Solid-liquid phase change materials for the battery thermal management systems in electric vehicles and hybrid electric vehicles – A systematic review

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Abstract

With the rapid pace of advancement in the electrification of vehicles in the automotive industry, temperature changes in Lithium-ion batteries as the power source of electric vehicles and hybrid electric vehicles are the primary concern that affects the reliability, safety, efficiency, and lifespan of batteries. Therefore, an efficient battery thermal management system (BTMS) is essential to alleviate the impacts of temperature change by maintaining the temperature in a reasonable range. These days, BTMSs benefit from the features of phase-change materials (PCMs) to control the temperature of batteries in passive or semi-passive systems. This paper provides an extensive review of various types of PCMs, essential factors for selecting an appropriate PCM in BTMSs, the merits and demerits of solid-liquid organic PCMs in BTMSs, several recently used solutions to tackle the possible problems of solid-liquid PCMs, and finally further research studies.

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Keywords: Solid-Liquid, Phase-Change Material, Electric Vehicle, Hybrid Electric Vehicle, Battery Thermal Management System, Cooling System

Abbreviations

BTMS	Battery Thermal Management System
CPCM	Composite Phase-Change Material
EV	Electric Vehicle
EG	Expanded Graphite
HEV	Hybrid Electric Vehicle
Li-ion	Lithium-ion
L-TEI	Linear Thermal Expansion Increment
MWCNT	Multi-Walled Carbon Nanotube
PCM	Phase-Change Material
SMMT	Society of Motor Manufacturers and Traders
SBS	Styrene Butadiene Styrene
TES	Thermal Energy Storage

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1 Introduction

The environmental issues due to the rapidly growing demand for energy derived from limited fossil fuels have attracted the attention of many researchers to develop technically feasible and environmentally friendly technologies. To attain this goal, Electric Vehicles (EVs) and Hybrid Electric Vehicles (HEVs) have achieved widespread popularity among researchers, scientists, and industries as viable alternatives for conventional internal combustion engines. Therefore, EVs provide green means of transport, equipped with battery packs as the primary energy source [1]. While transportation accounted for 28% of the carbon footprint in the UK in 2017, the UK Department for Transport aims to sell almost zero-emission vehicles, covering 50% to 70% of cars by 2030 and 40% of vans by 2040 [2]. The sales figures reported by the Society of Motor Manufacturers and Traders (SMMT) to date for 2021 show that EVs have a 6.9% market share compared to the 2.9% market share in 2020 [3].

Despite the increasing popularity of EVs, automotive manufacturers must overcome several difficulties. The most crucial obstacle for EVs is to develop a power supply with a longer lifespan that can provide quick charging and high-speed driving [4]. These days, rechargeable Lithium-ion (Li-ion) batteries are extensively utilized in EVs due to their advantages over other batteries, such as high specific energy, a large number of charge-discharge cycles, low self-discharging rate, fast charging capability, greater recyclability, and reasonable cost. Safety is the critical hurdle to the large-scale development of Li-ion batteries in EVs. The recharge of a typical battery in EVs will take up at least 30 minutes to reach 80% capacity. Fast charging increases the battery temperature due to internal resistance, known as ohmic heating or joule heating [5-8]. In addition, these batteries generate much heat during rapid acceleration or long-discharge cycles [9]. The temperature rise in Li-ion batteries may lead to capacity fade, self-discharge, thermal runaway, fire or even explosion [5]. Thermal runaway is the uncontrollable self-heating process related to Li-ion batteries, which is one of the main risks usually caused by mechanical, electrical or thermal abuse [10], the materials used, or cell design [11]. Furthermore, the uneven temperature distribution in a battery pack dramatically decreases its cycle life [12].

The temperature changes in Li-ion batteries are almost always inevitable because these batteries are influenced by environmental factors and heat emission due to chemical reactions such as electrochemical reduction and redox reactions during charging and discharging [13]. Since the performance, lifespan, and safety of Li-ion batteries depend heavily on the temperature, it is vital to have an efficient battery thermal management system (BTMS) for almost all cell materials to prevent adverse effects of temperature changes [12]. The most crucial feature of the BTMS is that it maintains an optimum average operating temperature and temperature uniformity within the battery pack, especially at high charge and discharge rates and high ambient temperatures [14]. The optimal working temperature range for the Li-ion batteries is between 25°C and 40°C with a temperature difference of less than 5°C in the battery pack [15].

Various systems are used in BTMS to maintain the appropriate temperature range, including air cooling systems [16], liquid cooling systems [17], boiling cooling systems [18], and phase change materials (PCMs) [19]. These systems are categorized based on different factors such as the power consumption or the medium used. Power consumption is a criterion to divide these systems into passive and active systems. While passive systems only use natural air convection, PCMs, heat pipes, or fins, active systems utilize an external energy source, such as forced air, liquid, or thermo-electric-based systems [20]. In 2000, Honda Insight and Toyota Prius applied an active system with forced air convection for cooling or heating of HEVs [21]. In 2012, the absence of an optimised BTMS in Nissan LEAF EVs, which relied on an active air cooling system using a single fan, caused battery capacity loss at a fast rate and a cut in half in the driving range in about 50-60°C ambient temperature [22].

Traditional cooling or heating systems such as air and liquid-based systems have some drawbacks. These include the design complexity, high cost, additional space, and power-wasting components such as fans and pumps [23]. Energy and volume are considered two of the most critical obstacles for BTMSs [22]. Hybrid cooling systems, which integrate two or more battery cooling systems, are currently of great interest among researchers to reduce the possibility of thermal runaway because of the high demand for more energy density and shorter recharge time in batteries [24-27]. Ling, Cao [28] investigated a hybrid system integrating liquid cooling and PCM/expanded graphite (EG). An optimized design was proposed, considering the system’s weight, volume, and cooling capacity, saving the PCM mass and volume by 94.1% and 55.6%, respectively.

While many reviews on the PCMs have been published [29-32], this paper provides a comprehensive review on the advantages and disadvantages of using solid-liquid PCMs for BTMSs in EVs and HEVs. An updated review of capabilities and challenges of using these passive cooling or heating systems for battery applications along with available approaches to tackle possible issues are discussed in this paper. First, a fundamental clarification and classification of thermal energy storage systems and PCMs were discussed. The following section presents solid-liquid PCMs with important criteria to be considered for their application in BTMSs. Then, the properties and characteristics of different types of solid-liquid PCMs with their advantages and drawbacks in automotive applications and the solutions to deal with their challenges were provided, followed by a conclusion on this field of research.

2 Thermal Energy Storage Systems and PCMs

Thermal energy storage (TES) systems are classified as sensible heat, chemical energy, and latent heat (shown in Figure 1) [33]. Sensible heat stores thermal energy by using the specific heat capacity. Most materials have a specific heat capacity two times lower than their latent heat, making a larger storage space necessary when using sensible heat. Therefore, sensible heat storage systems have become outdated in TES [22, 34]. In another category of

TES, the energy is stored through chemical processes. Energy storage through chemical processes can store large amounts of energy for long periods. However, the system is quite bulky and complicated because it has reaction chambers, causing limited use of chemical energy storage in practical applications [35]. The third classification of TES is PCMs, which save heat energy in sensible and latent heat forms, mostly in latent form, because of their high latent heat storage capacity [36]. PCMs utilise the latent heat energy to change from one phase to the other phase at an almost fixed temperature. Therefore, PCMs have an excellent capability for heating and cooling in BTMS [37, 38]. The main characteristics of these three categories are also shown in Table 1, showing PCMs with high efficiency and moderate costs compared to the other two categories.

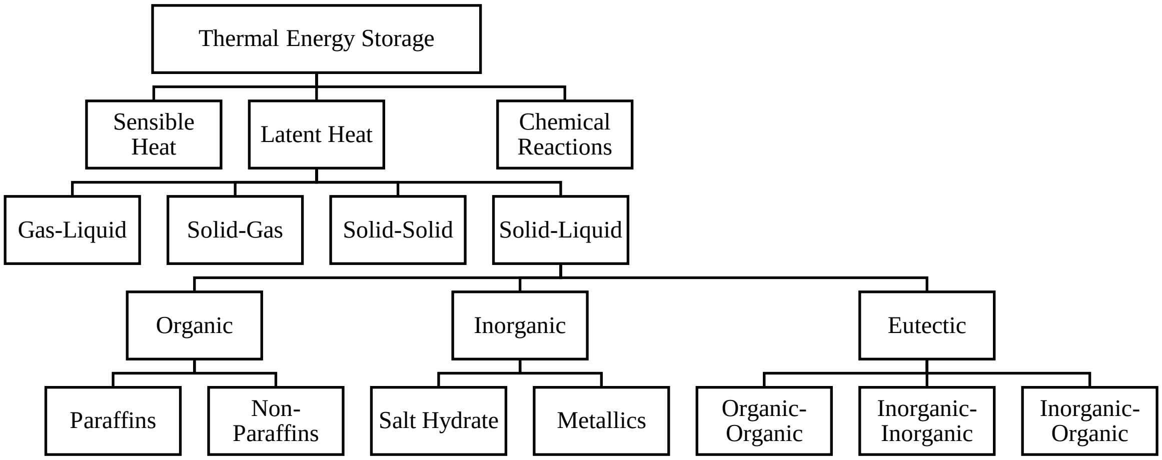

Figure 1. Various types of TES and solid-liquid PCMs [22]

Table 1.
Characteristics of three different TES systems [39]

TES System	Capacity (kWh/t)	Power (kW)	Efficiency (%)	Storage Period (h, d, m)	Cost (€/kWh)
Sensile Heat	10 – 50	1 – 10000	50 – 90	d/m	0.1 – 10
Latent Heat	50 – 150	1 – 1000	75 – 90	h/m	10 – 50
Thermo-chemical Heat	120 – 250	10 – 1000	75 – 100	h/d	8 – 100

PCMs are widely used for either thermal protection or energy storage. While thermal protection aims to reduce or transfer systems' heating or cooling loads, energy storage is designed to store the thermal energy from systems. PCMs are used in many industrial and engineering applications, including electronic industry, buildings, the automotive industry, solar energy systems, textiles, food industry, and medicine. Recently, PCM applications for thermal management of Li-ion batteries in EVs and HEVs are growing to safeguard and thermally stabilize the battery cells throughout charge and discharge cycles. [40].

PCMs can be classified as gas-liquid, solid-gas, solid-solid, and solid-liquid PCMs. While the latent energy in gas-liquid transition is generally one order greater than that in the solid-liquid transition, the gas-liquid phase change is less practical due to the higher volume expansion, which is usually used for heat extraction in heat pipes [23, 41, 42]. The solid-gas phase transition requires substantial latent heat energy, which sometimes makes it impractical in BTMS. Solid-solid PCMs are also not used much in reality due to the high phase change temperature. Solid-liquid PCMs are the most commonly used type, especially in automotive applications, due to large latent heat capacity and small volume changes throughout the phase transition process [22, 43]. When the temperature increases, solid-liquid PCMs absorb heat energy in an endothermic process and varies from a solid phase to a liquid phase. The heat transfer phenomenon during material phase transition is a highly complex process due to different variables such as the moving interface between the solid and liquid phases, the differences in the density and conductivity of the phases, and generated motions in the liquid phase [38]. Solid-liquid PCMs are classified into three main categories: organic (paraffins and non-paraffins), inorganic (salt hydrates and metallics), and their eutectic mixtures (organic-organic, inorganic-inorganic, and inorganic-organic mixtures). The organic and salt hydrate PCMs are suitable for applications below 100°C, such as thermal management systems of Li-ion batteries, while eutectic PCMs can be used at temperatures up to 250°C [44, 45].

3 PCM Selection for BTMS

In recent years, PCM as a novel cooling system is extensively used due to its high energy storage density, high efficiency, compactness, and lightweight. They are also less bulky, less complicated, and cheaper than air cooling or liquid cooling systems [22]. PCMs can be utilized as additional support in BTMSs to hinder thermal runaway or other thermal issues. PCMs, based on their characteristics, can restore thermal energy for an extended period, which is advantageous for EVs [46]. Al Hallaj and Selman [47] developed the PCM application in BTMS for the first time due to its simplicity. PCMs have many other advantages such as simple structure, sustainable life span, high latent heat, low cost, temperature control, property preservation in consecutive cycles, and no need for any external energy consumption such as pumps, leading to extensive research on PCM-based BTMS. However, they have some negative aspects such as low thermal conductivity, volume change, leakage, and PCM collapse at high temperatures [30, 43, 48, 49]. It is also important to be aware that greater PCM thickness in BTMSs can increase heat accumulation within the PCM, leading to adverse effects because of poor heat conduction [49].

Factors that should be considered when choosing an appropriate PCM in BTMS include high thermal conductivity, high latent heat, high specific heat, low volume changes during phase transition, melting temperature, easy availability, reasonable cost, chemical stability, high recyclability, low environmental impact, non-poisonousness, non-flammability, non-explosiveness, and little or no subcooling and supercooling. The phase transition temperature of the PCM should correspond with the working temperature of the controlling system in the selection of an appropriate PCM for a specific application. The phase change temperature of the PCM should be lower than the acceptable maximum temperature of Li-ion batteries to control the adverse effects of temperature increase during charge and discharge [22, 23, 38, 50]. While the ideal PCM should have a melting point between 30°C and 60°C [47], the 30°C melting point is excellent for Li-ion battery applications since batteries degrade rapidly above this temperature [51, 52]. In terms of stability, PCMs must show stable and predictable behaviour over repetitive melting and solidifying cycles to have practical application in thermal management systems. Some techniques have been proposed to improve the thermal, mechanical, and chemical stability of PCMs, including metal-matrix components such as PCMs enhanced by EG and copper foam [53] or PCMs improved by aluminum nitride (AlN) [54].

Some PCMs such as organic PCMs are flammable and emit harmful gasses resulting from combustion [55]. Several techniques can control this issue, including using layers of non-flammable coating, fire-resistant materials, or adding flame retardants and self-extinguishing products [38]. Zhang, Li [56] developed a flame-retarded composite PCM comprised of paraffin, EG, ammonium polyphosphate, red phosphorus, and epoxy resin. The results indicated that the fire retardant PCMs reduced the maximum temperature by 44.7% and 30.1% compared to the air cooling and pure paraffin cooling at 25°C during the 3C discharge rate, respectively. The temperature difference was also shown to be below 5°C even at an environment temperature of 45°C, making it suitable for BTMS, energy storage systems, and other applications.

The freezing behavior of PCMs also plays a prominent role in PCM selection for the desired applications. A PCM may not be appropriate if it shows incongruent freezing, extremely slow crystallisation, or viscous mixture formation. Another serious issue of many PCMs, particularly salt hydrates, is supercooling, which occurs when the temperature decreases considerably under the melting point before the solidification starts, usually because of the slow nucleation rate (the formation rate of a small cluster of the new phase). Consequently, the melted PCM does not solidify at the melting point, leading to the reduced efficiency of PCMs by preventing heat extraction. Supercooling is typically related to the viscosity of the melted PCM at the melting point, which has low diffusion coefficients. Problems related to nucleation can be solved by either homogenous nucleation with no added materials through techniques such as ultrasonic waves or heterogeneous nucleation, where impurities in the melted PCM act as a nucleating agent. The impurities should have a melting point more than the maximum temperature reached by the PCM, be water-irresolvable at all temperatures, and not be chemically reactive to the hydrate [38].

4 Solid-liquid PCMs in BTMS

The solid-liquid PCMs are used in BTMS in three different ways. One technique is to embed the batteries or battery packs into the PCMs directly. Another technique is the indirect contact between PCMs and batteries, which can prevent some issues arising from the direct contact between the PCM and battery such as the corrosion effect of the PCM to the battery surface or the battery short circuit [57]. Zhao, Gu [58] also proposed an internal passive cooling system for Li-ion batteries by placing PCM filled mandrel (n-octadecane and n-eicosane) into the batteries. This system with less PCM achieved lower temperature increases and more uniform temperature distributions compared to external BTMSs, which could also use the full latent heat capacity of the PCM to control the generated heat from the battery. The features of solid-liquid PCMs, which are categorized as organic, inorganic, and eutectic PCMs, are explained in sections 5, 6, and 7.

5 Organic PCMs in BTMS

Organic PCMs are classified as paraffins and non-paraffins. They have a melting point in the range of 0 to 200°C, usually latent heat in the range of 128-200 kJ/kg, and are mostly stable at high temperatures due to their carbon and hydrogen contents [59]. Furthermore, organic PCMs usually have lower densities (less than 1g/cm^3) than most inorganic PCMs, resulting in lower volumetric latent heat of fusion than inorganic PCMs. They also have outstanding thermal reliability regarding changes in thermal properties during thermal cycling [36].

Paraffins comprise straight-chain alkanes with the chemical formula of C_nH_{2n+2}, where $n \leq 40$. Increasing the number of carbon atoms or chain length increases the melting temperature of paraffin wax and latent heat of fusion. Since pure paraffin is expensive and moderately flammable, hydrocarbons mixtures with different chain lengths are usually utilised due to their safety, non-corrosiveness, low cost, wide availability, broad phase transition temperature ranges compared to pure paraffin, and freezing without supercooling [38, 60]. Paraffins have high latent heat, slight volume change during phase change, and low vapour pressure in the liquid phase. They are also chemically inert and stable without phase segregation under 500°C, making them dependable and predictable in most practical applications and academic research [38, 57, 61]. Furthermore, they have latent heat of about 200 kJ⁄kg, low thermal conductivity of about 0.2W⁄m°C, and no compatibility with plastic containers [38, 62]. While the low thermal conductivity of pure paraffin restricts its application in the automotive industry, it has been used in many studies on BTMS [63-66]. Table 2 shows a list of different paraffins with the number of carbon atoms, their thermo-physical properties, and price range.

Table 2.
The number of carbon atoms, properties, and price range of different paraffins [45]

Paraffins	C number	Density (kg/m^3)	Melting Temperature (°C)	Specific Latent Heat (kJ/kg)	Energy Density (kWh/m^3)	Minimum Price (£/kWh)	Maximum Price (£/kWh)
n-Tetradecane	14	764	5.5	228	48.39	8.27	10.33
n-Pentadecane	15	769	10	205	43.79	9.13	11.42
n-Hexadecane	16	770	16.7	237.1	50.71	7.89	9.86
n-Heptadecane	17	777	21.7	213	45.97	8.70	10.88
n-Octadecane	18	777	28	244	52.66	7.60	9.49
n-Nonadecane	19	786	32	222	48.47	8.25	10.32
n-Eicosane	20	792	36.7	246	54.12	7.39	9.24
n-Henicosane	21	792	40.2	200	44.00	9.09	11.36
n-Docosane	22	794	44	249	54.92	7.28	9.10
n-Tricosane	23	797	47.5	232	51.36	7.79	9.73
n-Tetracosane	24	799	50.6	255	56.60	7.07	8.83
n-Pentacosane	25	812	49.4	238	53.68	7.45	9.31
n-Hexacosane	26	803	56.3	256	57.10	7.00	8.76
n-Heptacosane	27	802	58.8	236	52.58	7.61	9.51
n-Octacosane	28	807	61.6	253	56.71	7.05	8.82

Non-Paraffins include fatty acids, alcohols, ester, and glycols. In general, non-paraffins are known for their high latent heat, not being flammable, low thermal conductivity, slight toxicity, and instability at high temperatures. Fatty acids, the most prominent non-paraffin PCMs, have higher latent heat than paraffins, no supercooling or subcooling, chemical stability, minor volume change, high energy density, no phase segregation, no toxicity, and compatibility with container materials (no corrosion). However, they are more expensive than other PCM materials. Their price is about 2 to 2.5 times more than paraffins [38, 45, 59, 62]. In chemistry, fatty acids are frequently represented by the chemical formula $CH_3(CH_2)_nCOOH$, where n is the number of chains. Their behaviour is similar to paraffins but with greater cycling stability. Promising research has revealed that mixing different fatty acids can achieve melting temperature ranges from 20°C to 30°C [38, 55, 67].

5.1 Challenges of Organic PCMs and Solutions

While organic PCMs are used in BTMS due to their excellent properties, pure organic PCMs such as paraffin have some weaknesses such as liquid phase leakage, low thermal conductivity, volume change, poor mechanical properties, and volume resistivity, which may limit their applications [4, 54, 57, 68]. These weaknesses can be improved through various techniques such as using additives that will be discussed in subsections 5.1.1, 5.1.2, and 5.1.3. The PCMs modified with additives are composite phase change materials (CPCM) or enhanced PCMs [43, 69].

5.1.1 PCM Volume Change and Leakage and Possible Solutions

In PCM-based BTMS, the solid-phase PCM melts by absorbing heat from the batteries. Therefore, PCMs undergo a volume change during the phase change process, which might lead to PCM leakage and affect the heat extraction capability of PCM. Several techniques are applied to tackle the PCM leakage problem during PCM melting.

One solution to molten PCM leakage is to embed porous materials in PCMs to absorb liquid PCMs and mitigate the leakage of melted PCMs [57]. Lv, Yang [70] proposed a novel BTMS using aluminum fins on the CPCM made of EG, as a porous material, paraffin, and low-density polyethylene. The CPCM significantly prevented liquid PCM leakage. The fins also enhanced surface heat transfer capability. Using porous materials such as EG cannot wholly prevent PCM leakage. Adding EG will also decrease the volume fraction of pure PCM per unit volume, which will reduce the latent heat of PCM. To overcome the leakage problem, PCMs can be encapsulated using organic or inorganic materials for capsules to restrict the PCM volume change during solid-liquid phase transition to the capsule size to effectively avoid the leakage of melted PCM [57]. In addition, the capsules can stop interaction between PCMs and the environment. Various technologies can be used to encapsulate the PCMs, which depend on the capsule materials and chemical properties of the PCMs, including spray drying, interfacial polymerization, suspension polymerization, emulsion polymerization, and coacervation techniques [68].

Another technique to control PCM leakage is using nano-silica (Nano-SiO_2). CPCM enhanced by Nano-SiO_2 could considerably prevent PCM leakage and volume change during the PCM melting process in BTMS [71]. Lv, Situ [71] investigated Nano-SiO_2 enhanced CPCM to control the volume change of CPCM. The volume change was tested by measuring the linear thermal expansion increment (L-TEI) using a thermo-mechanical analyzer. The L-TEI of CPCM without Nano-SiO_2 and Nano-SiO_2 enhanced CPCM were 18.2 and 10.7 mm/m at the temperature of 60°C, respectively. Therefore, the volumetric thermal expansion in CPCM without Nano-SiO_2 was quadruple that of Nano-SiO_2 enhanced CPCM. This proposed CPCM also provided improved cooling performance and a long-lasting battery cooling system. Furthermore, Fathabadi [72] proposed a hybrid cooling system consisting of air cooling and PCM/EG composite. It has been reported that the volume of the CPCM with EG increased by 20% during the melting process. Therefore, a volume expansion receptacle attached to the battery module was used to store the expanded liquid CPCM (shown in Figure 2).

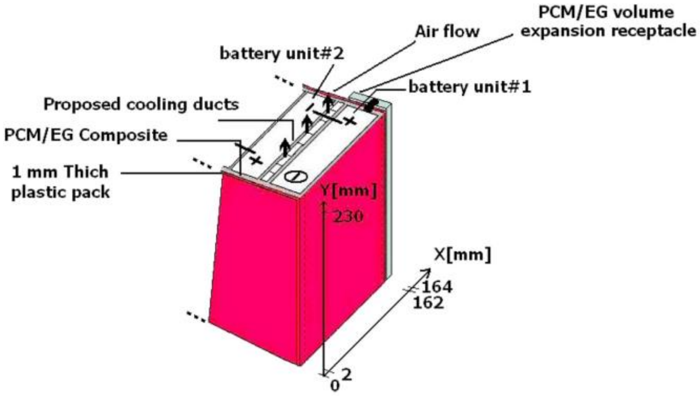

Figure 2. Battery pack with the volume expansion receptacle for the hybrid BTMS [72]

5.1.2 Low Thermal Conductivity of PCMs and Possible Solutions

Although organic PCMs in BTMS suffer from low thermal conductivity, there are several techniques to enhance the thermal conductivity. The first strategy to deal with low thermal conductivity is to choose an alternative for organic PCMs, such as inorganic PCMs or hydrate PCMs, which have higher thermal conductivity than pure organic PCMs [57]. However, other features of these materials should also be evaluated to choose an appropriate PCM, depending on the application. Other techniques to improve the thermal conductivity of organic PCMs include using PCM encapsulation, metallic materials such as copper, aluminum, nickel, and stainless steel in various forms such as fins and honeycomb as well as using carbon-based materials, nanomaterials, and porous materials [73]. These techniques will be discussed in the following subsections.

5.1.2.1 PCM Encapsulation

As discussed in subsection 5.1.1, PCM encapsulation is a technique to control PCM leakage during the PCM melting process. The encapsulation technique can also cause the thermal conductivity enhancement of PCMs using some methods. While the capsule walls are mostly made of organic polymer materials, including polymethylmethacrylate, polyethylene, and styrene-maleic anhydrous copolymer [74], the majority of organic polymers showed low thermal conductivity [75]. Some optimization measures such as adding magnetic iron (II, III) oxide (Fe_3O_4) nanoparticles to PCM nano-capsules [76] or adding nano-titanium dioxide (TiO_2) [77] have been proposed to enhance the thermal conductivity of the capsules. PCM capsule technology is not widely used in battery heat control, mainly because of its high manufacturing costs [57].

5.1.2.2 Metallic Materials

A solution for enhancing the thermal conductivity of PCMs is to add metallic materials, such as metal meshes, metal fins, metal fibers, metal foils, metal compounds, and metal containers. Therefore, the thermal conductivity of PCMs significantly increases with a slight change in the latent heat [78]. This technique with low manufacturing cost is worth developing and applying in BTMS. However, the possible electric leakage in the battery module due to the metallic materials is a risk [57].

Wu, Yang [78] developed a CPCM made of paraffin and EG with copper mesh, which enhanced the system’s mechanical properties and thermal conductivity (shown in Figure 3). The results showed that the maximum temperature of the battery pack with PCM plate enhanced by copper mesh and PCM plate was 61.6°C and 65.5°C at 5C discharge rate, respectively. Furthermore, Azizi and Sadrameli [37] experimentally and numerically investigated a BTMS with Poly Ethylene Glycol 1000, as the PCM, enhanced by aluminum wire mesh plates to control the batteries’ temperature at a high environmental temperature of 50-55°C. As a result, the maximum temperature of the battery cell surface decreased by 19%, 21%, and 26% at 1C, 2C, and 3C discharge rates, respectively.

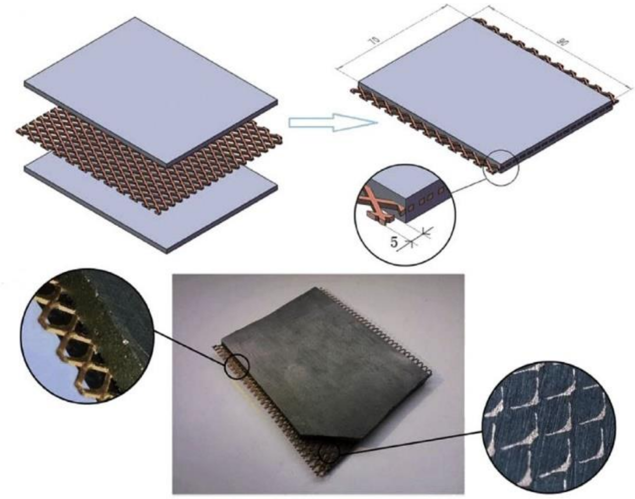

Figure 3. The structure of the CPCM plate enhanced by copper mesh [78]

Fins have been widely used to enhance PCM thermal performance for different applications such as electronic devices, space, solar heat storage, and BTMS, which improve the heat transfer area, have a simple structure, and are easy to fabricate. Fin structures improve conduction and natural convection heat transfer, which can help to use the whole latent heat capacity of PCM [66]. Metal fin structures are combined with organic PCMs in two ways, embedding the fins within the PCM or placing them on the outer surface of the PCM. The immersed metal fins can decrease the thermal resistance, resulting in cooling performance enhancement [57]. Essential factors for using metal fins include fin material, fin quantity, fin size, and fin shape.

While fins are usually made of copper or aluminum because of their high thermal conductivity, aluminum is mainly chosen due to its low cost and weight [49]. Increasing the quantity of fins and fin size in a battery module with limited space does not necessarily improve the fin structure efficiency due to the decrease in the amount of PCM material, leading to a decrease in the energy storage capacity of the PCM. Therefore, an optimized quantity and size of fins should be determined [57, 66, 79]. Four rectangular fins showed the best performance for cooling a cylindrical Li-ion battery cell [61]. Choudhari, Dhoble [49] also proposed an optimized PCM module with four I-shaped fins. Longitudinal fins within the PCM showed better convection heat transfer to the ambient air, while circular fins showed better conduction heat transfer due to the increased surface area for heat transfer [61]. Weng, Yang [80] found that cylindrical batteries with a PCM cooling system cannot efficiently dissipate heat from the bottom, leading to an almost higher temperature at the bottom. Therefore, a combination of four longitudinal fins and two circular fins were designed in the upper and lower parts of the battery cell, respectively, to achieve an improved heat balance in the battery cell (shown in Figure 4) [61].

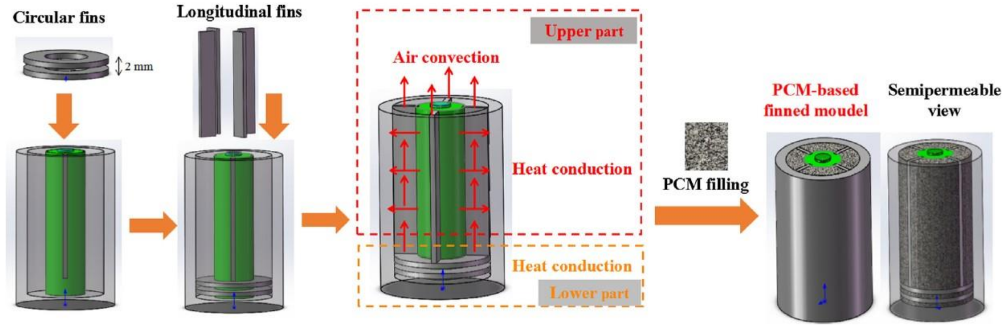

Figure 4. PCM module enhanced by longitudinal and circular fin structures [61]

The melted PCM, which usually has a lower density compared to the solid PCM, occupies the top part of the container, while the solid PCM with greater density stays at the bottom of the container [81]. Since buoyancy forces and gravity affect the PCM melting, Shojaeefard, Molaeimanesh [81] numerically investigated the effect of fin alignment within paraffin wax in an aluminum container with a 6 mm thickness and 40°C constant temperature. The alignment of fins affected the breaking state of the solid PCM, and horizontal fins showed the best cooling performance with the most significant liquid fraction. Karami and Kamkari [82] numerically investigated the effect of different inclination angles (from 0° to 180°) of rectangular enclosures on the buoyancy-driven convection flows. The enclosure was filled with lauric acid PCM (suitable for medium-temperature applications) with one and three aluminum fins, which were heated from one side. PCM melting time decreased by reducing the inclination angle, with the minimum melting time in a horizontal enclosure with heating from the bottom surface due to the intensification of the natural convection flows and increase in the number of vortices in the liquid PCM. The total heat increased by increasing the inclination angle and decreasing the quantity of fins. Furthermore, Ping, Peng [66] investigated the effects of three different paraffin waxes (n-docosane, n-tetracosane, and n-pentacosane) with and without fin structures for BTMS in a prismatic ($LiFePO_4$) battery module. The thermal contact resistance between different materials, which is primarily affected by the roughness, the interstitial air, and the contact pressures, were considered to enhance the accuracy of the numerical simulation. The results revealed that the optimized design of the PCM with fins kept the maximum temperature of the battery module below 51°C at 3C discharge rate.

Although incorporating fins into PCM improves PCM thermal conductivity, it also hastens the PCM melting process. Therefore, the heat absorption capability of PCMs diminishes quickly during the melting process, and PCMs function as an insulator once they have melted completely. Further research is needed to increase the heat absorption capability of PCMs for a longer duration. While a hybrid BTMS can help prolong the PCMs melting process or solidify them repeatedly, it might increase the system’s weight and maintenance costs. Another technique to lengthen the PCM melting process, improve solidification of the liquefied PCM, and enhance heat transfer is to utilize internal-external fins, which have already been applied in solar systems [49].

The application of metal fibre in BTMS is almost few [57]. Pan et al. [83] developed CPCMs by using the cutting copper fibre sintered skeleton in the paraffin (shown in Figure 5). The experiments revealed that the CPCM enhanced by copper fibre effectively improved the heat transfer performance compared to pure paraffin and paraffin enhanced by copper foam, with the temperature difference within 5°C.

(a) Sample of the pure paraffin (b) Sample of the copper foam/paraffin

(c) Sample #1 of the CCFSS/paraffin (d) Sample #2 of the CCFSS/paraffin

Figure 5. Samples of pure PCM, PCM enhanced by copper foam, and PCMs enhanced by copper fibre [83]

Karimi, Behi [84] wrapped aluminum grid foil around a battery module consisting of two prismatic Li-ion battery cells to improve the paraffin thermal conductivity (shown in Figure 6). Aluminum foil increased the systems’ cost, volume, and weight, which were undesirable in vehicles, emphasizing the need for optimization to balance system efficiency and constraints. A PCM thickness of 7 mm was almost ideal in terms of expense, weight, and volume. The results revealed that aluminum foil decreased the maximum temperature of the battery module by 20% and 13%

compared to forced-air convection and pure paraffin, respectively. It would be helpful to explore other materials with a higher thermal conductivity compared to aluminium for use as the grid foil in PCM-based BTMS, considering a balance between the system efficiency and restrictions.

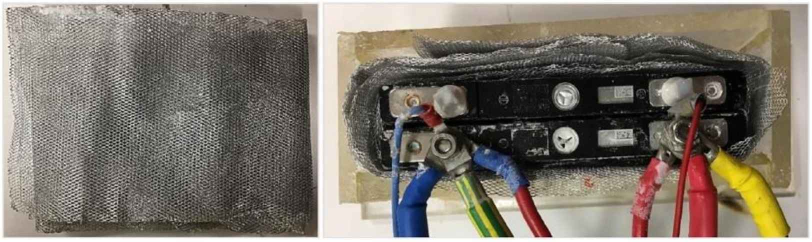

Figure 6. The aluminum foil (left picture) and aluminum foil wrapped around the battery module (right picture) [84]

5.1.2.3 Carbon-Based Materials

Carbon-based materials such as graphene [85, 86], carbon fiber [87], and graphite [88] can be used to improve the thermal conductivity of pure PCM in BTMS.

The thermal conductivity of graphene is very high, in the order of 2000-3000 W/(m·K). Graphene enhanced CPCMs have excellent mechanical properties and low thermal expansion coefficients [36]. The thermal conductivity of CPCM enhanced by graphene is about double that of pure PCM [85]. However, CPCM enhanced by graphene cannot absorb the melted PCM. Since graphene has a high manufacturing cost, other low-priced materials with high thermal conductivity are added to the suitable amount of graphene to enhance the PCM thermal conductivity for electric vehicle applications. While adding graphene to pure PCM increases the PCM thermal conductivity, the latent heat of PCM decreases. Therefore, an optimization between the thermal conductivity and latent heat of CPCM is required. When CPCM melts, graphene is likely to accumulate at the bottom of the container because of its higher density compared to pure PCM, thus limiting its application [57]. It is also important to be aware that the thermo-mechanical properties of graphene-enhanced CPCM become weaker at higher temperatures [36].

Carbon fibers have high thermal conductivity, excellent chemical stability, large specific surface area, and low density, making them suitable to be used as thermally conductive materials [87]. Samimi, Babapoor [87] numerically studied the thermal performance of CPCM enhanced by carbon fibre for cooling of a Li-ion battery cell. The results showed that a more uniform temperature distribution was achieved by increasing the mass fraction of carbon fibre with the minimum and maximum thermal conductivity improvement of 85% and 155%, respectively. Furthermore, Babapoor, Azizi [48] added different sizes and mass fractions of carbon fibres to paraffin to act as a heat transfer promoter. The results showed that CPCM with 2mm-long and 0.46% mass fraction of carbon fibre reduced the maximum temperature rise by up to 45%.

Graphite sheets have high thermal conductivity, low thermal expansion ratio, and low surface hardness. Graphite sheets and EG matrix were used to improve the low thermal conductivity of pure paraffin to control the increase in the battery temperature and improve the battery temperature uniformity (shown in Figure 7) [88]. As a result, the maximum temperature differences were less than 5°C at 1C and 2C discharge rates, using graphite sheets and EG matrix.

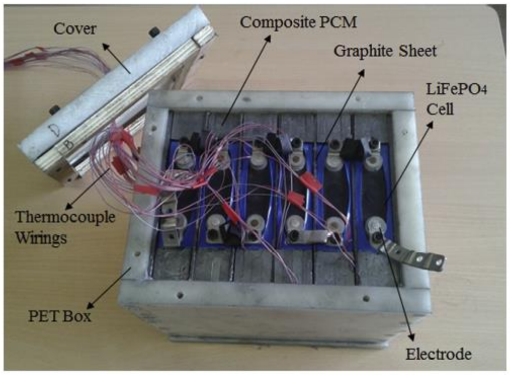

Figure 7. PCM-based BTMS enhanced by graphite sheets and EG matrix [88]

5.1.2.4 Nanomaterials

Nanomaterials such as nanocarbon materials, nano oxide materials, and metal nanomaterials are used to improve the thermal conductivity of pure PCMs. Nanocarbon materials include carbon nanotubes, carbon nanofibers, graphite nano-powder, and graphene nanosheets. Nano oxide materials comprise nano metal oxides and nano non-metallic oxides [57, 90-93]. Despite improved thermal conductivity due to the addition of nanomaterials to PCMs, it might not be economical to use nanomaterial-enhanced PCMs in electric vehicles due to the high cost of nanomaterials [57].

Khodadadi, Fan [89] concluded that carbon-based nanostructures and carbon nanotubes showed significant thermal conductivity improvement compared to the metallic or metal oxide nanoparticles because of their high aspect ratio. Zou, Ma [93] experimentally investigated the effect of adding various proportions of multi-walled carbon nanotubes (MWCNT), graphene, and MWCNT/graphene to the pure paraffin on the thermal conductivity improvement to prevent a rapid temperature increase in liquid PCM. As a result, the MWCNT/graphene CPCM with a mass ratio of 3/7 showed the best heat transfer enhancement in Li-ion powered BTMS. Adding MWCNT/graphene to the pure paraffin increased the thermal conductivity by 31.8%, 55.4%, and 124% compared to paraffin enhanced by graphene, paraffin enhanced by MWCNT, and pure paraffin, respectively.

Metal nanomaterials such as copper, silver (Ag), and Fe_3O_4 nanoparticles have been used to improve the thermal conductivity of pure PCM. CPCM enhanced by Ag nanoparticles showed better thermal performance compared to other CPCMs [94]. Mousavi, Siavashi [95] numerically investigated the thermal performance of a vertical cylindrical thermal energy storage system filled with PCM enhanced by aluminum oxide (Al_2O_3) nanoparticles with various nanoparticle concentrations and different quantities of horizontal radial copper fins. Consequently, CPCM with 5% of Al_2O_3 nanoparticles reduced the melting time by up to 5.5%. Furthermore, the nanoparticle-enhanced PCM with 5% of Al_2O_3 and three fins showed the best thermal performance, decreasing the melting time by 28.3%. Rao, Wen [96] proposed copper nanoparticles and copper foam to improve the thermal conductivity of a microencapsulated PCM. As a result, microencapsulated PCM with 5 wt.% of copper nanoparticles (with 50 nm diameter) and microencapsulated PCM with copper foam (30 pore quantity per inch) showed the thermal conductivity to be 1.12 and 3.46 times compared to the microencapsulated PCM at the temperature of 20°C, respectively.

The possible adverse effects of using nanomaterials to enhance the thermal conductivity of PCMs are:

- The liquid PCM viscosity increases by increasing the nanoparticles volume fraction, which affects the convection heat transfer of the PCM with the environment [97].
- The amount of pure PCM per unit volume of the composite materials decreases by adding more nanomaterials to the pure PCM, leading to the reduced latent heat of CPCM [57].
- The density of nano oxide materials and metal nanomaterials is greater than that of pure PCMs. Therefore, the nanoparticles will settle at the bottom of the container due to the gravity when PCMs melt. After multiple solid-liquid phase changes of PCM, most nanomaterials settle at the bottom of the pure PCM, which leads to an uneven distribution of nanomaterials in the PCM and significantly influences the heat transfer improvement by nanomaterials [57, 97]. Therefore, alternative techniques such as fins might be preferred to improve the thermal performance of PCMs [65, 98], and further studies should be carried out on the CPCM enhanced by nanoparticles to explore more homogenised structures [49].

5.1.2.5 Porous Materials

Porous materials such as metal foam and EG are commonly used in BTMS to enhance pure PCM properties. The crucial factors for choosing porous materials are pore size and pore density. The advantages of porous materials include the low cost and relatively simple manufacturing procedure. The enhanced PCM with desired thermal conductivity and latent heat can be manufactured by controlling the porosity and pore size, which can be used in electric vehicles. However, the danger of electric leakage in the battery system can increase [57, 99, 100].

Commonly used metal foams are copper foam, aluminum foam, and nickel foam. The advantages of metal foams are the small relative density, high porosity, good thermo-physical properties, large specific surface area, and mechanical strength. In addition, metal foams can also accelerate the melting process of PCMs. Due to the presence of pores in metal foams, the molten PCM will be adsorbed by capillary forces, maintaining the morphology of PCM [57]. The results from [101, 102] revealed the improved thermal conductivity of paraffin wax using aluminum foam for battery systems. The thermal conductivity of paraffin wax was also enhanced to 70 W/(m·K) using copper foam, resulting in higher heat transfer and temperature uniformity [103].

EG, a porous carbon material, is embedded into the pure PCM, resulting in the improved thermal conductivity and mechanical properties of the pure PCM. After several melting and solidifying processes, the CPCM shape will keep stable, and the leakage of the liquid PCM decreases considerably [57]. Jiang, Huang [104] prepared the CPCM sample by absorbing liquid paraffin into the EG with mass fractions of 3%, 6%, 9%, 12%, 16%, 20%, and 30% by capillary forces and surface tension. By increasing the EG mass fraction to 30%, the PCM thermal conductivity increased to 13.85 W/(m·K), and the liquid PCM leakage ratio decreased to 0.38 wt.%. Therefore, the capability of EG to enhance the PCM thermal conductivity and prevent liquid PCM leakage makes porous matrix a suitable candidate for temperature control of Li-ion batteries in BTMS. However, EG-enhanced CPCM cannot control the temperature of batteries at high discharge rates and long-term discharges. Furthermore**,** using large amounts of EG will highly decrease the latent heat of EG-enhanced CPCM due to the decreased amount of pure PCM per unit volume. Therefore, further improvement of the thermal conductivity of CPCMs is crucial at high discharge rates and long-term discharges. [57].

5.1.3 Poor Mechanical Properties and Volume Resistivity of PCMs and Possible Solutions

Mechanical properties and volume resistivity of PCMs are other important factors to consider while designing PCM-based BTMS. There are several techniques to improve the mechanical properties of PCMs. Huang, Li [105] proposed a flexible CPCM for BTMS to enhance the PCM mechanical properties. Styrene butadiene styrene (SBS), as a flexible supporting material, paraffin, and EG, as a thermal conductivity promoter, were used. The effect of different mass ratios of SBS on the mechanical properties of CPCM was studied. Six different paraffin/EG were mixed with mass ratios of SPS: paraffin/EG at 1:1 and 2:1. SBS improved the flexibility and elastic properties of the CPCM. The bending strength of the 2:1 and 1:1 CPCM was in the range of 0.2 to 0.3 MPa and 0.5 to 0.75 MPa, respectively. Furthermore, the 2:1 CPCM significantly enhanced the impact resistance. The maximum temperature of the battery surface and the temperature difference were also maintained under 46°C and 4°C at 5C discharge rate, respectively. It was also concluded that 4% of EG in the CPCM is optimal to enhance the thermal conductivity of the materials, reaching 0.88 W/(m·K). Furthermore, Alrashdan, Mayyas [106] showed that as mass fraction of paraffin wax in the EG-enhanced CPCM increased, tensile strength, compression strength, and burst strength improved while testing at room temperature, but the tensile strength, compression strength, and burst strength decreased when testing at higher operating temperatures.

While there are other solutions to improve the mechanical strength of pure PCMs, other issues such as volume resistivity have to be considered. Volume resistivity is the resistance to electric current and is closely related to the proportion of additives and chemical stability, which affects battery pack safety [54]. Zhang, Li [54] investigated the effect of various mass fractions of AlN in the CPCMs on the thermal conductivity, mechanical properties, and volume resistivity of CPCM used for thermal management of Li-ion batteries (shown in Figure 8). Consequently, the maximum thermal, mechanical, and electric insulation enhancement was achieved by adding 20 wt.% of the AlN to the PCM. Furthermore, Yuan, Yang [107] developed a silicon carbide (SiC)/EG CPCM. As a result, adding 15 wt.% of SiC increased the volume resistivity and thermal conductivity of the CPCM from 4.9 to 12.5 10^{11} Ω.cm and from 1.1 to 2.0 W/(m·K), respectively, which also enhanced the mechanical strength.

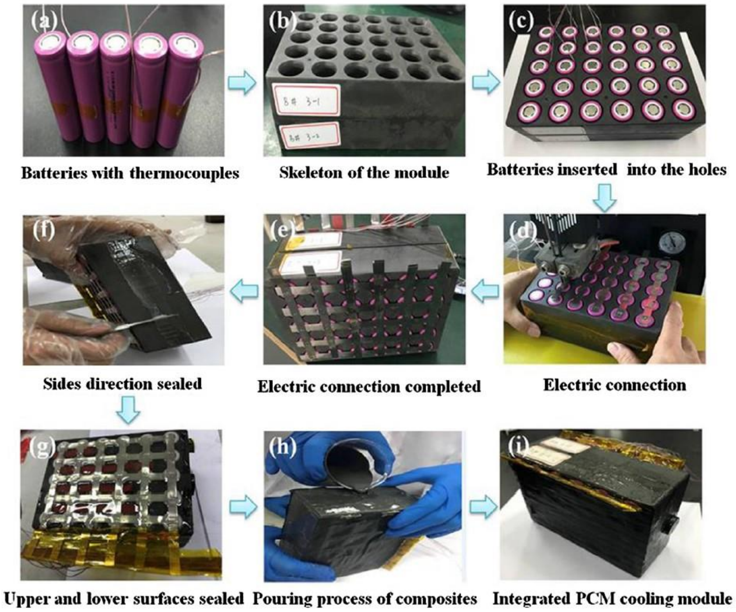

Figure 8. Assembly procedure of the battery module with the CPCM [54]

6 Inorganic PCMs in BTMS

Inorganic PCMs are divided into salt hydrates and metallics, which are made up of inorganic molecules. Inorganic materials have a wide melting temperature range of more than 5-130°C, sharp melting points, and high latent heat of more than 220 kJ/kg. They are also thermally safe and can have thermal conductivities up to three times higher than organic PCMs [6, 38, 108]. These properties make them suitable for a wide range of applications. However, the poor stability of inorganic PCMs' properties due to thermal cycling and corrosion between the PCM and the container has limited their application as latent heat energy storage systems [6, 38, 109].

Metals are a broad class of inorganic PCMs with low latent heat of fusion per unit mass, high latent heat of fusion per unit volume, high thermal conductivity, low specific heat, low vapour pressure, stable melting behaviour, and excellent cyclic performance, making them suitable for various applications [38, 110]. However, metallic PCMs are rather expensive, almost 2 or 2.5 more expensive than paraffins [22]. One of the most important issues of metallic PCMs is the chemical interactions between the molten metallic PCMs and containers, which can be resolved by encapsulation of PCMs [110]. Even though metals are heavy and have relatively high melting points, which are not favourable for PCM applications, they have become more practical due to the recent development in nanomaterial fabrication. Most metal salts such as nitrates, carbonates, and sulfates have high melting temperatures and are appropriate for thermal energy storage applications at 200°C to 1000°C [111, 112]. The most appealing metallic PCMs are bismuth (Bi), lead (PB), indium (In), Tin (Sn), cadmium (Cd), and their alloys because of their low melting temperatures [38]. Liu, Saman [113] reported a list of metals and metallic alloys, which have the potential to be used as PCMs. Table 3 shows some of the compounds in this list as well as their thermo-physical properties.

Table 3.
A list of metals and metallic alloys that can be used as PCMs [113]

Compound	Melting temperature (°C)	Latent heat (kJ kg^{-1})	Density (kg/m^3)		Specific heat (kJ kg^{-1} K)		Thermal conductivity (W/m·K)	
			Solid	Liquid	Solid	Liquid	Solid	Liquid
Pb	328	23						
Al	660	397						
Cu	1083	193.4	8800				350	
Mg–Zn (46.3/53.7 wt%)	340	185	4600					
Al–Mg–Zn (59/33/6 wt%)	443	310	2380		1.63	1.46		
Al–Cu–Mg–Zn (54/22/18/6 wt%)	520	305	3140		1.51	1.13		
Al–Si–Cu (65/5/30 wt%)	571	422	2730		1.30	1.20		
Al–Si (12/86 wt%)	576	560	2700		1.038	1.741	160	
Zn–Cu–Mg (49/45/6 wt%)	703	176	8670		0.42			

Hydrates contain water molecules (the host compound) and other compounds (the guest compound). The guest compounds determine the thermal properties of hydrates [114]. The application of hydrate PCMs in BTMSs is currently few, which will enhance in the future by synthesising hydrates using the guest complex with suitable thermo-physical properties. Hydrates include organic hydrates such as trimethylolethane hydrate as well as inorganic salt hydrates [57].

Salt hydrates are a mixture of salt and water molecules. When they solidify, their structures form a crystalline matrix [22]. Salt hydrates have high latent heat of fusion per unit volume, high thermal conductivity of up to two times that of the paraffin wax, small volume change during phase transition, compatibility with plastics, very low toxicity, and higher densities compared to paraffins, making them great candidates for many applications [38]. However, salt hydrates can be unstable, prone to segregation and corrosion, and can accumulate at the bottom of the container in a considerable supercooled phase [115]. The solid-liquid phase transition temperature of most salt hydrates is under 60°C [57, 116, 117]. Salt hydrates such as disodium hydrogen phosphate, dodecahydrate, and sodium sulfate decahydrate have great potential as PCM materials due to their wide melting temperature range of 5°C-130°C [38, 59, 108]. The application of salt hydrates in the automotive industry and BTMS has not been thoroughly investigated in the literature [22].

7 Eutectic PCMs in BTMS

Eutectic mixtures are innovative PCMs, which combine organic and inorganic compounds that melt and solidify to form a mixture of component crystals during crystallisation [108]. Eutectics have higher latent heat and melting points than each compound [22], which have been reported in very few studies for their use in automotive applications. These mixtures can consist of only salts, salt hydrates, alkalis, or a mixture of two or all of them. The properties of the eutectic hydrated salt and salt hydrates are similar [57]. Due to the high melting temperatures, all compounds can melt and freeze at the same time. As a result, they can be used to develop specific melting points for certain applications. They usually melt and solidify without any segregation and change in components. While eutectic materials do not suffer from incongruent melting due to their miscible components, they are primarily made of inorganic salts that might lead to non-homogenous volume expansion, which can be harmful to the container or the internal structure. Eutectic PCMs are also more expensive than other PCMs with lower latent heat [38].

Liu and Yang [118] investigated the effect of different mass fractions of TiO_2 nanoparticles on the specific heat and latent heat improvement of eutectic hydrated salt PCM. As a result, adding 0.3 wt.% of TiO_2 nanoparticles to eutectic hydrated salt increased the latent heat and specific heat capacity by up to 6.4% and 83.5%, respectively. This CPCM offered a promising approach to achieve high heat energy storage systems and can be used in BTMS in the future.

8 Conclusion

Li-ion batteries, which are widely used in EVs and HEVs, are temperature-sensitive. Therefore, a reliable BTMS is crucial to prevent heat accumulation, thermal runaway, capacity fade, and self-discharge in the battery pack to ensure efficient performance within a suitable temperature range. Considering the effectiveness of BTMSs and analyzing their thermal performance at different operating conditions are crucial for developing viable novel BTMSs. Therefore, the constraints and advantages of thermal management systems should be taken into full consideration before designing the systems. PCMs, as practical and low-cost passive BTMSs, mainly depend on the latent heat capacity to absorb the heat produced by the battery cells. PCM is used as pure PCM, CPCM, or PCM-based hybrid systems.

Solid-liquid PCMs are the commonly used PCMs in automotive applications due to large latent heat and small volume change during phase change. This review paper discussed different types of solid-liquid PCMs, including organic (paraffins and non-paraffins), inorganic (salt hydrates and metallics), and eutectic PCMs (combination of organic and inorganic PCMs), as well as their characteristics. An appropriate PCM should be selected based on the targeted application, considering factors such as high thermal conductivity, high latent heat, high specific heat, low volume change, melting temperature, easy availability, reasonable cost, chemical stability, high recyclability, low environmental impact, non-poisonousness, non-flammability, and little or no subcooling and supercooling.

The effectiveness and some of the challenges of solid-liquid organic PCMs for thermal management of Li-ion batteries have also been discussed. Easy leakage, low thermal conductivity, poor mechanical properties, and volume resistivity of solid-liquid organic PCMs have limited their application in some fields, especially in battery systems. There are several techniques to overcome these weaknesses. The volume change and liquid-phase leakage of organic PCMs could be resolved by using porous materials such as EG, Nano-SiO_2 enhanced PCMs, PCM encapsulation, and volume expansion receptacle. However, PCM encapsulation is not common in automotive applications due to high manufacturing costs. Furthermore, low thermal conductivity of organic PCMs can be improved through techniques such as adding metallic materials, carbon-based materials, nanomaterials, and porous materials. Poor mechanical properties of organic PCMs can also be improved by using additives such as SBS or increasing the mass fraction of paraffin wax in the EG-enhanced CPCM at room temperature. The volume resistivity of organic PCMs can also be enhanced by adding AlN or SiC to the PCM.

There is limited research on the application of non-paraffin organic PCMs, salt hydrate inorganic PCMs, and eutectic mixtures in BTMSs. Mixing different fatty acids for automotive applications are promising future research areas. More research is also needed to increase the heat absorption capacity of PCMs for a longer duration. Further optimized low-cost techniques with improved thermo-physical properties of pure PCMs should be provided to commercialize these passive BTMSs.

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