where the distribution of array-CNT in the PCM is tailored by uniaxial compression (Figure 7a). By introducing n-eicosane (C20) into a porous carbon nanotube network, the thermally conductive material was created. The developed PCCs have up to 90% C20 loading. Li et al. [80] developed a phase change composite-based stearic acid/graphite foam that is testified to have a great potential application in thermal energy storage in tankless solar water heaters (Figure 7b). Zhang et al. [81] described a phase change composite composite of paraffin wax as a PCM and copper foam with a porosity of 95% using the melt impregnation technique (Figure 7c). The obtained material can be an effective candidate to increase the thermal comfort of the building type and lower its energy consumption.



Figure 7. (a) CNT arrays with compressible alignment and PCC [79], (b) [80], and (c) the process of fabrication of copper foam/paraffin PPC [81]. Reproduced with permission.

2.2.2. Vacuum Impregnation

Melt impregnation and vacuum impregnation are two identical processes. As a result, vacuum impregnation necessitates first vacuuming the porous material and then adding it to the impregnated solution. Several studies in the literature have shown the effectiveness of using this method for the preparation of porous material composites [82,83].

Takahiro Nomura et al. [84] developed a phase change composite-based octadecane PCM into mesoporous SiO₂ for building materials; the PCCs were obtained by vacuum impregnation. The ratio of impregnation was above 0.95. Zhiwei Yang et al. [85] developed phase change composite porous carbonized woods (PCWs) by delignification and carbonization treatment of sycamore wood as encapsulation materials of lauric acid (LA) phase change materials (PCMs) (Figure 8a). The resulting PCC demonstrated exceptional thermal dependability, chemical compatibility, and thermal stability. Teppei Oya et al. [86] developed a phase change composite (PCC) using nickel porous metal, and the phase

change material (PCM) was chosen to be erythritol. The PCC was fabricated by vacuum impregnation (the microstructures of the obtained PCCs are shown in Figure 8b). The resulting PCCs have a variety of features that may be used for high heat transfer and effective exploitation of solar heat and industrial waste heat. Ruilong Wen et al. [87] developed PCCs using the vacuum impregnation method. The composite phase change material (PCM) comprising stearic acid (SA) and carbonized sunflower straw (CSS) is shown in Figure 8c. The obtained material is found suitable for solar heat energy storage system and energy conservation buildings.



Figure 8. (a) Preparation of LA/PCWs ss-CPCMs illustration. (b) (1) and (2) are the SEM pictures of porous nickel before vacuum impregnation, and (1') and (2') are the SEM pictures of porous nickel after vacuum impregnation. (c) The experimental procedure for manufacturing SA/CSS composite PCM.

In addition to the described methods above, there are many other methods used for creating porous composite PCM. For instance, the self-diffusion method and vacuum pressure impregnation method are described and used in previous works elsewhere [88,89]. Table 3 represents a summary of previous studies using hybrid confinement methods.

Method of Fabrication	Material	Reference
	1-octsdecanol@HPP	[90]
	SA@MOS	[91]
Melt impregnation	PEG@PC	[92]
	C20@CNTA	[79]
	PW@CNT	[93]
Self-alfrusion method	PW/triglycerides@CNT	[94]
	PEG@MCC/GNP	[95]
	PEG@SWNT	[96]
Vacuum impregnation method	on method	[97]
PW@GO		[98]
Vacuum pressure impregnation	PW@CNT	[88]
purification method	Dodecanoic acid@CNT	[89]

Table 3. Summary of some examples of PCCs by impregnation methods.

2.3. PCMs Polymerization Method

In addition to the previously described methods, PCM polymerization is an efficient process based on molecular bond rebuilding. This process is based on the fact that the crystallizable parts of the PCMs are incorporated into the basic structure of the polymer macromolecules through chemical bonding and inhibit their free movement in the noncrystalline form [99,100]. This method is widely used by previous works, such as polyurethane (PU) [101]. Copolymerization of PU is based on the process of PEG being introduced into a macromolecular backbone ("soft segment") by "block copolymerization" [102–104], "side-chain grafting" [105,106], or "cross-linked copolymerization" [107] (Figure 9).



Figure 9. Polymeric SS-PCM phase transition characteristics [107].

This section provides a quick overview of the three most frequent methods.

2.3.1. Blocked Polymers

Soft polymers have created new potential for the design and implementation of latent thermal energy storage devices. Polymers with complex compositions and structures may be produced using controlled polymerization processes. Among these, the group of block copolymers is particularly interesting. During the block copolymerization process, flexible polymeric and crystalline lamellar sheets are alternatively polymerized to generate linear block structures [104]. Wu et al. [108] developed PCCs by using dual polymer and graphite nanoplatelet networks as the functional matrix of PCMs. Researchers were able to create extremely thermally conductive, flexible, and leakage-proof PCCs (Figure 10a). The PCM in the composites is paraffin wax; the macromolecular olefin block copolymer (OBC) forms a cross-linked polymer network to enclose the molten PCM and provide flexibility to the composite film; and the expanded graphite (EG) with a long-chain structure forms an aligned and interconnected graphite nanoplatelet percolation network to enable the high thermal conductivity of PCCs.

2.3.2. Grafted Polymers

A graft copolymer is a form of the copolymer in which one or more homopolymer blocks are grafted as branches onto the main chain, resulting in a branched copolymer with one or more homopolymer side chains linked to the backbone of the main chain [109]. Reconstructing the grafting architecture has evolved into a flexible approach for creating polymers with stable and fixed forms while also managing their physical characteristics. Poly(ethylene glycol) or poly(ethylene oxide) are linear chains that act as "soft segments" and are grafted to a polymeric backbone that acts as a "hard segment" [110]. When the temperature rises to the phase change temperature of the soft segments, heat storage is initiated by the phase transition component's latent heat absorption process. Simultaneously, the soft segments' movement is limited since they are still connected to the strong backbone, which keeps the PCC firm. Several groups have reported high-molecular-weight grafting copolymers. PVAc-g-PLLA "Poly(vinyl acetate)-graft-poly(l-lactide" graft chains, for example, were prepared by ring-opening polymerization with lactide [111] (Figure 10b).

2.3.3. Cross-Linked Polymers

Cross-linked copolymerization is another appealing method for producing solid–solid PCCs with nonlinear topologies [112]. To create leak-proof 3D copolymer networks, a crystallizable and linear polymer is linked to a backbone as a link between molecular bonds [113]. Because of the homogeneity of the resulting polymer, cross-linking copolymerization is favored throughout grafting and blocking. Jaehyun Wie et al. [88] developed a PCM using polyethylene glycol (PEG) and boron nitride (BN) as a thermally conductive phase change material. Because of the poor interfacial adhesion between BN and PEG, the authors grafted onto the surface of BN polyvinyl alcohol (PVA), as well as cross-linked, due to hydrogen bonding between the hydroxyl groups in PVA and the oxygen atoms in PEG, which increased the thermal conductivity. Wang et al. [114] used a condensation polymerization procedure that produced no small molecule byproducts to create a PCM using PEG and triphenylmethane-tri-isocyanate (TTI). Furthermore, functionalized graphene was equally spread in the PCM, allowing the PCCs to gather light and heat (Figure 10c).



Figure 10. (a) PW-OBC/EG composite synthesis and characteristics. (b) The PVAc-graft-PLLA copolymer synthesis strategy; microscope pictures taken at 130 °C show melted crystallized copolymer with PLLA contents of 90%, 80%, and 70%. (c) Surface-functionalized graphene, PCM, and surface-functionalized graphene/PCM nanocomposites synthesis. Reproduced with permission from [108,111,114].

To reduce the polymeric solid–solid PCCs' latent heat, a new "semi-interpenetrating" system topology was constructed by Zhang et al. [115]. An instance of this technique is used by Zhang et al. [116] by impregnating PW into a crosslinked PU framework using an in situ doping technique. Unlike prior research, the structure of cross-linked PU was created as a support matrix for the PWs. As a result, 210.6 J/g was obtained for the phase change enthalpy at 74 wt% PW. Another study created temperature-controlled latent thermal PCMs with a semi-interpenetrative network structure using a PU structure cross-linked with PEG [115].

A summary of different techniques used for PCM polymerization is represented in Table 4.

Methods	PCC	References
	Polyurethane PCM	[117]
Blocked	Thermoplastic polyurethane PCM2000-8000	[118]
	Hyperbranched polyurethane copolymer	[102]
	Polystyrene-g-PA	[106]
Craftad	Polyacrylonitrile-g-maleic acid polyethylene glycol	[119]
Glatted	Styrene-co-acrylonitrile-g-PA	[120]
	Cellulose-g-PEG2000	[121]
	PEG/4,40-diphenylmethane diisocyanate/Poly(vinyl alcohol)	[122]
Cross-linked	methoxy polyethylene glycol	[123]
	poly(decaglycerol-co-ethyleneglycol)	[124]
	Polyurethane-ionomer-10	[125]
Others	pentaerythritol/butane tetracarboxylic acid/PEG4000	[126]

Table 4. Summary of different PCCs using different techniques of PCM polymerization.

3. Thermal Conductivity Improvement of PCCs

To overcome the problem of low thermal conductivity of PCCs, many solutions have been explored and can be divided into two categories. The first solution consists of adding fins to the heat storage system to intensify the heat transfer during the solid-liquid phase change [127,128]. The second type of remedy is to improve the thermal conductivity of PCCs. This is achieved by adding conductive carbon-based additives such as carbon nanotubes, carbon fibers, and graphite or by adding conductive metal particles (TiO₂, aluminum nitride ...) [127,129]. Improving PCC thermal conductivity can also be accomplished by impregnating PCCs with conductive porous materials [75,127].

3.1. Improvement of the Thermal Conductivity by Graphite Foams

High-porosity graphite foams present a promising solution for intensifying the low thermal conductivities of PCMs for heat storage. They have the advantages of low density; their high thermal conductivities, which reach 200 W.m⁻¹.K⁻¹; their thermal stabilities; their chemical compatibility with PCMs; and their resistance to corrosion [127]. Graphite foams are obtained by the PU foam method, the blowing method, and by compression [130]. They are subsequently soaked in liquid PCM to ensure the impregnation of the PCM into their pores. A graphite foam/PCM composite material combines the heat storage properties of PCMs with the high thermal conductivity of graphite. Many works in the literature have been devoted to the study of this type of composite.

Karthik et al. [131] studied the graphite foam/erythritol composite prepared by impregnation according to the protocol shown in Figure 11a. They highlight the filling of the pores of the graphite foam by the PCM. The results of the thermophysical characterization of the studied composite demonstrated that the thermal conductivity of graphite/erythritol is 3.77 W.m^{-1} . K^{-1} , which is five times higher than that of pure erythritol (0.72 W.m^{-1} . K^{-1}), with an impregnation factor of 75%. Tao et al. [132] used kerosene with a melting temperature of 51.4 °C and a thermal conductivity of 0.22 W.m^{-1} . K^{-1} to fill the pores of graphite foam. They next cut the PCM-impregnated foam into 1–3 mm spheres. These spheres were sprayed with epoxy resin and pressed with 1 MPa pressure to make the composite PCC. According to the findings of this investigation, the introduction of graphite foam increased the thermal conductivity of the composite PCC to 4.98 W.m⁻¹.K⁻¹.

Another work by Karthik et al. [133] focused on the synthesis and assessment of the thermophysical characteristics of a graphite/paraffin composite. The results show that the thermal conductivity of the composite examined in the solid state is 2.6 W.m^{-1} .K⁻¹ and

 1.8 W.m^{-1} .K⁻¹ in the liquid state. The introduction of graphite foam has therefore resulted in an increase in heat conductivity of 980% in the solid state compared to kerosene and 1530% in the liquid form.

3.2. Impregnation of PCMs in Metallic Foams

Metal foams with open porosity are promising possibilities for improving PCMs' poor thermal conductivity. This is due to their high heat conductivity, up to 95% porosity, high permeability, and huge surface areas per unit volume [134,135]. They also benefit from low weight and good mechanical qualities such as stiffness [135]. These foams have a cellular structure formed by pores connected by metallic ligaments. The most-used foams, illustrated in Figure 11b–d, are the foams of copper, aluminum, and nickel [83].

The principle of using metal foams is based on the impregnation of their pores by a PCM to form a composite material. Prepared metal foam/PCM composites are attracting increasing interest in heat storage systems. They cover a wide range of applications that include electronic cooling [136], thermal management of Li-ion batteries [137,138], heat exchangers [139,140], and solar energy storage [141,142].



Figure 11. (**a**) Impregnation technique of erythritol–graphite composite foam preparation. Reproduced with permission from [143]. Metal foams of: (**b**) copper, (**c**) nickel, and (**d**) aluminum [83].

Many other methods are used to improve PCM's thermal conductivity. Because of their excellent thermal conductivity and wide availability, carbon-based materials are attractive adjuncts for the fabrication of enhanced PCCs [144,145]. Carbon-based materials exhibit morphological structures that may be classed as 1D, 2D, and 3D, as shown in Figure 12 [146]. For instance, the 1D nanostructure was used for the first time by Wang et al. [96]; they created PEG-based PCCs with increased thermal conductivities using SWCNTs. The resulting PEG/SWCNT (2 wt%) PCC has a thermal conductivity of 0.312 W/mK, which is

roughly 116.9% of that of pure PEG. For 2D nanostructure, Mehrali et al. [147] realized that, due to their high intrinsic thermal conductivity, a range of PCCs can be created by dipping PA in various commercially available PNGs (300, 500, and 750 m²/g). The obtained thermal conductivity of 91.94 wt% is 2.11 W.m⁻¹.K⁻¹ (eight times higher than the pure PA). For 3D nanostructure, Chen et al. [124] were the first to demonstrate a PCC in which CNTs were utilized as a supporting structure to contain PW and boost the thermal conductivity of the PCC. With 80wt% PW, the thermal conductivity of the PCC reached roughly 1.2 W.m⁻¹. K⁻¹, representing a nearly six-fold improvement over pure PW.



Figure 12. Illustration of the fundamental carbon nanostructures: (**A**) 1D carbon nanostructure (1D CNTs and CFs), (**B**) 2D nanostructure (graphene, graphite nanoplates, EG, and GO), and (**C**) 3D nanostructure (carbon foam, and graphite/graphene foams and aerogels) [146].

4. Applications

4.1. Application of PCM Composite in Thermal Energy Storage

4.1.1. Solar Energy Conversion

Given its purity and availability in many regions of the planet, solar energy is a viable renewable energy source [148]. Because solar energy is discontinuous, the use of thermal energy storage is critical in the solar energy system to extend the running hours of the energy storage system [149,150]. Latent heat storage is a common type of thermal energy storage, and solar latent heat energy storage is gaining popularity due to its compactness, high energy storage density, and stable operating temperature [151]. As a result, PCM, as a potential energy storage material, might be employed to store latent heat energy in solar energy systems such as solar water heating and solar collectors [152].

The solar water heater is the most prevalent piece of solar energy-gathering equipment since it is inexpensive and simple to make and operate. According to the literature [153,154], solar energy is stored inside PCMs as latent heat. The solar water heater operates in the following manner: For the solar water heater, heated water is removed and replaced with cold water that absorbs energy from PCM. The PCM completes the phase shift from liquid to solid as a result of the energy release. Prakash et al. [155] investigated the thermal performance of a novel type of solar water heater with a bottom layer of PCM. The PCM layer is used to obtain hot water during nonsunny hours. The solar collector is a one-of-a-kind energy exchanger that may convert solar irradiation energy to thermal energy in solar thermal applications or straight to electronic energy in photovoltaic applications [156]. In another study by Zheng et al. [157], phase change composites (PEG)-based polyimide (PI)/phosphorene (PR) hybrid aerogel and polyethylene glycol were fabricated using freeze-drying and thermal imidization procedures were used, followed by vacuum impregnation of PEG as a phase transition material into the aerogel framework. This choice of material and procedures presents a viable method for the design and development of high-performance and lightweight composite PCMs to fulfill the requirements of solar energy applications.

4.1.2. Building

Because of their high latent heat, stable chemical characteristics, integration, and compatibility with building materials, PCMs were used for thermal storage in structures built before 1980 [158]. Microencapsulated and macroencapsulated PCMs may be directly integrated into building structures such as wallboards, ceiling boards, and floors [159]. The goal of using PCMs in the structure is to store or release energy under various conditions. Energy storage and release are required in engineering applications to balance availability and demand in terms of time and power.

Figure 13 explains the fundamental notion of temperature regulation using PCMs incorporated into the building structure [160]. As observed, surplus heat from the room may be retained in the PCM to maintain a lower room temperature during the day's high temperatures. During low-temperature periods of the day, such as at night or in the morning, stored heat might be removed by active or passive heat exchange. Furthermore, microencapsulated PCM is commonly used in wallboards, whereas macroencapsulated PCM is mostly used in ceiling boards and flooring.



Figure 13. PCM is included into the construction of the building [160].

Royon et al. [161] conducted experimental and numerical research on the employment of PCM in building constructions (i.e., flooring). A novel polymer PCM composite employing paraffin was created and inserted into the floor panel to enhance the thermal inertia of the building structure. The results showed that using PCM can reduce the amplitude of the surface wall temperature and boost thermal energy storage. Ahmad et al. [162] evaluated the thermal performance of a test-cell with a unique wallboard construction that used PCMs, as well as a comparison with a test-cell that did not use PCMs. They recommended that a vacuum insulation panel be used in conjunction with the PCM panel to increase wallboard efficiency. They observed that this innovative wallboard might enhance the apparent heat capacity of the structure while decreasing the thickness of the wallboard in comparison to convectional wallboards. Zhao et al. [163] developed encapsulating poly (ethylene glycol) (PEG)/organic diatomite (O-Dt) as a latent heat storage agents and wood fiber (WF)/high-density polyethylene (HDPE) as the matrix. The obtained performance of heat storage WPCs demonstrated high heat storage dependability and chemical stability.

4.1.3. Off-Peak Electricity Storage

The difference in power usage between day and night is determined by industrial, commercial, and residential activity. As everyone is aware, different pricing systems may be developed in response to variations in power consumption during peak and off-peak hours. Electricity demand shifts from peak to off-peak hours, which improves economic gain. The development of thermal energy storage systems allows for the storing of surplus

energy until it is released when it is needed. Some approaches have been studied [164]. For example, a common residential storage heater is made of ceramic bricks and structural cement that is heated at night by electrical heating wires or hot fluids. During the day, heat is obtained from the heater by natural convection, radiation, or forced convection.

Some literature [158] has advocated and developed the use of PCM storage technology in off-peak power. PCM might melt and store electrical energy in the form of latent heat thermal energy, as well as release heat as needed. As a result, combining the latent heat thermal energy storage system with the active system can lower the peak load. This technology can also help to minimize the cost of power generating. To take advantage of off-peak power, Agyenim and Hewitt [165] created a finned PCM storage system. To investigate heat transfer properties, they created an experimental setup that included a longitudinally finned phase transition material in a horizontal cylinder. Furthermore, the latent heat method of heat storage will contribute to substantial weight reduction in storage heaters.

Bruno et al. [166] used a PCM system's performance represented by the number of transfer units representation to simulate instantaneous heat transfer when paired to an inverter chiller cooling system. As indicated in Figure 14, the household cooling system included a chiller, a PCM thermal storage unit, and fan coil units. With an ice-based system, 85% of cooling energy use could be transferred to off-peak periods, but energy demand increased by 7.6%. Furthermore, the experiments revealed that utilizing a PCM with a melting point of 4 °C might result in energy savings for cooling. A PCM with a melting point of 10 °C may save around 13.5%.



Figure 14. Domestic cooling system schematic diagram with PCM [166].

4.2. Application of PCM Composite in Thermal Management System 4.2.1. Electronic Devices

Several research have been undertaken to investigate the possible use of PCM in the cooling of high-power devices. Tan and Tso [167] investigated the cooling performance of mobile electronic devices (such as personal digital assistants and wearable computers) in which a heat storage unit containing PCM was employed within the device. Figure 15 depicts a schematic representation of the heat storage unit in which the heaters were utilized in an experimental setting to simulate the electrical packages found within personal digital assistants. They also evaluated the influence of the orientation heat storage unit on the heat dissipation of chips. Hodes et al. [168] conducted an experimental study on the feasibility of transient thermal management of handsets using the PCM in which the handset's base temperature was measured using an infrared camera.



Figure 15. Heat storage unit including PCM [167].

Because of PCM's poor thermal conductivity, the cooling rate of the heat sink utilizing PCMs may be reduced. As a result, certain methods of augmentation have been proposed and developed. Metal foam/PCM composite is a viable solution for improving heat sink cooling performance. For example, Rehman et al. [136] evaluated the thermal behavior of a heat sink employing a metal foam/PCM composite to cool electronic components. They discovered that the heat sink with 95% porosity performed better in terms of cooling than the heat sink with 97% porosity.

4.2.2. Electric Vehicles

In recent years, the battery thermal management system based on PCM has received increasing attention [32,169]. Hallaj et al. [170] presented and investigated a novel battery temperature management system based on PCM for use in electric vehicles. Figure 16 depicts the PCM-based battery module schematic. The results demonstrated that the thermal management system based on PCM was successful in thermally sensitive batteries such as Li-ion and the majority of Li-polymer batteries. The temperature change of the battery cell incorporated into the module design with PCM was noticeably smoother than that of the module design without PCM. Javani et al. [171] quantitatively studied the heat transfer performance of phase change material in electric car battery thermal management. They stated that the highest temperature and temperature excursions in the PCM battery cell were decreased.

Cell capacity = 100 A-h





Zou et al. [172] investigated the effect of graphene and multiwalled carbon nanotubes (MWCNT) as additions on improving the thermal performance of lithium-ion power battery thermal management systems. By comparing experimental data, they discovered that a PCM composite including MWCNT/graphene with a mass ratio of 3/7 may demonstrate the greatest enhancing role. The MWCNT/graphene composite had the greatest temperature increase/decrement rate, which could be reduced by 63.3% and 50.0% when compared to pure PCM. Finally, the PCM composite with graphene and MWCNT as additives had a considerable impact on the thermal management of lithium-ion power batteries. According to the preceding literature, all of the PCMs used in the thermal management system are

paraffin because of their inexpensive cost and constant melting temperature. A novel battery thermal management system (BTMS) was presented that combines composite phase change material (CPCM) with water cooling via double s-shaped micro-channels by Gao et al. [173]. The cPCM is based on paraffin wax-doped expanded graphite (EG) to improve the thermal conductivity.

4.2.3. Cold Energy Storage and Transport

Transporting and storing low temperature-sensitive items is a worldwide concern due to the need to preserve quality and extend shelf life [174]. In recent years, around 4 million refrigerated vehicles have been utilized worldwide, with the use of refrigerated transport continuing to expand. Refrigeration uses around 40% of total energy consumed during product transportation, which encourages associated studies on lowering energy consumption for the refrigerated transport system. Cold energy storage might be an effective approach to bridge the gap between cold chain demand and consumption.

A sophisticated energy technique is cold energy storage employing phase change material (PCM). Many researchers have examined the use of PCM in residential refrigeration, freezers, and refrigeration facilities to save energy and enhance energy efficiency [175,176]. Gin et al. [177] conducted an experiment on the use of PCM in a household freezer. They evaluated the impacts of the freezer's door opening, defrost cycle, and power outage with and without PCMs. Cheralathan et al. [178] used encapsulated PCM to conduct an experimental assessment of the cooling performance of an industrial refrigeration system. The effects of intake temperature on the cooling system were investigated in the experiment. They observed that a suitable evaporator temperature and charging system at a lower condensation temperature might improve cold storage system thermal behavior.

Tan et al. [179] conducted experimental research of the cold storage system using water as the PCM for cold energy recovery of LNG (liquefied natural gas)-refrigerated vehicles, as shown in Figure 17. Michel et al. [180] investigated the insulating walls of a refrigerated vehicle utilizing PU-PCM composite in an experimental and numerical study. It was decided that the composite material should be positioned close to the vehicle's external walls to improve cooling performance.



Figure 17. PCM refrigeration schematic illustration [179].

5. Conclusions

A possible alternative for thermal energy storage is phase change material (PCM). However, its limited heat conductivity limits its widespread applicability. Many approaches have been investigated to improve the thermal performance of PCMs, including encapsulated PCMs, the addition of a high thermal conductivity additive, the insertion of a metal fin, and the embedding of metal foam.

This review provides an overview of PCM composite research, covering manufacturing processes, thermal performance enhancement, and use in thermal energy storage and thermal management systems. It has been discovered that metal foam, as a thermal

enhancer, plays an essential role in improving the thermal performance of PCM. Many scholars have already examined metal foam with a stochastic structure in the literature.

The primary goal of PCCs is to create thermal energy storage or thermal management systems that are safe, highly efficient, and cost-effective. Nonetheless, despite recent significant advances in the preparation, thermal conductivity, and use of PCCs, some crucial concerns demand more investigation:

- (i) Novel low-cost strategies for manufacturing PCCs with excellent thermal stability and energy storage density are required. The optimal design of materials can increase the density of energy storage. Previous studies show that the mix of PCMs and additives has a significant impact on PCC thermal performance. The integration of additive surface modification and novel synthesis techniques is critical for obtaining high energy storage density and homogeneity.
- (ii) PCCs with excellent thermal conductivity are required for PCC devices with high energy density. Whereas a microchannel tube heat exchanger loaded with PCCs may produce a high-power density during thermal charging and discharging, the complexity and high cost of the heat-exchanger construction precludes future use. Three-dimensional porous materials have a greater potential for increasing the form stability and thermal conductivity of PCCs than one-dimensional or two-dimensional additions. It is desirable to use 3D porous materials, which have a greater potential to increase the form stability and thermal conductivity of PCCs than 1D or 2D additions. The creation of massive heat transfer networks aligned within PCCs, in particular, is a viable technique to increase PCCs' thermal conductivity while retaining heat storage density.
- (iii) It is necessary to build PCCs with multiple applications for energy harvesting and storage. Despite very effective energy conversion and storage, large-scale deployment of PCCs at high temperatures still difficult. New designs to prevent heat loss and increase energy collecting should be investigated to further improve energy collection efficiency.
- (iv) The current emphasis of research is on high-temperature PCM cooling and heat absorption. With the spread of new energy vehicles in colder areas, it is vital to further investigate heat storage by cPCMs at low temperatures.
- (v) Future study on cPCMs' ecological preparation and cost-effective technology is required to encourage its use in the fields of energy efficiency and environmental preservation.

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