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Review

Organic and ionic liquid-based phase change materials: Advances in property tailoring and applications

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ABSTRACT

The growing use of intermittent renewable energy sources has increased the need for efficient thermal energy storage (TES), with phase change materials (PCMs) offering high latent heat (LH) storage within a narrow temperature range. This review critically examines conventional organic PCMs, their property-enhancement strategies, emerging ionic liquid (IL)-based PCMs, computational design approaches, and major TES applications. Experimental and computational studies are synthesized to evaluate nanoparticle incorporation, encapsulation, ionic liquids (ILs), functionalized ionic liquids (FILs), deep eutectic solvents (DESs), and computer-aided molecular design (CAMD) methods. The review provides an integrated perspective linking material properties, molecular design, computational screening, and application-specific requirements. The literature shows that nanoparticle incorporation can markedly improve thermal conductivity, although its effects on LH and long-term stability depend on PCM-additive combination and additive loading, while encapsulation helps reduce leakage, protect the PCM from the surrounding environment, and improve heat-transfer performance. ILs, FILs, and DESs exhibit tunable phase-change behavior, with molecular structure, functional groups, and hydrogen-bonding interactions influencing their LH storage capacity. QSPR and machine-learning models enable rapid prediction and screening, but remain limited by scarce high-quality data and validation. Applications in buildings, solar energy storage, and battery thermal management further highlight the need for application-specific PCM design and long-term performance evaluation. Collectively, these findings identify key directions for the rational development and practical deployment of advanced PCMs.

1. Introduction

Against the backdrop of growing concerns over global energy security and carbon neutrality goals, renewable energy sources (e.g., solar power) have been increasingly adopted as viable alternatives to fossil fuels during the past decades. However, the large-scale utilization of such energy still suffers from the inherent intermittency caused by natural factors (e.g., diurnal cycles) and climatic conditions (e.g., cloud cover and precipitation), which severely undermines the stability of energy supply [1]. To address this critical bottleneck, thermal energy storage (TES) technology has emerged as a pivotal solution in bridging the demand-supply gap by delivering heat during the trough and peak, thereby garnering significant attention from both industrial and academic communities [2–5].

TES technology is generally categorized into three main types based on its energy storage mechanism, i.e., sensible heat storage (SHS), latent heat storage (LHS), and thermochemical heat storage (THS) relying on reversible chemical reactions [6,7]. Each type exhibits distinct technical

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characteristics and application scopes; among them, LHS is particularly prominent due to its exceptional high latent heat (LH) storage capacity and nearly isothermal operation within a narrow temperature range [8]. The core medium enabling LHS is phase change materials (PCMs), to be specific, PCMs absorb or release substantial amounts of LH during phase transitions (predominantly solid-liquid transitions), providing thermal energy storage support for heating or cooling systems. Owing to their unique ability to mitigate thermal mismatches and improve energy utilization efficiency, PCMs are widely recognized as promising candidates for diverse energy-related and thermal management applications, such as building energy efficiency, solar thermal energy storage, waste heat recovery, battery thermal management, microelectronic device temperature control, and photovoltaic/thermal systems [9–12].

Ensuring efficient thermal energy storage (TES) systems critically contingent on the rational selection of appropriate PCMs, as the performance of PCMs directly determines the energy storage density, operating stability, and cost-effectiveness of TES systems. Typically, the desirable PCMs exhibit the following key characteristics: (1) an appropriate phase change temperature that matches the temperature range of the target application; (2) high latent heat capacity to maximize energy storage and high thermal conductivity (TC) to facilitate rapid heat transfer; (3) excellent thermal, chemical and structural stability to ensure long service life; (4) minimal supercooling to avoid energy loss and ensure stable phase change; and (5) cost-effectiveness to enable large-scale commercialization [2,13–16].

Given the vast diversity of available PCMs, a systematic classification is essential to guide material selection and optimize application-specific design. Broadly, PCMs are divided into two primary types based on their chemical composition: inorganic PCMs and organic PCMs [17]. Inorganic PCMs (e.g., salt hydrates, anhydrous salts and metallic compounds) possess high TC, high volumetric LH and non-flammability, while most inorganic salts are plagued by inherent corrosiveness and significant supercooling, severely restricting their practical application [17–20]. In contrast, organic PCMs, predominantly carbon-based compounds (e.g., paraffins, fatty acids, alcohols, glycols, esters), offer distinct merits including non-corrosiveness to most substrates, minimal supercooling and high gravimetric LH capacity. Yet their poor TC, moderate flammability and potential volatility pose constraints on their widespread adoption in TES systems [21–23]. These drawbacks of conventional PCMs have spurred intensive research into advanced high-performance heat storage materials, an endeavor central to enhancing thermal management efficiency and encouraging practical sustainable energy solutions.

Ionic liquids (ILs), organic salts composed of organic cations paired with organic or inorganic anions, have been thrust into the limelight as novel promising TES materials [24,25]. Compared with the aforementioned conventional PCMs, ILs exhibit a number of unique and highly tunable properties that address key limitations of traditional PCMs, positioning them as a compelling alternative for advanced TES systems. Notably, their low volatility, high thermal stability, and low flammability render ILs exceptionally safe candidates for TES applications in demanding high-temperature scenarios, such as battery thermal management and fuel cell thermal regulation [26–28]. Furthermore, their excellent solvation capacity enables them to serve as carriers or matrices for functional additives (e.g., thermally conductive nanoparticles), providing a viable strategy to mitigate the poor TC of conventional organic PCMs. Additionally, as ILs consist exclusively of ions, they have an extended liquidus range (from room temperature to over 300 °C), making them well-suited for TES systems requiring operation across broad thermal regimes [29–31]. More importantly, the key thermal properties including the IL melting temperature (T_m) and LH, can be precisely designed and fine-tuned via the judicious combination of cationic and anionic moieties [24,25,32,33].

Leveraging the inherent tunability of ILs, functionalized ionic liquids (FILs), a subclass of ILs modified by grafting specific functional groups (e.g., hydroxyl, carboxyl) onto cations or anions, have emerged to further optimize TES-critical properties, such as boosting TC via improved filler compatibility or enhancing LH through regulated intermolecular interactions [29,34–37]. Complementing FILs, deep eutectic solvents (DESs), representative IL analogues comprised of hydrogen bond donors (e.g., urea, glycol) and acceptors (e.g., choline chloride), stand out as cost-effective, eco-friendly alternatives to ILs for TES, offering simple ambient-temperature synthesis and tunable T_m values [38–42]. Together, these IL-based material expand the portfolio of advanced TES media, and growing research efforts have been devoted to exploring their feasibility as high-performance PCMs for TES applications.

Despite the promising performance of ILs and their derivatives (FILs, DESs) as TES media, a critical challenge persists in their practical application as PCMs. Specifically, the vast number of possible cation-anion combinations for ILs renders experimental evaluation of all potential PCM candidates impractical, creating a bottleneck for rapid material screening and development. As a consequence, the effective development and application of ILs rely heavily on theoretical design and computational screening strategies. In recent years, computer-aided molecular design (CAMD) method has been widely used and extended as a reliable tool for tailoring optimal IL molecular structures for diverse applications, including thermal energy storage [43–47] Within this framework, quantitative structure-property relationship (QSPR) models, and machine learning approaches have been developed to predict key physical and thermodynamic properties, such as LH and T_m [48–53]. By specifying target properties as design objectives, optimal IL structures can be rapidly identified through reverse-design logic.

Previous reviews have provided important but largely topic-specific perspectives on TES materials and systems. Extensive reviews have examined conventional organic PCMs, their synthesis, property enhancement, encapsulation strategies, and applications [18,19,54–56]. In addition, recent reviews have explored the potential of ILs as designer PCMs and CAMD approaches for predicting and tailoring the physicochemical properties of ILs across various process applications [25,33]. Collectively, these studies establish solid foundations for each individual area; however, the existing review literature remains largely fragmented across conventional organic PCM modification, emerging IL-based materials, computational property prediction, and end-use applications. Consequently, a unified critical perspective that bridges these areas is valuable for understanding how material-level property tailoring and computational screening can jointly support the development and application-specific selection of high-performance PCMs.

A structured literature search was conducted using Web of Science, ScienceDirect, and Google Scholar to identify studies relevant to the scope of this review. The search considered literature available up to 2026, with emphasis on recent publications, while earlier important studies were retained when required to establish fundamental PCM properties, material classifications, and the development of computational prediction methods.

Given the critical challenges and research needs outlined above, particularly the demand for systematic guidance on PCM selection, the potential of IL-based materials, and the reliance on computational design, this work aims to present a comprehensive review of conventional organic PCMs and IL-based PCMs, covering their fundamental properties, technical advancements, computational screening strategies, and typical applications. The whole review is organized as follows. Section 2 provides a brief overview of organic PCMs and comprehensively explores recent advancements in techniques, including nanoparticle inclusion and encapsulation, to mitigate their inherent drawbacks. Section 3 provides an introductory overview of three representative types of IL-based PCMs, namely common ILs, FILs, and DESs, with emphasis on their structural features and phase change behaviors relevant to TES. Following this, section 4 presents a thorough review of physical property prediction models (e.g., QSPR and thermodynamic models) and comprehensive screening methods within the computer-aided molecular design (CAMD) framework, focusing on their

utility for accelerating IL-based PCM development. Section 5 comprehensively reviews recent advances and innovations in PCM applications, highlights current challenges, and outlines future research directions required for the effective and efficient use of PCM-based systems. Finally, Section 6 concludes this review and discusses future perspectives in this field, identifying key research gaps and directions for advancing the development and practical application of PCMs in sustainable TES. Schematic overview of the review's structural framework is presented in Fig. 1.

2. Organic phase change materials

Generally, organic PCMs are divided into two categories: paraffins and non-paraffin organic PCMs [57]. Furthermore, the non-paraffin organic PCMs include sugar alcohols, fatty acids, esters, and polyethylene glycol (PEG) [58]. Paraffin belongs to the n-alkane family, which comprises long-chain hydrocarbons generally represented by the chemical formula $C_nH_{2n+}2$ [59]. Paraffins are promising materials that offer remarkable properties, including high LH, good chemical and thermal stability, limited supercooling, low vapor pressure, low toxicity, low corrosivity, and low cost. However, paraffins have several limitations, including low TC, substantial volume changes during phase transitions, and flammability. A number of studies have been conducted to address these limitations, including low TC and substantial volume changes. Researchers have investigated the use of stable metal matrices and the incorporation of various additives, such as nanomaterials, into paraffin to address these issues [60–67].

Sugar alcohols are compounds that contain hydroxyl group (-OH) and are suitable for low to medium temperature TES [68]. Similar to paraffins, the phase-change temperature and LH of sugar alcohols generally increase with increasing carbon-chain length. Fatty acids are long-chain compounds that contain a carboxylic group and can be obtained from plant and animal sources; thus, they are considered renewable materials. Esters are generally produced through the esterification of fatty acids with alcohols. Owing to their generally lower melting temperatures compared with those of the corresponding fatty acids, they show promising potential for low-temperature TES applications [69]. PEG is a biocompatible polyether used in numerous applications, including as a PCM, and is represented by the general chemical formula $C_{2n}H_{4n+2}O_{n+1}$ [70]. Non-paraffin PCMs also exhibit several advantages, including high LH, limited supercooling, and renewability, depending on the material class. However, some non-paraffin PCMs have limitations such as low TC, volatility, limited chemical stability, high cost, corrosivity, and flammability. A comparative analysis of paraffin and non-paraffin PCMs for TES is presented in Table 1.

The intrinsic limitations of conventional organic PCMs, such as low TC, high volatility, substantial volume changes during phase transitions, and flammability, pose significant challenges to their effective use in various TES systems. The subsequent sections discuss advanced techniques, such as nanoparticle incorporation and PCM encapsulation, to address these limitations.

Table 1
Comparative analysis between paraffin and non-paraffin PCMs [14,68,69,71–74].

Property	Paraffin	Non-paraffin
LH	Generally high; typically ~150–250 kJ kg^{-1}	Moderate to high; strongly dependent on the chemical class
Thermal stability	Usually good within the intended operating temperature range	Variable; dependent on molecular structure and operating temperature
Chemical stability	Mostly high, with good cycling stability	Variable; fatty acids are mostly stable, whereas some other classes may undergo degradation during prolonged thermal cycling
TC	Low	Generally low; varies with material class
Volume change during phase transition	Moderate; appropriate accommodation is required in containment design	Material-dependent and may be significant for certain compounds
Supercooling	Generally low	Ranges from low to high depending on the material; sugar alcohols may exhibit substantial supercooling
T_m range	Broad and strongly dependent on carbon-chain length	Very broad owing to the diversity of fatty acids, alcohols, esters, glycols, and sugar alcohols
Vapor pressure	Generally low	Usually low to moderate, depending on the specific compound
Corrosivity	Typically non-corrosive toward common containment materials	Mostly low but material-dependent; fatty acids may cause mild corrosion of certain metals
Inflammable	Flammable; fire-safety concerns may limit certain applications	Strongly composition-dependent; many non-paraffin organic PCMs are combustible
Cost and availability	Relatively inexpensive and widely available commercially	Highly variable; purified or specialty compounds may be more expensive

2.1. Properties of organic PCMs

The effective design of TES systems requires the selection of PCMs with favorable thermophysical, chemical, and kinetic properties, as well as low toxicity and reasonable cost. Comprehensive criteria for the selection of an appropriate PCM for a TES system are presented in Fig. 2. Organic PCMs are promising materials that offer high LH, good thermal

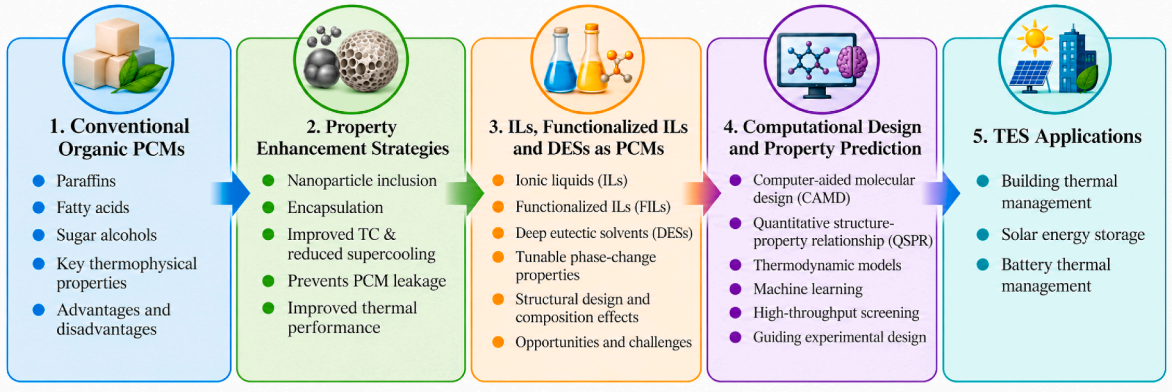


Fig. 1. Schematic overview of the review's structural framework.

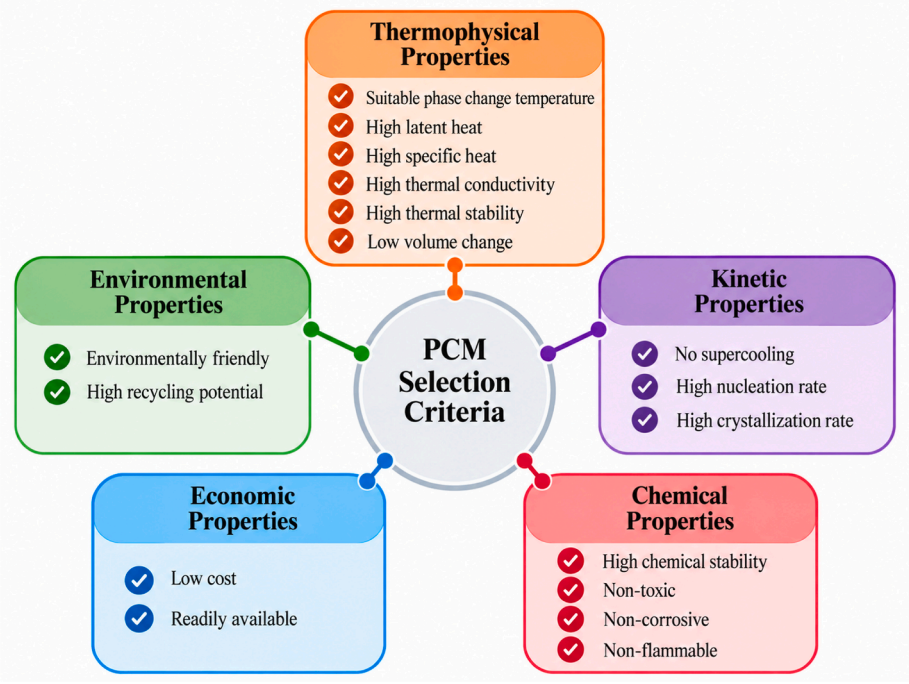


Fig. 2. Comprehensive criteria for selecting a PCM for a TES system.

stability, and low supercooling but suffer from low TC.

Nanoparticles are categorized into three main categories based on their composition: organic, inorganic, and carbon-based nanoparticles. Inorganic nanoparticles include metals and metal oxides, such as Cu, Ag, Al, TiO_2, Al_2O_3, etc., while carbon-based nanoparticles primarily consist of carbon-based structures, including graphite nanoplatelets, carbon nanotubes, graphene, etc. Inorganic and carbonaceous nanoparticles are widely used by researchers to modify the properties of PCMs [75]. In this section, the effects of nanoparticle incorporation on the key properties of organic PCMs are examined.

2.1.1. Latent heat

LH is considered an important thermodynamic property in selecting a suitable PCM for a TES system since it directly affects the gravimetric energy storage density. It signifies the energy required to transform a unit mass of a material from the solid to the liquid state at a constant temperature. Based on the published literature, organic PCMs exhibit high LH values ranging from 143 to 356 kJ/kg, as tabulated in a Table 2. Nevertheless, their low TC significantly limits their applicability in various TES systems; thus, nanoparticle incorporation has been investigated by researchers to mitigate this problem [76,77]. The incorporation of nanoparticles not only improves TC but also influences the LH of the corresponding pure organic PCMs. The reported effects of nanoparticle incorporation on the LH of different organic PCMs are summarized in Table 3. Based on the studies, nanoparticle incorporation into paraffin can either increase or decrease LH, depending on the type of nano-additive and its concentration. In contrast to metal and metal oxide nanoparticles, carbon-based additives have shown the potential to maintain or even enhance the LH of paraffin.

2.1.2. Thermal conductivity

When employing PCMs in TES applications, high thermal charging and discharging rates are desirable. TC is a crucial property of a PCM that influences its thermal charging and discharging rates [77]. The typical range of thermal conductivity for the pure organic PCMs is 0.1-0.3 $W\,m^{-1}\,K^{-1}$, as tabulated in Table 2. Researchers have implemented advanced techniques such as fins, porous structures, and nanomaterials to improve the TC of PCMs [115–123]. However, the use of fins and metallic foams can cause a significant increase in the weight of the TES system; thus, the incorporation of highly thermally conductive nano-additives is considered as efficient method for enhancing TC

Table 2
Thermophysical properties of organic PCMs.

PCMs	T_m (°C)	λ (W/mK)	ΔH (kJ/kg)	Ref.
Paraffin $C_{12}H_{26}$	−12	0.21	189	[78]
Paraffin $C_{13}H_{28}$	−6	0.21	216	[79]
Paraffin $C_{14}H_{30}$	4.5–5.6	0.15	227	[80]
Paraffin $C_{16}H_{34}$	18.2	0.21	238	[78]
Paraffin $C_{18}H_{38}$	28	0.35	245	[81]
Paraffin $C_{30}H_{62}$	65	-	252	[78]
Paraffin $C_{34}H_{70}$	75.9	-	269	[78]
Stearic acid	70	0.172	203	[81]
Myristic acid	54	-	187	[81]
Dimethyl adipate	9.7	0.358	164.6	[82]
Caprylic acid	16.3	0.149	148	[83]
Lauric acid	44	0.22	212	[84]
Polyethylene glycol	33	0.29	143	[85]
Capric acid	31.5	0.149	153	[86]
Adonitol	101.6	-	247	[87]
Acetic acid	16.45	0.26	170	[88]
Erythritol	117.7	0.326	339.8	[88]
Benzamide acid	125.65	0.397	175.2	[89]
Palmitic acid	58–62	0.21	222	[69]
Xylitol	93	-	240	[90]
D-sorbitol	97.6	-	153	[91,92]
D-Iditol	80	-	169.9	[93]
L-Iditol	80	-	169.9	[94]
Methyl stearate	39.04	-	179.1	[95]
Glycerol	18.8	-	159.6	[96]
D-threitol	90	-	225	[97]
D-arabitol	102.9	-	230.1	[98]
Pentaerythritol	261	-	300	[99]
L-arabitol	100.9	0.93	283.9	[100,101]
d-mannitol	167.6	1.32	296.3	[101,102]
D-dulcitol	187.3	-	296	[102]
Inositol	225.5	-	351.6	[103]
Hexadecane	18.1	-	236	[104]
Formic acid	8	0.3	277	[105,106]
Oxalic acid	105	-	356	[107,108]
Adipic acid	152	-	275	[109]
1-Tetradecanol	37	0.252	227.8	[110]
n-Eicosane	33	0.372	152.7	[111]

Table 3
Impact of nanoscale particle incorporation on the LH and TC of organic PCMs.

Base PCM/ nano-additive	Additive loading	Method	TC(W/mK)	LH (kJ/kg)	Cycling stability	Ref.
paraffin/ Graphene	38.46 wt% (20 vol%)	TC: Transient Plane Source; LH: DSC	0.25 to 7.25 (Solid)	278.20 to 314.53	-	112
paraffin/ MWCNT	37.37 wt% (20 vol%)	TC: Transient Plane Source; LH: DSC	0.25 to 2.33 (Solid)	278.20 to 289.50	-	112
paraffin/Al	43.41 wt% (20 vol%)	TC: Transient Plane Source; LH: DSC	0.25 to 0.79 (Solid)	278.20 to 282.50	-	112
paraffin/ TiO_2	54.58 wt% (20 vol%)	TC: Transient Plane Source; LH: DSC	0.25 to 0.44 (Solid)	278.20 to 283.09	-	112
Paraffin wax/ Al_2O_3	4 wt% (1 vol%)	TC: modified transient plane source; LH: DSC	0.277 to 0.307 at 23 °C, 0.132 to 0.146 at 35 °C, 0.149 to 0.167 at 50 °C	215.92 to 207.43	TC remained stable for 10 cycles, then declined to near-pure paraffin levels by ~45 cycles	66
Paraffin wax/ Al_2O_3	11 wt% (3 vol%)	TC: modified transient plane source (MTPS); LH: DSC	0.277 to 0.338 at 23 °C, 0.132 to 0.157 at 35 °C, 0.149 to 0.179 at 50 °C	215.92 to 170.15	-	66
OM37 paraffin/ Al_2O_3	1 wt%	TC: TEMPOS thermal property analyzer; LH: DSC	0.16 to 0.30 at 20 °C (Solid), 0.14 to 0.17 at 65 °C (liquid)	185 ± 15 to 182 ± 15	No significant degradation or property variation over 10 cycles	113
OM37 paraffin/ CuO	1 wt%	TC: TEMPOS thermal property analyzer; LH: DSC	0.16 to 0.32 at 20 °C (solid), 0.14 to 0.23 at 65 °C (liquid)	185 ± 15 to 141 ± 15	No significant degradation/ property variation over 10 cycles	113
OM37 paraffin/ Cu	1 wt%	TC: TEMPOS thermal property analyzer; LH: DSC	0.16 to 0.36 at 20 °C (solid), 0.14 to 0.29 at 65 °C (liquid)	185 ± 15 to 157 ± 15	No significant degradation/ property variation over 10 cycles	113
Paraffin wax/ CQDs	1 wt%, 5 wt%, 10 wt%, 15 wt%, 20 wt%	LH: DSC; thermal stability: TGA	-	1 wt%: 145.35 to 240.00; 5 wt%: 145.35 to 204.11; 10 wt%: 145.35 to 194.52, 15 wt%; 145.35 to 174.27; 20 wt%: 145.35 to 160.53	LH retained 99.48% after 300 cycles (1 wt%)	114

[75]. Recent advances in improving the TC of organic PCMs through the incorporation of nanoparticles are summarized in Table 3. According to the reported results, the synthesized NePCMs show higher thermal conductivity than their corresponding base PCMs. NePCMs prepared with optimal loadings of carbon-based nanoparticles have shown greater TC enhancement than those incorporating inorganic nanoparticles. Increasing the concentration of nanoparticles can enhance the TC of the PCM, but it can also increase its viscosity, which can adversely affect natural-convection heat transfer; thus, it is crucial to select an optimal concentration of nano-additives to balance TC enhancement and viscosity [124]. In addition, nanoparticle settling and agglomeration have been observed after repeated thermal charging and discharging cycles of NePCMs, which can reduce the thermal performance of the TES system [120,125].

2.1.3. *Melting temperature*

Selecting a suitable PCM for a specific TES system requires that the melting temperature (T_m) of the PCM fall within the operational temperature range required for heating and cooling. It refers to the temperature at which the PCM begins to transform from the solid to the liquid state. Houshi et al. classified different PCMs into three distinct categories based on T_m [126]. These categories include low-, medium-, and high-temperature PCMs. The temperature ranges for different PCM categories are presented in Fig. 3. If a PCM has a T_m below 220 °C, it falls into the low-temperature category; however, if its T_m exceeds 420 °C, it belongs to the high-temperature category. In addition, if T_m falls between 220 and 420 °C, the PCM is classified as a medium-temperature material. According to Table 2, all listed organic PCMs fall into the low-temperature category, except pentaerythritol and inositol. Owing to their wide range of T_m, organic PCMs are considered suitable for a wide range of TES applications. Recent studies investigating the effect of nanoparticle addition on the T_m of organic PCMs are listed in Table 4. The reviewed studies indicate that the inclusion of nano-additives can cause slight changes in the melting and solidification temperatures of

Table 4
Influence of incorporation of nanoparticles on T_m.

Nano-enhanced PCM	Conclusion	Ref.
d-mannitol with Al_2O_3	The melting and solidification temperatures of d-mannitol-Al_2O_3 are tuned by ±3.5 °C by varying mass fraction and particle sizes of Al_2O_3.	127
Paraffin wax with Al_2O_3	The melting and solidification temperatures of paraffin wax increase following the incorporation of 2% Al_2O_3.	128
Paraffin wax with CQDs	Different composite PCMs are prepared by dispersing varying wt.% of CQDs into paraffin wax, and it is concluded that the T_m range of the synthesized composite PCMs is enlarged compared to pure paraffin wax, thus making them suitable for solar water and air heating systems. Moreover, the resulting melting points range is 61-64 °C for all synthesized composite PCMs.	114
Paraffin wax-Alumina (Al_2O_3)	The incorporation of 4 wt% Al_2O_3 nanoparticles into paraffin wax reduces its Tm by 0.17 °C.	66

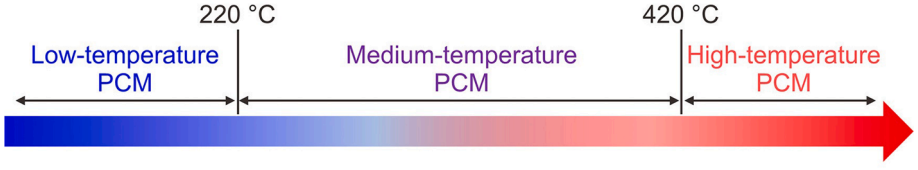


Fig. 3. Classification of PCMs based on T_m. Reproduced with permission from Ref. [71].

organic PCMs; thus, the selection of nano-additives should also consider the temperature requirements of a specific TES application.

2.1.4. Supercooling

A TES system requires efficient storage and release of LH through phase transitions; however, some PCMs exhibit supercooling, which delays LH release and therefore adversely affects the thermal performance of the TES system [129]. Supercooling refers to a condition in which a PCM remains in the liquid state at a temperature below its solidification temperature. Factors that can significantly influence supercooling include: (1) Percentage of impurities (2) Surface roughness of container (3) Cooling rate (4) Addition of nucleating agents (5) PCM liquid volume (6) Repeated thermal cycling of PCMs (7) Mechanical shocks (8) TC [130]. The incorporation of nanoparticles into organic PCMs has been widely investigated as a strategy for mitigating supercooling. Recent studies investigating the reduction of supercooling through nanoparticle incorporation are summarized in Table 5.

2.1.5. Thermal stability

For TES systems to operate efficiently over the long term, it is vital that PCMs retain their thermophysical properties after repeated thermal cycling [133]. Thermal cycling stability refers to a PCM's ability to retain its thermophysical properties during repeated cycles of energy storage and release. Several factors can significantly influence the thermal stability and cyclic performance of PCMs, including (1) encapsulation (2) physical and chemical stability of PCMs (3) thermal cycling conditions (4) incorporation of additives (5) environmental exposure [72]. Emeema et al. [114] studied the effect of incorporating CQDs nanoparticles on the thermal stability of paraffin wax. The results showed that the onset temperature of thermal degradation increased linearly with increasing CQD loading; thus, the composite PCMs exhibited greater resistance to thermal decomposition than pure paraffin. In addition, thermal cycling tests revealed that the composite PCM retained its LH, T_m, and degradation temperature more effectively over 300 cycles than pure paraffin. Kalidasan et al. [133] synthesized various composite PCMs by varying the amount of Ag nanoparticles dispersed in paraffin wax. The findings showed that the composite PCMs prepared by adding 0.2-0.8 wt% Ag nanoparticles exhibited higher degradation temperatures than pure paraffin. Subsequent thermal cycling stability tests showed that the composite PCM effectively retained its LH after 500 cycles and exhibited an increase in degradation temperature after cycling. These studies indicate that optimal nanoparticle dispersion can improve the thermal stability and cycling performance of paraffin-based PCMs by helping them retain their thermophysical properties during repeated thermal cycling.

2.2. Encapsulation of PCMs

PCMs possess high LH, which makes them promising materials for energy storage systems. Nonetheless, their application in TES systems is greatly affected by challenges such as low TC, leakage, volume expansion during phase transitions, and the possibility of reactions with surrounding materials. Encapsulation helps to address these challenges by enclosing a PCM within a shell [72,134]. The shell provides a strong and flexible protective barrier for the core material against environmental influences. Typically, shell materials are made of metals, ceramics, and polymers. According to the capsule's diameter, encapsulation methods are divided into three categories: macro-, micro-, and nanoencapsulation [135]. Containers are available in various shapes, including rectangular, spherical, cubic, tubular, and cylindrical configurations, and the selection of a suitable shape depends on the intended application. Different container geometries can result in different thermal performance characteristics. To enable a direct comparison of these approaches, their sizes, advantages, disadvantages, and typical applications are summarized in Table 6. This section summarizes recent advances in PCM encapsulation techniques aimed at improving the thermal performance of TES systems.

Table 5
Recent advances to mitigate the supercooling through nanoparticles addition.

Nano-enhanced PCM	Conclusion	Ref.
RT54HC with boron nitride	The degree of supercooling is significantly reduced by more than 50% via dispersion of 2 wt% of cubic boron nitride particles into the RT54HC.	131
d-mannitol with Al_2O_3	The findings revealed that supercooling was reduced by 25.2% through the incorporation of Al_2O_3 nanoparticles into d-mannitol. The mass fraction and particle size of Al_2O_3 nanoparticles were identified as key factors in reducing supercooling.	127
PEG2000 with EG-Co_3O_4	The subcooling of PEG2000/EG-Co_3O_4 was reduced from 15.7 °C to 9.6 °C.	132

2.2.1. Macro-encapsulation

Macro-encapsulation is a well-established and widely implemented technique in various TES systems. Macrocapsules are typically manufactured from low- and high-density polyethylene, polypropylene, tin-plated metal, and mild steel. The capsules are readily available in the markets and typically have diameter greater than 1 mm. This technique requires careful selection of the shell material, shape, and size to improve heat-transfer performance [136]. High specific surface area is achieved by macro-encapsulation which significantly improves the heat transfer; however, sufficient space must be provided within the capsules to accommodate volume expansion during phase transitions.

Researchers have widely applied this technique in various TES systems to improve heat transfer and prevent PCM leakage. Ajay et al. [137] constructed a novel TES system by macro-encapsulating a bio-derived organic PCM containing expanded graphite in a multipass tube arrangement for domestic water heating. The results showed that heat-transfer rates during both charging and discharging were significantly improved, with 90% of the stored thermal energy released within 90 min. Ali Reza et al. [138] performed numerical simulations to investigate the influence of encapsulating n-octadecane in fountain-shaped macrocapsule on heat transfer. The findings showed that the proposed TES design reduced the PCM charging time by 43% compared with the pear-shaped configuration. Achmad et al. [139] designed a novel macro-encapsulation strategy in which the PEG-Fe_3O_4

Table 6
Comparison of macro, micro, and nano-encapsulation of PCMs.

Type	Typical size	Advantages	Disadvantages	Applications
Macro	>1 mm	Simple and relatively low-cost; easy handling; high PCM capacity; suitable for large-scale TES	Lower surface-area-to-volume ratio and slower heat transfer; bulky; leakage possible if container is damaged	Large TES units, solar thermal storage, building panels
Micro	1 μm–1 mm	Higher surface area and improved heat transfer; good leakage protection; readily incorporated into other materials	Complex and costly fabrication; shell reduces effective PCM loading; shell degradation	Buildings, coatings, and textiles
Nano	<1 μm	Very high surface-area-to-volume ratio; rapid heat transfer; suitable for compact systems; improved PCM confinement	Complex and relatively expensive synthesis; scale-up, agglomeration, and long-term stability remain challenges	Nanofluids, electronics cooling, smart coatings, and compact TES

composite was encapsulated within copper tubes, and their study concluded that macro-encapsulation of PEG-Fe_3O_4 in copper tubes increased the thermal energy storage capacity of concrete.

Abid et al. [140] investigated three macro-encapsulation geometries, namely, circular, hexagonal, and square, to improve the thermal and electrical performance of battery thermal management systems (BTMS) incorporating PCMs. Their findings demonstrated that the hexagonal-shaped configuration significantly enhanced the thermal performance of the BTMS by increasing the heat-transfer rate between the PCM and the battery compared to other geometries.

2.2.2. Micro-encapsulation

Owing to their higher surface-area-to-volume ratio, micro-encapsulated PCMs can provide higher heat-transfer rates in energy storage applications than macroencapsulated PCMs. In addition, microencapsulation helps prevent PCM leakage and direct contact with the surrounding environment [141,142]. Microcapsules enclose a PCM core and typically range from 1 μm to 1 mm in diameter. Baolian et al. [143] employed a chemical precipitation technique to micro-encapsulate paraffin using SiO_2 as the shell material, and the results indicated that the addition of 10% micro-encapsulated PCM to emulsion paints considerably increased the heat conservation time of film by 19.44%. The effects of incorporating microencapsulated PEG-1000 PCM on the thermal and mechanical properties of lightweight concrete (LWC) were investigated by Davood et al. [144]. The study demonstrated that the addition of microencapsulated PEG-1000 improved the compressive strength, decreased the TC of the LWC mixture, thereby improving its thermal insulation performance, and considerably reduced its water absorption. Yuanchao et al. [145] synthesized micro-encapsulated capsules using a single-step-in-situ polymerization process. The microcapsules consisted of an n-octadecane core and a silicone shell, and the influence of the shell-to-core ratio on the properties and morphology of the microencapsulated material was investigated. Their findings showed that the organic silicon micro-encapsulated phase change materials (OS-MPCMs) exhibited favorable thermophysical and mechanical properties, indicating their potential suitability for TES applications such as thermoregulatory textiles and thermal energy storage in buildings.

2.2.3. Nano-encapsulation

Nanocapsules typically have diameters of less than 1 μm; thus, they provide a higher surface-area-to-volume ratio than larger capsules. Nano-encapsulation has considerable potential to reduce PCM leakage, enhance TC, and improve the mechanical properties of the resulting composite. Owing to its numerous benefits, nano-encapsulation has attracted considerable global attention. Soumen et al. [146] used a sol-gel technique to synthesize silica-shelled n-octadecane nanocapsules and investigate their thermophysical properties. Silica was used as the shell material to encapsulate n-octadecane, and a shell-to-core ratio of 3:1 was identified as optimal under the investigated conditions. The characterization results indicated that the nano-capsules exhibited good thermal cycling stability, with only a slight change in enthalpy after 300 cycles, and exhibited latent heats of storage and release of 186.82 and 183.14 J/g, respectively. Furthermore, the incorporation of the synthesized nano-capsules into cement-based materials improved the mechanical properties of the resulting concrete. Ghufran et al. [147] synthesized 15 paraffin-SiO_2 nanocapsule formulations with different core-to-shell ratios to investigate their thermal performance. Their findings showed that the nanocapsules exhibited good thermal cycling stability, with their measured thermal properties remaining approximately unchanged after 100 thermal cycles. The reported encapsulation ratio was 81.8%, while the latent heats of storage and release were 115.07 and 114.19 J/g, respectively.

Overall, reducing capsule size increases the surface-area-to-volume-ratio and generally improves heat transfer performance, however fabrication complexity, cost, and scale-up challenges become more significant for micro- and nano-encapsulation.

3. Ionic liquid-based phase change materials

PCMs are pivotal to thermal energy storage and management, with wide-ranging applications in solar energy utilization, building temperature regulation, and electronic device cooling [5,9,14,15]. However, traditional organic PCMs (e.g., paraffins, fatty acids) suffer from severe disadvantages such as unstable physico-chemical properties, while phase segregation and sub cooling for the inorganic ones (e.g., salt hydrates) [19–22]. To address these inherent limitations, ILs and their analogues have recently emerged as novel and highly designable class of PCMs [24,29,33]. This section focuses on reviewing four core aspects of these advanced PCMs: (1) the intrinsic advantages of ILs for TES applications; (2) the development of FILs to enhance key performance metrics (e.g., TC, cyclic stability); (3) the promising potential of DESs as cost-effective, eco-friendly alternatives to both conventional PCMs and neat ILs; and (4) the latest progress of composite IL-based PCMs.

3.1. Advantages of ILs as PCMs

Structures of common cations and anions of ILs are depicted in Fig. 4. Owing to their unique ionic structure and molecular designability, ILs presented distinct advantages as PCMs, which can be summarized as follows.

(a) **High thermal stability.** The decomposition temperatures of ILs are generally comparable to those of paraffin waxes, although some ILs exhibit thermal stability at temperatures exceeding 200 °C [148,149].
(b) **Nonvolatility and Low-flammability.** The ionic interactions endow ILs with negligible saturated vapor pressure, eliminating the volatilization loss common in organic PCMs; meanwhile, their inherent low-flammability and resistance to phase separation greatly enhance operational safety and long-term service life.
(c) **Broad liquidus range.** Through rational selection of cation alkyl chain lengths and anion types, the phase change temperature of IL-based PCMs can be tailored from −50 to 300 °C, making them suitable for diverse applications ranging from low-temperature cold chain storage to medium-to-high-temperature industrial waste heat recovery [150,151].
(d) **Promising LH.** Introducing dicationic structures or hydrogen bonding functional groups (e.g., hydroxyl and amide groups) strengthens intermolecular interactions, enabling ILs to achieve LH values of 150~220 kJ/kg, comparable to those of high-performance paraffin-based PCMs [29,34,152].
(e) **Excellent chemical stability.** Many ILs exhibit high chemical stability, which ensures good compatibility with various substrates, including metals and polymers, thereby supporting the development of stable composite PCMs.

The feasibility of ILs as PCMs was first explored by Zhu et al., in 2009 through systematic measurements [27]. They characterized the T_m and LH for 10 imidazolium-based ILs, identifying [C_{16}MIM][Br] and [C_{16}MMIM][Br] as promising PCM candidates. Specifically, [C_{16}MIM][Br] exhibited a moderate melting temperature of 63.91 °C and a high LH of 152.56 kJ/kg, while [C_{16}MMIM][Br] had a melting temperature of 98.55 °C and an LH of 126.16 kJ/kg. These T_m fall within the optimal range for TES scenarios (e.g., industrial waste heat recovery and building temperature regulation), and their LH values are comparable to those of commercial paraffin-based PCMs. Notably, the decomposition temperatures of [C_{16}MIM][Br] and [C_{16}MMIM][Br] were approximately 230 and 250 °C, respectively, significantly higher than their T_m, conforming their high resistance to thermal degradation. Additionally, the heat capacities of these ILs were found to outperform that of commercial heat transfer fluid Therminol® VP-1 [26,35,153].

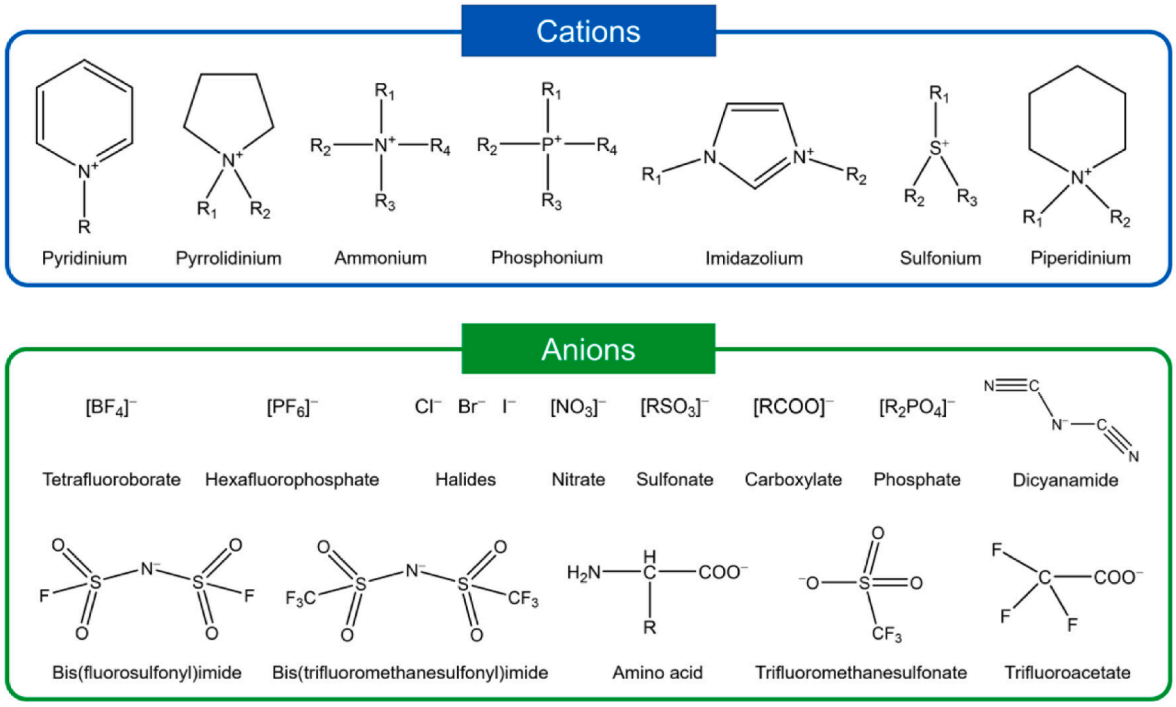


Fig. 4. Structures of common cations and anions of ILs. Reproduced with permission from Ref. [24].

Similarly, Matuszek et al. [26] reported 8 pyrazolium-based ILs, all solid at room temperature, with melting points ranging from 91 to 168 °C and LH values between 40 and 160 kJ/kg. Among these, [Pzy][$MeSO_3$] stood out with a high LH of 160 kJ/kg alongside a high melting point (168 °C), and its excellent thermal stability was further validated through 130 consecutive thermal cycles within the temperature range of 120-175 °C. Very recently, the same group reported the thermophysical properties of 10 pyridinium ILs as PCMs for intermediate-temperature TES, and this spectrum of ILs were also experimentally proven to be promising candidates as PCMs [34]. Table 7 presents a summary of the thermal properties of certain potential ILs documented in existing literature.

These experimental findings collectively confirm that ILs hold substantial promise as PCMs for TES systems. However, in terms of LH

Table 7
Information of reported ionic liquids in literature.

ILs	Abbreviation	T_m (°C)	ΔH (kJ/kg)	Ref.
1-ethyl-3-methylimidazolium dicyanamide	[EMIM][DCA]	−46.82	82.4	33
1-akyl-3-methylimidazolium bromide	[C_{10}MIM][Br]	74.43	66.789	33
1-hexadecyl-3-methylimidazolium bromide	[C_{16}MIM][Br]	63.91,65.4	152.563,158.6	33
1-butyl-2,3-dimethylimidazolium bromide	[C_4MMIM][Br]	76.51	66.977	33
1-akyl-2,3-dimethylimidazolium bromide	[C_{10}MMIM][Br]	68.2	75.395	33
1-hexadecyl-2,3-dimethylimidazolium bromide	[C_{16}MMIM][Br]	98.55	126.616	33
1-butyl-3methylimidazolium chloride	[C_4MIM][Cl]	53.42	59.004	33
1-butyl-2,3-dimethylimidazolium chloride	[C_4MMIM][Cl]	92.74	76.384	33
1,3-dimethylimidazolium tosylate	[C_1MIM][TOS]	94.4	170.3	33
cetyltrimethylammonium bromide	CTAB	243	151.6	33
guanidinium methylsulphonate	[Gdm][$MeSO_3$]	208	190	37
guanidinium benzenesulfonate	[Gdm][$C_6H_5SO_3$]	210	138	37
guanidinium hydrogensulfate	[Gdm][HSO_4]	100	133	37
guanidinium triflate	[Gdm][TfO]	160	130	37
guanidinium tosylate	[Gdm][TOS]	227	91	37
guanidinium chloride	[Gdm][Cl]	188	150	37
guanidinium bromide	[Gdm][Br]	190	107	37
guanidinium nitrate	[Gdm][NO_3]	217	174	37
guanidinium tetrafluoroborate	[Gdm][BF_4]	173	103	37
1,2-bis(3-ethylimidazolium-1-yl) ethane bromide	[C_2(eim)$_2$][Br_2]	184.21	125.17	154
1,4-bis(3-methylimidazolium-1-yl) butane bromide	[C_4(mim)$_2$][Br_2]	115.45	115.69	154
1,4-bis(3-ethylimidazolium-1-yl) butane bromide	[C_4(eim)$_2$][Br_2]	131.34	102.13	154
1,4-bis(3-vinylimidazolium-1-yl) butane bromide	[C_4(C_2H_4im)$_2$][Br_2]	124.86	159.35	154
1-hexadecyl-3-methylimidazoliumchloride	[C_{16}MIM][Cl]	63.44	159	154
pyrazolium methylsulphonate	[Pzy][$MeSO_3$]	168	160	26
pyrazolium benzenesulfonate	[Pzy][$C_6H_5SO_3$]	137	105	26
pyrazolium ethylsulphonate	[Pzy][$EtSO_3$]	113	108	26
pyrazolium hydrogensulfate	[Pzy][HSO_4]	122	115	26
pyridinium benzenesulfonate	[PyH][$C_6H_5SO_3$]	135	123	34
3-methylpyridine methylsulphonate	[3-MepyH][$MeSO_3$]	99	136	34
2-hydroxypyridine methylsulphonate	[2-OHpyH][$MeSO_3$]	160	190	34
2-aminopyridine methylsulphonate	[2-NH_2pyH][$MeSO_3$]	125	111	34

storage capacity, current IL candidates still lack distinct advantages compared with high-performance conventional organic PCMs (e.g., long-chain paraffins). The practical operating-temperature window of an IL-based PCM is determined by its melting and solidification behavior, as well as its thermal stability under the intended operating conditions. Effective charging requires complete melting of the PCM, whereas discharging involves cooling to a temperature that enables solidification. To ensure reliable performance over repeated cycles, the maximum operating temperature must remain below the temperature at which significant material degradation begins. Consequently, a high decomposition temperature does not necessarily indicate a correspondingly wide operating-temperature range for LH storage.

3.2. Functionalized ILs

Aiming to develop PCMs with higher gravimetric TES densities and superior thermophysical properties, researchers have identified that functionalizing ILs by introducing specific functional groups (e.g., hydroxyl, amino and phenyl) is a promising way to improve their TES performance. For instance, Matuszek et al. [26] developed hydroxyl-functionalized ILs ([2-OHpyH][$MeSO_3$]) as PCMs, which was experimentally proved to exhibit a substantially higher LH (190 vs. 77 kJ/kg) than the benchmark conventional IL [pyH][$MeSO_3$], with only a slight shift in T_m (180 vs. 160 °C). Additionally, these ILs showed excellent thermal stability with a decomposition temperature of 225 °C. Shi and his co-workers [32] theoretically demonstrated that introducing the hydroxyl group (-OH) into [MPy][TfO] and the methoxy group (-OCH_3) into [MIm][NO_3] were highly beneficial for increasing the thermal power of the parent convention ILs. Zhang et al. [29] synthesized a series of imidazolium-based ILs as novel PCMs by grafting functional groups (such as the ester, hydroxyl and carboxyl groups) onto the imidazolium cation, and these modifications effectively improved the ILs' TES performance, i.e., LH and heat capacity. Collectively, these findings confirm that performing functionalization on well-documented ILs could be a promising solution to enhance their thermophysical properties.

With respect to the underlying reasons, hydrogen bonding is hypothesized to play a key role [30,36,154]. Through targeted functional modification, introducing specific functional groups (e.g., –OH and –COOH) onto the side chains of alkyl-chain ILs enhances both the number and strength of internal hydrogen bonds. Notably, the modified ILs possess 2 – 3 times internal hydrogen bonds than their structurally analogous alkyl-chain counterparts, accompanied by a more stable hydrogen-bonded network. During phase transition, the energy consumed in breaking these hydrogen bonds directly contributes to the enhanced thermophysical properties and TES density of the ILs.

This feature could also provide a theoretical basis for molecular design, thus guiding the development of high-performance PCMs by tailoring the number of hydrogen bond donor and acceptor sites. Vijayraghavan et al. [155] synthesized a set of protic ILs using guanidinium as the cation, owing to its multiple hydrogen bonding sites. Experimental results demonstrated that these protic ILs exhibited favorable thermal stability and promising phase change behaviors. Among them, [Gdm][$MeSO_3$] was identified as an attractive candidate for intermediate temperature TES with a LH of 190 kJ/kg at 208 °C. Moreover, recent study confirmed that [Gdm][$MeSO_3$] maintains thermal stability up to 225 °C, with its T_m and LH remain unchanged after 400 thermal cycles, rending it suitable for ide-temperature-range applications [156]. Zhang et al. [154] developed a series of dication imidazolium-based ionic PCMs, of which the heat capacities and LH were significantly enhanced, leading to remarkably higher TES densities compared to most traditional monocationic ILs. Meanwhile, quantum chemical calculations indicated that dicationic ILs form complex hydrogen-bonding networks, contributing to their superior thermal performances.

Despite the progress made, research on the application of functionalized ILs as phase change energy storage materials remains at an early stage. Specifically, systematic and in-depth investigations into their solid-liquid phase transition processes, underlying mechanisms and the effects of PCM structures on thermophysical properties are still insufficient. Moving forward, integrating experimental characterization with theoretical analysis will be essential to elucidate the phase change energy storage mechanism and identify key factors regulating the TES performance of functionalized ILs, thereby facilitating the development of advanced IL-based PCMs.

3.3. IL-based composite PCMs

IL-based composite phase change materials (CPCMs) represent an emerging class of key materials for thermal energy storage and management systems. These materials employ ionic liquids as the phase change matrix, and are further engineered with functional additives such as porous scaffolds, nanoparticles, or polymer matrices [157–160]. By integrating the intrinsic physicochemical properties of ILs with the advantages of composite configurations (e.g., enhanced TC, leakage resistance, and cycling durability), IL-based CPCMs enable effective regulation of phase transition temperature and LH storage capacity. Such capabilities allow them to meet TES demands across a broad temperature range, thus positioning them as a prominent research focus in the field of advanced energy materials.

For instance, Nourizadeh et al. [161] developed a series of innovative, eco-friendly nano-enhanced CPCMs, consisting of the IL 2-hydroxyethylammonium stearate, d-mannitol and various nanoparticles (ZnO, ZrO_2, SiO_2 and HAP). In contrast to the pure IL, the LH of the resultant CPCMs increased to nearly twice the origin value (307.41 vs. 171.12 kJ/kg). Moreover, the incorporation of nanoparticles not only significantly improved the TES capacity and reduced the T_m of the CPCMs, but also retained excellent thermal stability. Lin et al. [162] fabricated a novel form-stable CPCM, where the IL 1-hexadecyl-3-methylimidazolium bromide served as the phase change component, with nano aluminum nitride and copolyimide 6/12 as the additive and supporting matrix, respectively. Experimental results demonstrated that this CPCM exhibited favorable LH, good thermal stability and reusability, along with a remarkable increase in TC of 165.2%. Zhang et al. [163] reported similar findings: by compounding ILs (1-butyl-3-methylimidazolium tosylate and 1,3-dimethylimidazolium tosylate) with tubular carbon fiber and stearic acid, the resulting composites displayed superior TC, satisfactory thermal stability, and outstanding mechanical stability.

Future studies should prioritize the systematic characterization of IL-based CPCMs, including TC, specific heat capacity, and phase transition behavior across a range of temperatures and operating conditions. Also, investigating the long-term stability and degradation of IL-based CPCMs under practical service conditions is of paramount importance for their industrial application.

3.4. DES as PCMs

DESs present physicochemical properties relevant to PCMs analogous to those of ILs. In addition, DESs offer cost competitiveness relative to ILs as PCMs. Specifically, DESs are prepared by simply mixing a hydrogen bond acceptor (HBA) with a hydrogen bond donor (HBD). As shown in Fig. 5, a large number of cheap compounds can be used as the HBA (e.g., ammonium and phosphonium salt) or HBD (e. g., organic alcohols and acids) to form DESs, making DES-based PCMs more affordable and sustainable [164].

Extensive prior studies have demonstrated that, from the perspective of TES density, the formed hydrogen bond network between HBA and HBD is conducive to LH enhancement, thereby improving energy storage performance [30,36,154]. Moreover, commonly used HBDs, e.g., polyols, organic carboxylic acids, and amides are known as conventional compounds exhibiting high phase change enthalpy (Table 8). In line

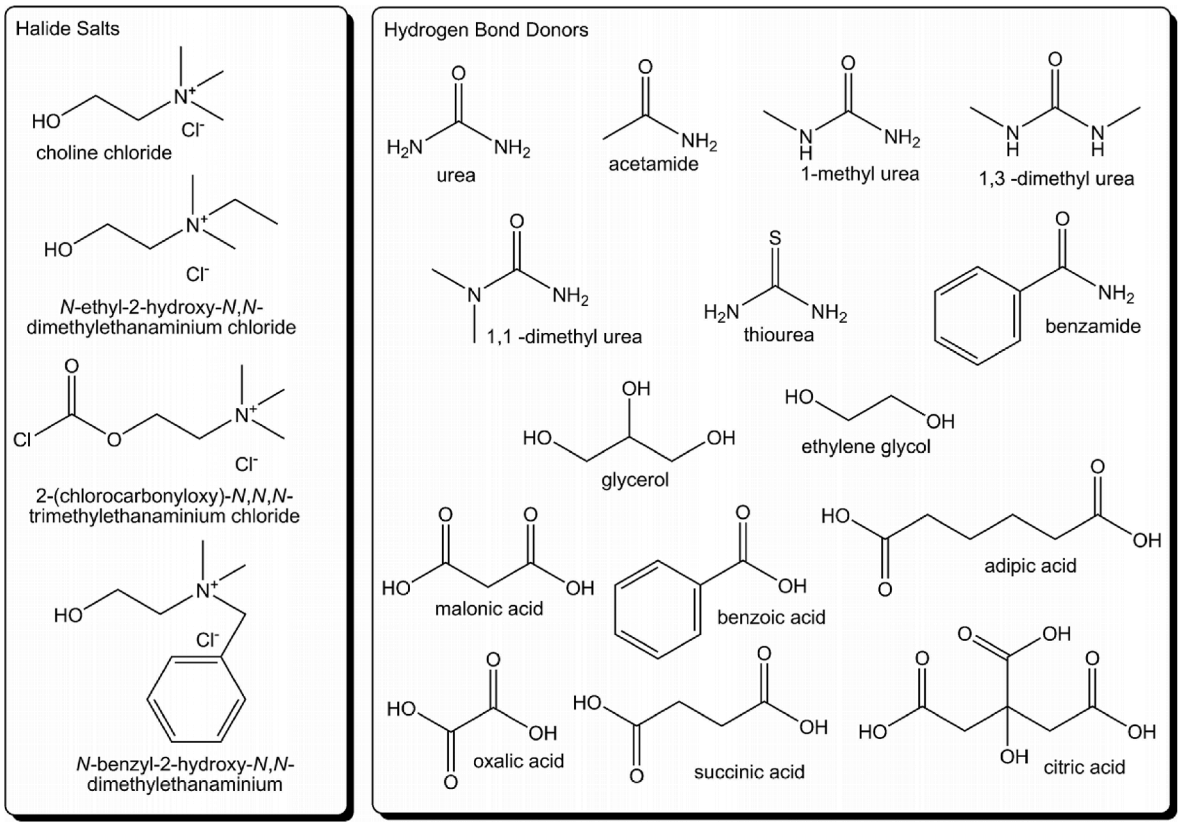


Fig. 5. Structure of typical HBAs and HBDs used to form deep eutectic solvents. Reproduced with permission from Ref. [164].

with the general principle governing the enthalpy of eutectic PCMs, employing such high-enthalpy compounds to form DESs is expected to yield systems with significantly enhanced gravimetric energy storage density [40,42,165].

With respect to T_m, DESs can achieve widely tunable range of phase transition temperatures (from −50 °C to over 180 °C) through the rational selection and combination of HBAs and HBDs. Specifically, this tunability is governed by a dual regulatory mechanism based on component selection and molar ratio adjustment, which allows precise control over phase transition behavior without modifying the molecular structure of the individual components [38,166]. As a result, this strategy offers notable advantages in terms of operational simplicity and low cost, making it highly suitable for the practical application of phase change energy storage materials. Furthermore, DES-based PCMs formulated at or near the eutectic composition may exhibit congruent melting and solidification behavior, which can reduce the tendency toward phase separation and improve thermal cycling stability; however, these characteristics remain dependent on composition and operating conditions.

Table 8
Phase change properties of common HBDs.

HBDs	Formula	T_m (°C)	ΔH (kJ/kg)
acetic acid	CH_3COOH	16.45	164
oxalic acid dihydrate	$C_2H_2O_4{\cdot}2H_2O$	101	398
malonic acid	$HOOCCH_2COOH$	135	224
amber acid	$HOOC(CH_2)_2COOH$	188	311
adipic acid	$HOOC(CH_2)_3COOH$	152	275
urea	$CO(NH_2)_2$	134	218
acetamide	CH_3CONH_2	82	233
propionamide	$CH_3CH_2CONH_2$	82	175
butyramide	$CH_3(CH_2)_2CONH_2$	117	212
decanamide	$CH_3(CH_2)_8CONH_2$	101	222
malonamide	$CH_2(CONH_2)_2$	162	229
hexanediamide	$(CH_2)_4(CONH_2)_2$	226	365
1,6-hexanediol	$HO(CH_2)_6OH$	44	194
1-decanol	$CH_3(CH_2)_9OH$	7	210
1-dodecanol	$CH_3(CH_2)_{11}OH$	24	216
inositol	$C_6H_6(OH)_6$	225.5	351.6
pentaerythritol	$C(CH_2OH)_4$	261	300
erythritol	$OHCH_2(CHOH)_2CH_2OH$	117.7	273
dipentaerythritol	$(CH_2OH)_3CCH_2OCH_2C(CH_2OH)_3$	220	263
d-mannitol	$OHCH_2(CHOH)_4CH_2OH$	167.6	296.3

From the above analysis, DESs satisfy the multifaceted requirements of TES performance and economic feasibility, rendering them promising candidates as PCMs. For the application, Wang et al. [167] developed a DES-based PCM composed of choline chloride (ChCl), urea, and nickel chloride hexahydrate, which was found to has high TC as well as excellent thermal stability, with a decomposition temperature of up to 215 °C. Shahbaz et al. [39] designed two ChCl-calcium chloride hexahydrate based DESs with suitable phase change temperature as well as reasonable LH, and excellent stability properties are tuned in their work, suggesting the high potential of DESs as PCMs. Peyrovedin et al. [42] synthesized 7 binary ChCl-based DESs, and most of these green DES-based PCMs present larger LH than the reference paraffin, as summarized in Table 9. Subsequently, they conducted a modified thermal cycling test for a solar thermal power plant system that employs a PCM storage tank to store solar energy during the day and releases it at night. By comparing the performance results of the studied DESs with those of the benchmark paraffin, it was found that some of the selected DESs outperformed the paraffin in terms of both energy and exergy analyses. Yang et al. [168] reported a myristic acid-palmitic acid-stearic acid (MA-PA-SA) ternary DES with a mass ratio of 52.2:29.4:18.4, and they subsequently synthesized the MA-PA-SA/expanded graphite (EG) composite PCM at a mass ratio of 13:1. Experimental results indicated that the composite DES-based PCM exhibited negligible supercooling, along with good thermal reliability and excellent stability. Our group

Table 9
Phase change behaviors of ChCl-based DESs [42].

DES	HBA	HBD	molar ratio	T_m (°C)	ΔH (kJ/kg)
DES1	ChCl	suberic acid	1:1	93	192.65
DES2	ChCl	Urea	1:0.9	80	216.89
DES3	ChCl	gallic acid	1:0.5	77	201.42
DES4	ChCl	4-hydroxybenzoic acid	1:0.5	87	219.22
DES5	ChCl	oxalic acid	1:0.8	73	187.42
DES6	ChCl	itaconic acid	1:1	57	175.13
DES7	ChCl	p-coumaric acid	1:0.5	67	190.11
paraffin	-	-	-	68	189

has recently developed a novel CTAB-urea DES-based PCM for application in solar water heating system targeting high-temperature water supply [40]. Experimental results indicated that the CTAB-urea DES combines the inherent merits of CTAB and urea, exhibiting a favorable melting point of 86.9 °C, a high LH of 174.9 kJ/kg, excellent shape stability, a low degree supercooling of about 3 °C as well as comparable TC. Moreover, this DES-based PCM demonstrated satisfactory thermal stability and reliability, which was confirmed by notably higher temperature of thermal decomposition over the operating condition and slight changes of phase change behaviors (i.e., T_m, LH) after 100 thermal cycles, validating its long-term service potential.

Currently, the research on DESs as PCMs is still in its infancy. To advance this field toward practical applications, two key challenges need to be addressed. Firstly, the current library of HBDs and HBAs remains relatively limited. Expanding this material library necessitates the systematic evaluation of critical thermophysical and chemical properties including melting/freezing points, supercooling degree, LH, TC, specific heat, decomposition temperature, volume expansion, cycling stability, and material corrosivity. Such a comprehensive assessment is essential for screening high-performance HBD/HBA pairs and developing advanced DES-based PCMs. Secondly, the fundamental mechanisms underlying energy storage behaviors and the evolution of thermophysical properties during phase transition remain incompletely elucidated. Specifically, the correlation between the hydrogen bond network structure (e.g., bond strength, coordination number) and energy storage capacity, as well as the dynamic changes in molecular interactions during melting/solidification, have not been fully clarified. Elucidating these intrinsic mechanisms is critical for developing robust and reliable theoretical models to predict the performance of DES-based PCMs, thereby guiding the rational design of high-performance systems.

4. Computer-aided IL design as PCM

Current research on IL-based TES materials has predominantly focused on the experimental characterization of their thermophysical properties. However, given the vast number of potential cation-anion combinations these experimental efforts have only covered a minute fraction of the entire IL chemical space [45]. The conventional experimental trial and error method is cost-expensive, time-consuming, and even unrealistic for this expansive chemical space to identify high-performance IL-based PCMs. In this regard, computational screening or rational design of ILs as PCMs based on reliable theoretical methods is of the essence. In recent years, the computer-aided ionic liquid design (CAILD) method has been widely used and extended as a reliable tool for tailoring IL molecular structures with pre-specified desirable properties [169–171]. Within the CAILD framework, reliable IL structure-property models are the prerequisite that computational IL screening or design can be realized. This section systematically reviews the key structural determinants governing the TES performance of IL-based PCMs, alongside state-of-the-art structure-property models and high-throughput screening methodologies for IL-based PCMs.

4.1. QSPR models

Structure-property models correlate the molecular structural features of materials with their physicochemical properties, enabling the prediction of material performance. These models have been increasingly applied in the field of ILs. In particular, by investigating the intrinsic microstructural properties of IL molecules and integrating experimental data, QSPR models have been developed to link the molecular structure with key properties [172–175]. To date, a substantial number of QSPR models have been reported in the literature covering a broad spectrum of ILs properties relevant to phase change behaviors, such as LH, TC, T_m, heat capacity etc.

4.1.1. T_m

T_m is one of the most important properties of IL-based PCMs, as it largely determines the temperature range over which they can be practically applied. Early QSPR studies focused mainly on bromide-based IL families and therefore captured only a limited range of cation–anion interactions [176–178]. Varnek et al. [179] extended the investigated chemical space and found that nonlinear methods, including ANN, SVM, and BPNN, generally performed better than linear approaches. Later group-contribution models incorporated a wider variety of IL structures while maintaining relatively low prediction errors, and advances in machine learning and deep learning enabled predictive modeling using datasets containing thousands of ILs [52,180–185]. Similar ML approaches have also been applied to DES melting-temperature prediction [186]. Notably, Carrera et al. [187] experimentally validated a QSPR-guided molecular-design approach by synthesizing six new guanidinium ILs, for which the measured T_m values showed good agreement with model predictions. A comparative analysis of various modelling approaches is summarized in Table 10.

4.1.2. LH

Compared with T_m, considerably fewer models have been developed for the LH of ILs. Zhu et al. [27] developed a QSPR correlation for 44 ILs and identified cation–anion interaction energy as an important descriptor, obtaining $R^2 = 0.9047$. Bai et al. [35] further showed that model performance depends strongly on the structural consistency of the dataset, with better prediction obtained for a more homogeneous imidazolium-halide subset. In contrast, the group-contribution model of Jose et al. [188] showed an AARD of 15.9%, which was attributed to limited and uncertain experimental data and the inability of a linear model to fully represent complex phase-transition behavior. More recently, Saliha et al. [189] developed an ML model for 42 protic organic salts, achieving $R^2 = 0.82$ and an SEE of 4 kJ/mol. These studies indicate that the limited availability and quality of LH data remain major constraints on model development.

4.1.3. TC

Data-driven approaches have also been applied to TC prediction. Chen et al. [184] developed the ILTransR model using 73 ILs and 454 thermal-conductivity data points, achieving an MAE of 0.0061. Cao et al. [190] combined group-contribution descriptors with several ML algorithms and found GC-XGB to provide the best performance, with $R^2 = 0.808$ and RMSE = 0.01 using an IL-based data-splitting strategy. Qiu et al. [185] subsequently applied ILBERT, obtaining $R^2 = 0.879$ and an MAE of 0.007 W/mK, followed by virtual screening of approximately eight million possible IL combinations. Nevertheless, further development and broader applicability of these models are hindered by the limited availability of high-quality experimental TC data, emphasizing the importance of stronger integration between computational modeling and experimental validation.

Despite being a powerful and reliable tool for predicting the thermophysical behavior of ILs, the utility of QSPR models in rational design and high-throughput screening is severely limited by the lack of experimental data on these thermal properties. It is vital to

Table 10
Comparison of computational models for predicting Tm, ΔH, and TC.

Property	Model	Data size	Validation method	Prediction performance	Ref.
T_m	QSPR	126 pyridinium bromides, T_m range 30–200 °C	Three-subset cross-validation (A + B→C, A + C→B, B + C→A)	6-descriptor model: $R^2 = 0.7883$, $R^2_{cv} = 0.7624$, s = 23.0 K. External subset predictions: $R^2 = 0.7570$–0.7785, prediction error = 24.05–27.10 K	177
T_m	QSPR–GFA	Set B: 75 tetra alkyl-ammonium bromides; Set C: 34 (n-hydroxyalkyl)-trialkyl-ammonium bromides; 126 pyridinium bromides (Set A) used for method validation	Leave-one-out validation	Set A, B, C: $R^2 = 0.790$, 0.775, 0.716 respectively	178
T_m	QSPR/ML models	717 bromides of N-containing organic cations: 126 pyridinium, 384 imidazolium/benzimidazolium, 207 quaternary ammonium	5-fold external cross-validation	RMSE = 37.5–46.4 °C (FULL); 26.2–34.8 °C (PYR); 38.8–45.9 °C (IMZ); 34.2–49.3 °C (QUAT)	179
T_m	CPG neural-network QSPR	101 guanidinium salts; T_m 46–322 °C	Independent test set, 5-fold cross-validation, y-randomization, and experimental testing of 6 salts	Test: $R^2 = 0.815$, RMSE = 23.95 °C, MAE = 20.33 °C; 5-fold CV: $R^2 = 0.742$	187
T_m	GC model	190 ILs	Model developed on 155 ILs and tested on 35 ILs	ARD = 5.86%, RMSD = 28.20 K, $R^2 = 0.8984$; external 35 ILs: ARD = 5.16%	180
T_m	GC model	799 ILs	Independent test set of 201 ILs	R [2] = 0.817, RMSE = 24.86 K	181
T_m	GC model	293 data points for 136 ILs	Assessed against literature values	ARD = 7.8%; AAD = 22.6 K	182
T_m	GA-optimized GC model	400 ILs; T_m = 194–567 K	Random 200/200 split into correlation and independent prediction sets	Correlation AARD = 7.97%; validation AARD = 6.16%; total AARD = 7.07%, $R^2 = 0.8841$	183
T_m	ML models	2212 ILs	Random 67:33 train/test split; 5-fold CV ×3; 500 y-randomization tests; external validation on 19 ILs	Best test model (GBM): $R^2 = 0.67$, RMSE = 45 °C, MAE = 33 °C	52
T_m	Transformer–CNN	2212 ILs; pre-trained on 9,434,070 IL-like molecules	10-fold cross-validation with IL-based dataset splitting	Test MAE = 11.15 K	184
T_m	Transformer–CNN	2673 ILs; pre-trained on ~31 million IL-like molecules	IL-based 5-fold CV, repeated 5 times	MAE = 26.03 ± 0.17 K; RMSE = 35.04 ± 0.25 K; $R^2 = 0.782 \pm 0.003$	185
T_m	ML	1648 data points	80:20 mixture-out train/validation split; 5-fold CV for model tuning	SVR: $R^2 \approx 0.85$, RMSE ≈ 30–31 K, MAPE ≈ 5.57%	186
ΔH	QSPR	44 ILs	10 ILs randomly selected as prediction set; remaining 34 used for training	R [2] = 0.9047; SD = 4.797 kJ/mol	27
ΔH	MLR-QSPR	40 ILs: 30 training + 10 external test. Subsets: A-all ILs; B-22 imidazolium; C-10 halide; D-9 imidazolium-halide ILs	Independent external test sets	Test MRE: A = 31.48%; B = 19.79%; C = 22.05%; D = 13.86%	35
ΔH	GC model	28 experimental ΔH from NIST-ILs	Model fitted using NIST-ILs data; no independent external validation reported for ΔH	AARD = 15.9%; maximum deviation = 34.7%	188
ΔH	ML	69 salts	Experimental external validation using 7 newly synthesized salts	Best statistical model MLREM_HF42: $R^2 = 0.82$, SEE = 4 kJ/mol	189
λ	Transformer–CNN	454 data points, 73 ILs	10-fold cross-validation with IL-based dataset splitting	Test MAE = 0.0061 W m^{-1} K^{-1}	184
λ	Transformer–CNN	606 data points, 44 ILs	IL-based 5-fold CV, repeated 5 times	MAE = 0.0021 ± 0.0001; RMSE = 0.0029 ± 0.0001 W m^{-1} K^{-1}; $R^2 = 0.9880 \pm 0.0010$	185
λ	GC-XGB	334 data points, 46 ILs; 273.15–390 K	IL-based 10-fold testing; 5-fold CV for hyperparameter tuning; modeling repeated 5 times	Test $R^2 = 0.808$; RMSE = 0.0100 W m^{-1} K^{-1}	190

systematically integrate experimental characterization with theoretical analysis to identify and understand the key factors that affect the thermophysical properties of ILs, thereby accelerating the rational design of high-performance IL-based PCMs. In addition, model performance should not be assessed solely on the basis of prediction error, as dataset size and structural diversity, validation strategy, applicability domain and uncertainty estimation also determine the reliability of predictions. Models evaluated using independent test sets or cross-validation provide stronger evidence of generalization than models assessed only against their training data. Furthermore, the reviewed studies rarely include formal applicability-domain analyses or quantitative estimates of predictive uncertainty, which limits confidence in extrapolations to unexplored IL structures. Future computational screening studies should therefore integrate rigorous external validation with explicit assessment of applicability domains and prediction uncertainty.

4.2. CAILD strategy workflow for IL-based PCMs

A comprehensive CAMD framework for IL-based PCMs should integrate molecular design, computational screening, and experimental validation [25]. The process begins by defining application-specific target properties, such as T_m, LH, specific heat, TC, viscosity, etc., followed by the generation of chemically feasible cation–anion combinations. Relevant thermophysical properties are then estimated using QSPR, group-contribution, thermodynamic, or machine-learning models, after which multi-property screening is used to select promising candidates for synthesis and experimental evaluation. Experimental results are subsequently compared with model predictions and can be incorporated into the models to improve their predictive accuracy, thereby establishing an iterative design–validation process [25, 185]. A systematic CAILD workflow for IL-based PCMs is presented in

Fig. 6. A similar strategy has been demonstrated for the QSPR-guided design of novel guanidinium ILs, for which measured T_m agreed well with predicted values [187].

5. Applications of PCMs

PCMs have enormous potential for application in numerous TES systems due to their wide melting-point range, and they possess a LH. Leveraging this potential, PCMs have been successfully implemented in an extensive range of applications, including energy-efficient buildings, solar energy storage, and battery thermal management. In this section, we comprehensively explore recent advances and innovations in PCM applications, identify current challenges, and outline future research directions required for the effective and efficient use of PCM-based systems.

5.1. Building thermal management

Typically, conventional building materials have excellent physical and mechanical strengths but poor thermal properties. PCMs excel at storing and releasing thermal energy; thus, integrating these materials into building components is a highly promising strategy for transforming conventional structures into energy-efficient buildings [71]. Qudama et al. [191] investigated the influence of paraffin wax integration into the building envelope on indoor temperature, CO_2 emissions, and energy saving. Two identical test rooms are constructed, filled with/without paraffin wax in their walls and roofs, and investigated in severely hot climate of Iraq. The research findings revealed that the test room with PCM showed improved indoor temperature by showcasing an average indoor temperature reduction of 2 °C, and substantial reduction in average heat gain by 56 W. The findings also exhibited substantial environmental and economic benefits, such as CO_2 emissions and energy cost reduction by 1.35 kg/day and 80.64 Iraqi dinar/day respectively. Furthermore, the author concluded that the PCM is more effective in roof as compared to walls.

Said et al. [192] studied the effect of inclusion of PCM on annual energy demand (AED) of a residential building in Egypt. The paraffin (M91/Q25) is chosen as PCM, and dynamic simulations are performed to evaluate the several configurations such as building with/without PCM, building with NePCMs, metallic fins-assisted with pure PCM/NePCM. The findings indicate that while pure PCM addition decreased the AED by 17.3%, the copper fin-assisted PCM-based system performed exceptionally well, lowering the AED by 28.7% compared to a conventional building without PCM (Fig. 7). Furthermore, implementation of copper fins with NePCM as hybrid system proved to be beneficial in reducing AED as compared other configurations.

To enhance the thermal performance of clay bricks, Samad et al. [193] integrated a binary eutectic mixture of Lauric acid and Palmitic acid as a PCM. The study is conducted in Islamabad to investigate the temperature regulation, heat flux, and energy saving across two individual test compartments (with conventional bricks/with PCM-enhanced bricks). The experimental test setup and the process for integrating the PCM into the clay bricks are presented in Fig. 8. The author concluded that the PCM-enhanced bricks successfully decreased the indoor temperature of the compartment by 4-5.5 °C under direct sunlight. Furthermore, the heat flux is decreased by 25-30%, alongside a substantial reduction in energy cost of 733 PKR/day.

Urbashi et al. [194] studied the influence of adding bio-composite PCM (mixture of organic PCM with water hyacinth biochar) into clay bricks for enhancing energy efficiency and environmental sustainability of buildings in hot climates. The experimental findings exhibited an indoor temperature decrease of up to 5.8 °C for the PCM-integrated cubicle as compared conventional cubicle. In addition, whole-building simulations emphasized the potential of the newly developed bio-composite PCM to save energy, lowering annual cooling demands by 31.4%. Therefore, the approach is considered economically feasible in energy savings, as it offers a short payback period of 9 year. A computational study is conducted by Farida et al. [195] to evaluate the thermal performance of eight-hole bricks enhanced separately by three organic PCMs and their combinations. Under hot Algerian climate conditions, the computational results revealed that an n-eicosane-capric acid mixture is the optimal PCM choice, lowering peak temperature and heat

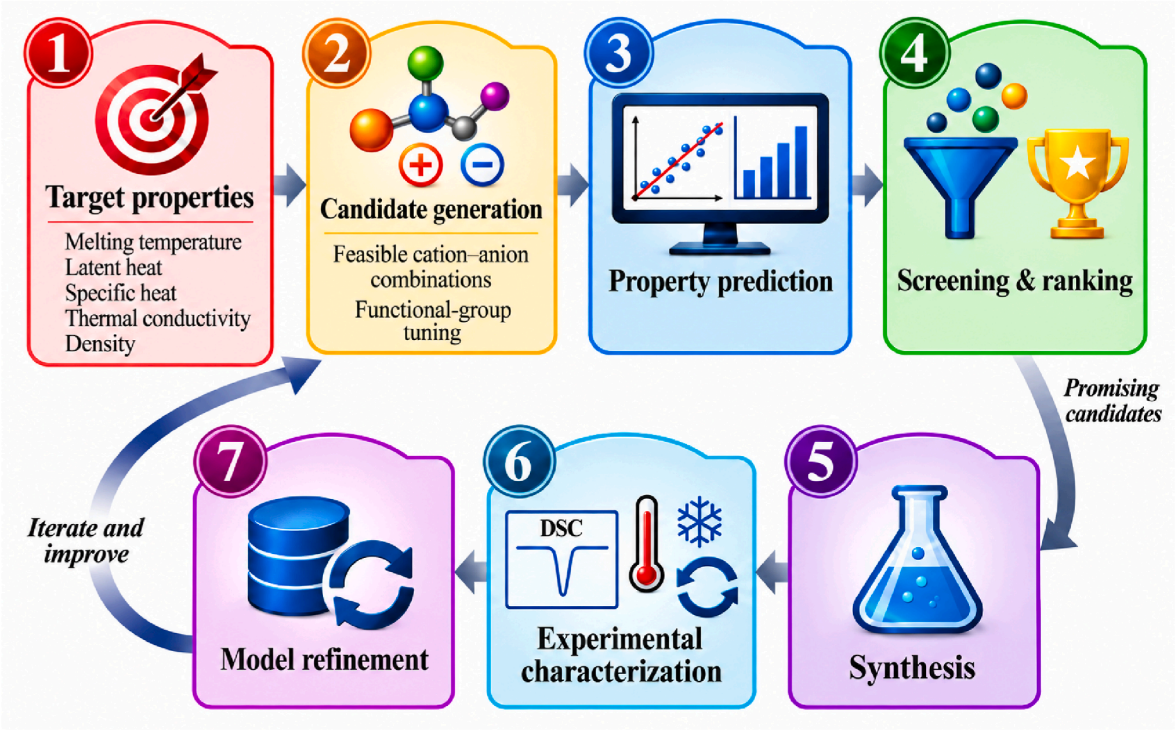


Fig. 6. CAILD strategy workflow for IL-based PCMs.

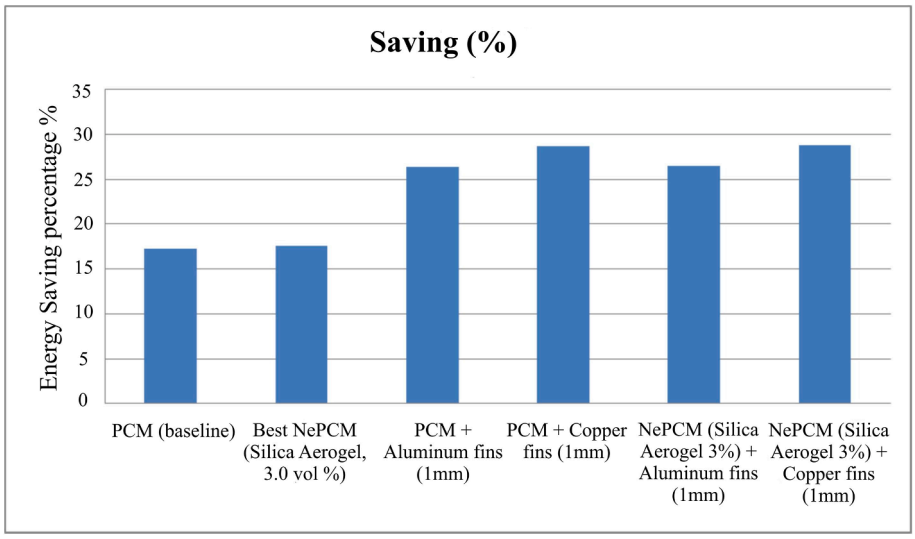


Fig. 7. Comparison of energy savings across different system configurations. Reproduced with permission from Ref. [192].

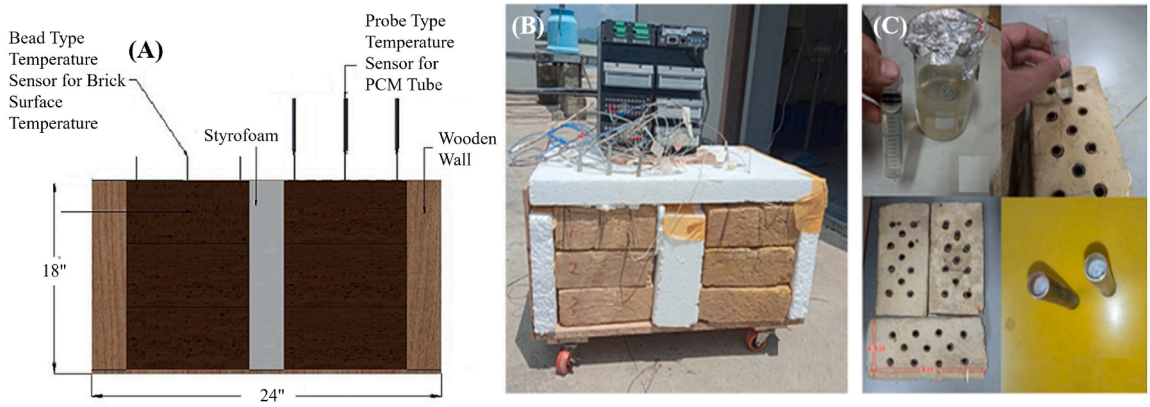


Fig. 8. Investigated experimental setup, (A) Front view of the model setup, (B) Setup under climatic conditions of Islamabad, (C) Step followed to integrate PCM into the clay bricks. Reproduced with permission from Ref. [193].

flux by 6.39% and 36.49% respectively.

In summary, current simulations and experimental studies clearly demonstrate the potential of PCMs to enhance building energy efficiency, optimize indoor thermal comfort, and promote environmental sustainability across diverse regions. Nevertheless, a significant gap remains between these findings and the widespread adoption of PCMs in practical building thermal management. Firstly, the computational studies often simplify building operations by assuming ideal and steady-state conditions, thereby neglecting the critical factors of real-world occupancy and localized meteorological deviations. On other hand, the experimental studies are conducted in small-scale test rooms for limited period of time in restricted environments. Consequently, the lack of long-term experimental findings regarding the thermal performance and stability of PCMs under changing weather conditions severely hinders their commercial viability for large-scale buildings. To overcome these bottlenecks, future research must prioritize the development of bio-based PCMs and advanced encapsulation strategies that significantly boost thermal performance. Furthermore, moving toward large-scale implementation will require the establishment of standardized procedures for PCM selection, alongside climate-tailored optimization strategies. Finally, integrating robust techno-economic analyses (TEA) and life cycle assessments (LCA) will be essential to evaluate the economic feasibility and long-term environmental benefits of PCM-enhanced architecture.

5.2. Solar energy storage

While the Sun represents an abundant, carbon-free source of renewable energy [196], the inherent intermittency of solar radiation poses a fundamental challenge [197]. This daily and seasonal variability threatens the reliable, continuous operation of various solar energy systems, including photovoltaics (PV), photovoltaic/thermal (PV/T) configurations, concentrated photovoltaics (CPV), concentrated solar power (CSP), solar thermal collectors, and solar desalination systems. Owing to their high LH, PCMs offer a promising solution by storing excess heat during periods of high solar irradiation and subsequently releasing it during periods of low or no irradiation.

Himanshu et al. [198] investigated a double-pass solar air heater (DPSAH) integrated with an organic PCM (OM46) and W-shaped artificial roughness to simultaneously enhance heat transfer and thermal storage. Both experimental and CFD simulation results demonstrated that the PCM-enhanced system extended thermal delivery by 4 h beyond sunset compared to the conventional baseline (Fig. 9-A). In addition, the influence of various flow rates on thermal efficiency and thermohydraulic performance parameter (THPP) is studied. As shown in Fig. 9-B, C, a maximum thermal efficiency of 94% and a thermohydraulic performance parameter (THPP) of 1.95 were obtained at a mass flow rate of 85 kg/h. Consequently, the PCM-integrated DPSAH demonstrated superior thermal performance and enhanced heat storage capacity

compared to the conventional system.

To address the issue of solar intermittency, Abhishek et al. [199] investigated the thermal performances of four configurations of nanofluid-based direct absorption solar collectors (DASC). The experimental setup along with various configurations, is presented in Fig. 10. The optimal architecture is achieved in Configuration 4, which comprises a 40 mg/L amorphous carbon nanofluid paired with a PCM and external thermal insulation. Under the optimal configuration, the nanofluid achieved a temperature of 65.2 °C after only 10 min of charging. Furthermore, it sustained a temperature above ambient for 175 min, marking a 3.12-fold increase in heat retention relative to the conventional baseline. Therefore, this approach proves highly viable in mitigating the inherent intermittency of solar radiation by providing continuous thermal support through the effective utilization of the stored thermal energy.

Tamim et al. [200] evaluated the thermal and exergy efficiencies of a stationary parabolic trough solar collector employing a flat metallic receiver integrated with a paraffin wax. Under optimal high-flow conditions, the thermal and exergy efficiencies of the PCM-integrated system increased by 16.6% and 47%, respectively, compared to the system without PCM. Thus, the paraffin wax as PCM substantially enhanced the thermal performance of the system, and reduced the temperature fluctuations during intermittency of solar radiations. The electrical efficiencies of PV modules are evaluated by Asim et al. [201] The researchers constructed three distinct configurations of PV modules, designated as module 1 (Conventional PV), module 2 (Paraffin-integrated PV), module 3 (NePCMs-integrated PV). The transient behavior of these modules, specifically regarding temperature, voltage, power output, and electrical efficiency over time, is illustrated in Fig. 11. The module 3 performed exceptionally well compared to the other modules. Specifically, during peak solar irradiance, it effectively regulated its temperature by decreasing it by 11.2 °C, while boosting power output and electrical efficiency by 23.1% and 2.26%, respectively, relative to module 1.

The 3D CFD model is developed by Singgih et al. [202] to investigate the influence of distinct PCM types and fins geometries on the thermal and electrical efficiencies of PV system in tropical region. The PV systems are designed by separately coupling of three distinct kinds of PCMs (Lauric acid, paraffin-ZnO, calcium nitrate tetrahydrate) with two kinds of fins geomentries including diamond and honeycomb. The findings exhibited that the Lauric acid inclusion enhanced the thermal and electrical efficiencies of the PV system by 7.04% and 12.31%, respectively. Additionally, ANOVA demonstrated that the thermal and electrical efficiencies of the PV systems were largely influenced by the choice of PCM; however, no effect was observed for fin geometries. Thus, this work concluded that the optimal PCM coupled with fins can provide an effective and efficient passive cooling strategy to significantly enhance both the thermal and electrical efficiencies of PV systems. Similarly, a computational study was conducted by Mohammed et al. [203] to evaluate the performance of a hybrid passive cooling strategy coupling circular and rectangular fins with a PCM (RT-28HC) for PV systems. The findings indicted that the designed hybrid passive cooling system efficiently decreased the average operating temperature by 19.39%, and boosted the average electrical efficiency of PV system by 5.34%. Consequently, this approach provides a highly effective solution for thermal management, thereby boosting the electrical performance of PV systems operating in hot climates. Jia et al. [204] studied the influence of PCM incorporation and optical concentration on the performance of a single-slope solar still in tropical region. The experimental setup of passive single-slope solar still, which incorporates a PCM and a Fresnel lens. Under optimized conditions of Fresnel lens size and PCM formulation, the passive solar desalination system successfully desalinated raw water with a salinity level of 30,000 ppm into freshwater meeting WHO drinking standards at a low cost of \$0.004 $(kg/m^2)^{-1}$. Furthermore, the system's thermal efficiency reached 35.34%, while attaining a daily freshwater yield of 3.30 kg/m^2. Therefore, the passive solar desalination system enhanced with a PCM and optical concentration demonstrated enormous potential to meet the demand for high-quality freshwater at a low cost. Pioneering this work, subbarama et al. [205] conducted a comparative study to assess the performance of a rectangular pyramid solar still (RPSS) integrated with PCM and fins against a conventional solar desalination system (CSDS) under the same climatic conditions. The experimental results illustrated that the freshwater daily yields for day 1 and day 2 were enhanced by 110.32% and 103.72%, respectively, alongside a two-fold increase in thermal efficiency relative to the CSDS baseline. Additionally, the RPSS yielded freshwater at a lower cost, and its quality met the requirements of the Bureau of Indian Standards. Thus, the modification of solar desalination systems with PCM and fins is considered a promising strategy to overcome the intermittency of solar radiation.

In summary, the experimental and simulation studies demonstrated the immense potential of utilizing PCMs in solar energy systems to overcome the inherent challenge of solar radiation intermittency. For solar thermal collectors, the inclusion of PCMs facilitates the storage of high thermal energy, stabilizes thermal fluctuations, and extends the operation, thereby significantly enhancing the thermal performance of the systems. PCM-enhanced PV systems also exhibited several advantages, including regulating operating temperatures, improving electrical and thermal efficiencies, and extending the lifespan of the systems. Furthermore, the PCM-integrated solar desalination systems showed the potential to mitigate the challenge of global freshwater scarcity by producing high-quality daily freshwater yields at a lower cost. The integration of PCMs into solar energy systems provides a sustainable solution to solar radiation intermittency; however, tailoring PCM properties for specific applications, integrating them into diverse solar systems, and minimizing implementation costs remain significant challenges.

5.3. Battery thermal management

Lithium-ion batteries (LIBs) have attracted the global attention due to their enormous advantages in energy storage especially in electric

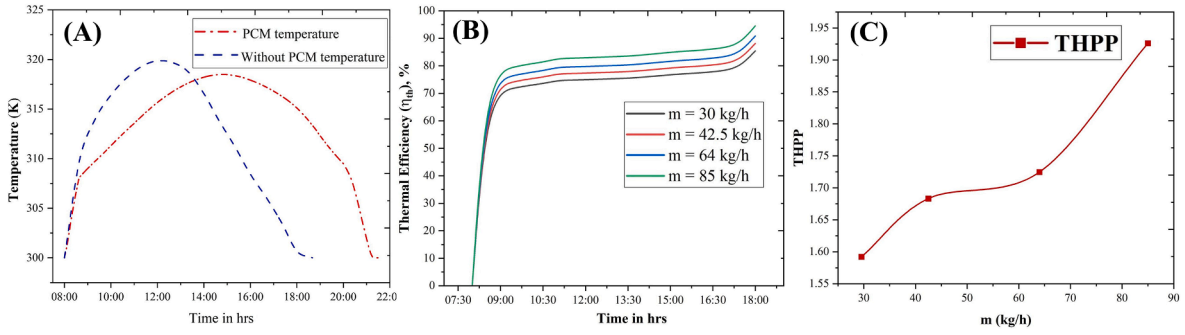


Fig. 9. Evaluation of thermal performance of the DPSAH, (A) Temperature over time, (B) Effect of flowrates on the thermal efficiency, (C) Effect of flowrates on THPP. Reproduced with permission from Ref. [198].

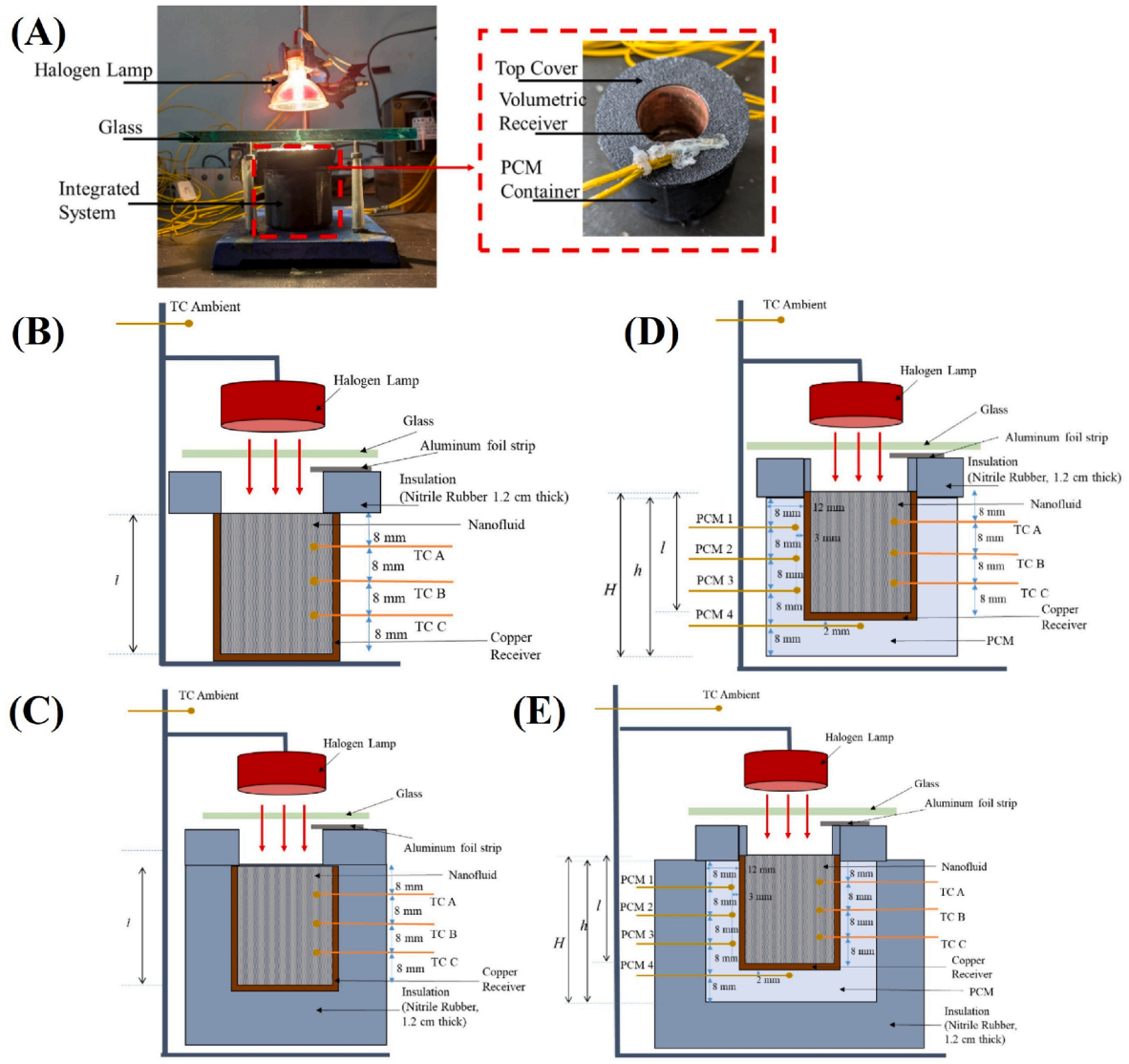


Fig. 10. Experimental setup to evaluate the thermal performances of distinct configurations of DASC, (A) Experimental setup based on configuration 3, (B) Configuration 1, (C) Configuration 2, (D) Configuration 3, (E) Configuration 4. Reproduced with permission from Ref. [199].

vehicles. A significant amount of heat is produced during the charging and discharging cycles of the battery which must be removed to maintain the temperature of the battery. The operational temperature must be from 20 °C to 50 °C for lithium-ion batteries to work effectively and efficiently; while, the temperature differences among all the cells in a battery pack should not be higher than 5 °C [206]. Therefore, it is very crucial to design an appropriate battery thermal management system (BTMS) that controls the temperature of the battery for its safe and efficient working. The passive PCM-based BTMSs have attracted widespread attention due to their ability to prevent issues related to overheating, thermal runaway, and capacity degradation in LIBs. To improve the passive BTMS for LIBs, Hossein et al. [207] constructed NePCMs by inclusion of graphene nanoplatelets (GnPs) into organic PCMs. The research findings showed that these optimized NePCMs are capable of reducing maximum cell temperatures (T_{max}) during charging and discharging of the battery. It can be observed from Fig. 12, that the T_{max} is decreased by 45.5% at a charge rate of 1.25C, likewise 38% reduction in T_{max} is observed during discharge rate of 2.5C. Consequently, the NePCMs integrated BTMS significantly enhanced the safety, efficiency, and service life of LIBs.

Praveenkumar et al. [208] prepared a novel binary nanoparticle-infused composite PCM (BNICPCM) by integrating nano-additives including zinc oxide, boron nitride, and silicon dioxide into paraffin wax for improving the efficiency of BTMS in LIBs. The optimized BNICPCM-4 formulation is employed for passive cooling in the BTMS, and the BNICPCM-4-enhanced BTMS effectively reduces the peak temperatures of LIBs during charging and discharging by 18.64% and 19.31%, respectively, compared with natural convection (Fig. 13). Therefore, the PCM-based passive BTMS ensured the efficient and safe operations of LIBs.

Similar study is conducted by Rajesh et al. [209] to develop an efficient passive BTMS for LIBs. The author constructed four composite phase change materials (CPCMs) based on the concentrations of nanomaterials into PEG. The CPCM-4 is utilized in BTMS based on its superior thermophysical properties, and effect of CPCM-based BTMS on peak surface temperature of the battery is studied in Fig. 14. It is concluded that CPCM-based BTMS successfully decreased the peaked temperatures during charging and discharging process of LIBs.

Pioneering this work, Salar et al. [210] constructed a CFD model to evaluate the thermal performance of hybrid battery thermal management system (HBTMS) for LIBs. The HBTMS comprise of PCM, metal foam, liquid cooling, and porous layers as shown in Fig. 15. The simulations are performed to study the influence of various factors, such as distinct configurations, PCM's thickness, and porous layers on thermal

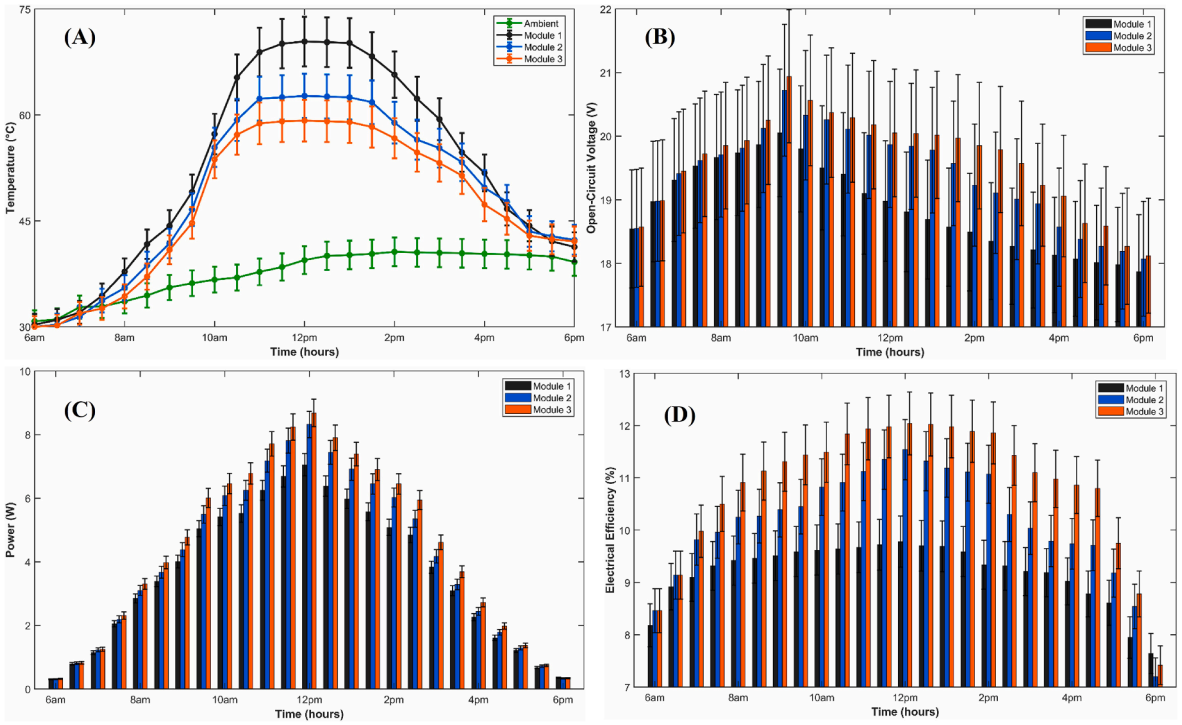


Fig. 11. Transient behavior of the modules, (A) Temperature over time, (B) Voltage over time, (C) Output power over time, (D) Electrical efficiency over time. Reproduced with permission from Ref. [201].

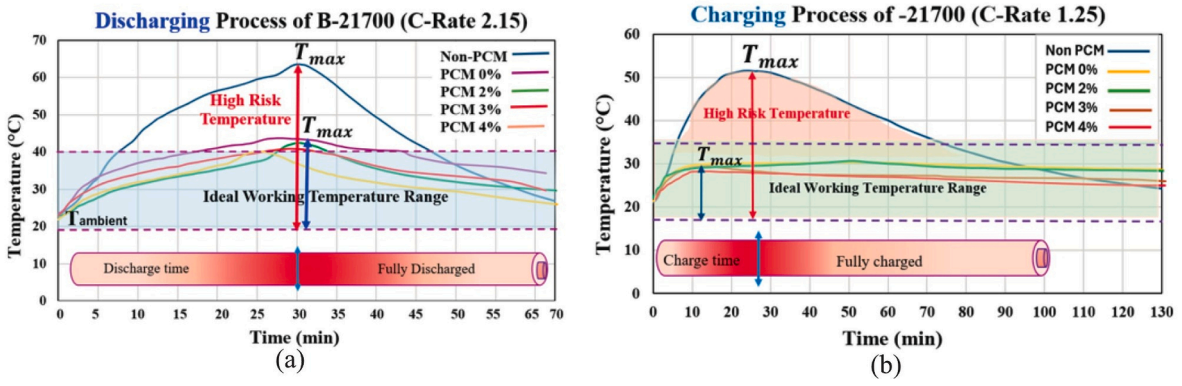


Fig. 12. Influence of NePCMs on the peak temperatures of the LIBs, (A) Discharging, (B) Charging. Reproduced with permission from Ref. [207].

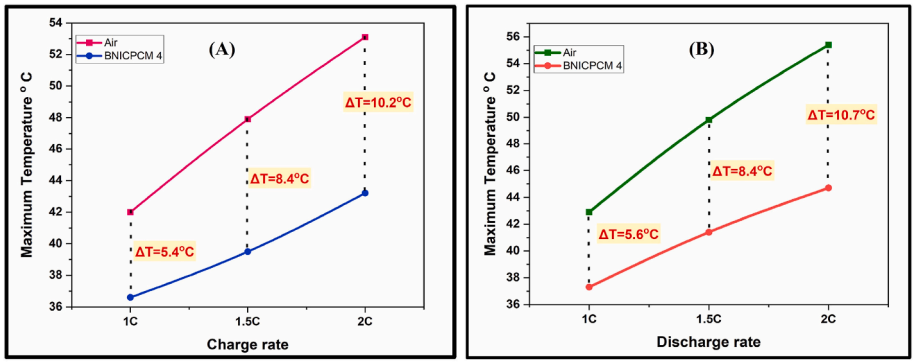


Fig. 13. Effect of PCM and air on the T_{max} of battery, (A) At various charging rates, (B) At various discharging rates. Reproduced with permission from Ref. [208].

performance of HBTMS. The results showed that the optimized HBTMS greatly decreased T_{max} at the discharge rate of 5C. The presence of metal foam, and liquid cooling significantly slow down the PCM's phase transition process, and porous layers further facilitates the heat dissipation through convective cooling. Thus, the HBTMS is considered more efficient as compared to traditional BTMS in enhancing the thermal

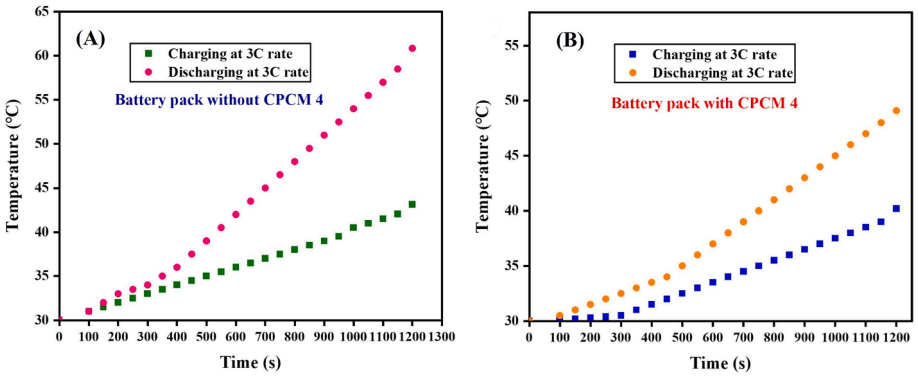


Fig. 14. Peak surface temperatures of 3S3P battery with and without CPCM-4 at 3C rates. Reproduced with permission from Ref. [209].

performance of LIBs.

Another author [211] constructed an advanced HBTMS integrating shape stabilized phase change material (SPCM) and immersion liquid cooling for efficient temperature regulation in LIBs. The SPCM is produced by paraffin, expanded graphite, boron nitride, and epoxy resin. The experimental results exhibited that the SPCM alone substantially lower the T_{max}, however implementing them along with immersion liquid cooling provide superior temperature control and uniformity across the battery pack. Specifically, the HBTMS decrease the T_{max} and ΔT_{max} to 35.8 °C and 1.9 °C respectively at a discharge rate of 3C.

Recent literature highlights the critical role of PCM-based BTMS in alleviating temperature-related hazards, including overheating, thermal runaway, and capacity degradation. Consequently, these systems not only enhance the efficiency and safety of battery operations but also significantly extend the lifespan of LIBs. However, the inherent low TC of PCMs is considered a major hurdle in heat dissipation from LIBs. To address the challenge of poor TC, the researchers integrated nanomaterials including inorganic and carbon-based into the PCMs. In addition to standalone passive PCM-based BTMSs, advanced HBTMSs which integrate both active and passive cooling mechanisms are evaluated to optimize battery thermal performance. Advanced HBTMSs outperforms the passive PCM-based BTMSs in maintaining the optimal operating temperatures for LIBs. Despite this progress, further exploration of diverse HBTMS configurations remains essential to maximize

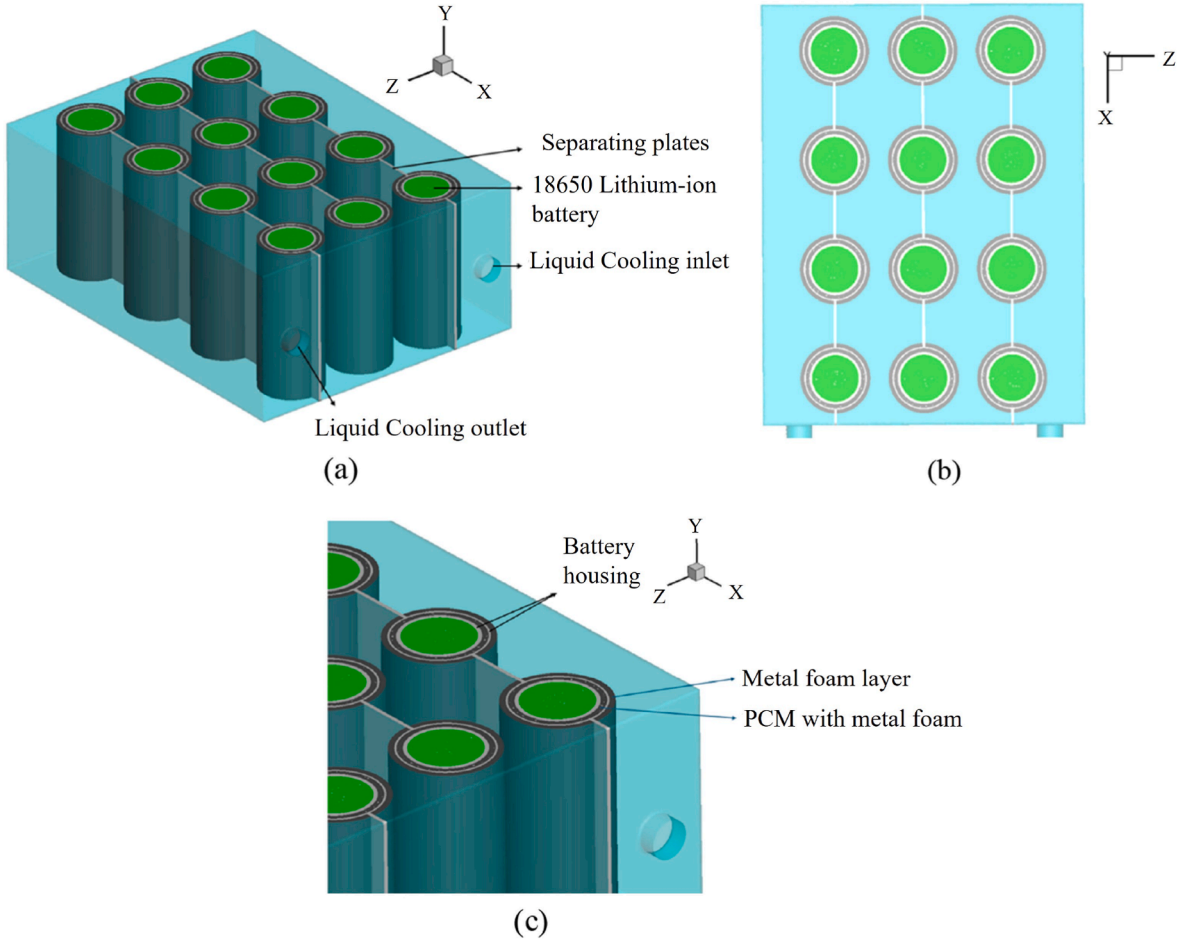


Fig. 15. Schematic diagram of the HBTMS investigated for the thermal management of LIBs. Reproduced with permission from Ref. [210].

battery performance. Furthermore, leveraging machine learning and advanced computational approaches to rationally design multifunctional PCMs and optimize their performance in LIBs represents a highly promising avenue for accelerating advancements in battery thermal management.

6. Conclusions and future perspectives

This review has systematically evaluated recent advancements in conventional organic PCMs and emerging IL-based alternatives, examining their fundamental properties, technical innovations, computational screening methods, and typical applications. Based on this review the following conclusions are drawn:

- Nanoparticle incorporation and encapsulation have been widely reported as effective strategies for mitigating key limitations of organic PCMs, including low TC, leakage, volume change, supercooling, and limited cycling stability. However, the magnitude of improvement strongly depends on the PCM-nanoparticle combination and nanoparticle loading. Carbon-based nanoparticles have been reported to be considerably more effective than their metal and metal-oxide counterparts in improving the TC of organic PCMs. Nevertheless, the influence of nano-additives on LH varies among systems and may be either favorable or unfavorable depending on the formulation. Nano-additive dispersion can also promote nucleation and reduce supercooling in organic PCMs. Furthermore, well-dispersed nanoparticles may enhance the thermal and cycling stability of paraffin-based PCMs by allowing them to retain their thermophysical properties after repeated number of cycles.
- ILs and DESs have emerged as highly tunable PCMs that address several critical shortcomings of conventional organic PCMs. Their outstanding thermal stability, negligible vapor pressure, low flammability, and phase change behavior can be finely adjusted through the diverse combinations of cations and anions. Functionalization of ILs, particularly with hydroxyl or carboxyl groups is considered a promising solution to enhance their LH (up to 190 kJ/kg). DESs are cost-effective and sustainable alternatives, with the added benefit of facile synthesis and tunable eutectic points, furthermore, they achieved LH comparable to or even higher than commercial paraffins.
- Both experimental and simulation studies have confirmed the potential of PCMs to improve indoor thermal comfort, enhance building energy efficiency, and promote environmental sustainability across diverse climatic zones.
- The utilization of PCMs in solar thermal collectors enhances the thermal energy storage, stabilizes temperature fluctuations, and significantly extends the operation. In PV systems, the incorporation of PCMs significantly improves thermal management, enhances electrical and thermal efficiencies, and prolongs system lifespan. Furthermore, PCM-enhanced solar desalination systems demonstrate longer hours of operation and produced higher yields of freshwater compared to conventional systems.
- PCM inclusion in BTMS greatly mitigates the temperature-related risks, such as overheating, thermal runaway, and capacity degradation, thereby significantly enhancing the safety, efficiency, and lifespan of LIBs.
- Finally, computational approaches such as QSPR and machine-learning models have demonstrated strong potential for predicting key properties of IL-based PCMs. By enabling fast and reliable property predictions, these methods allow researchers to efficiently screen the vast chemical space of ILs, thereby accelerating the discovery of high-performance IL-based PCMs and reducing costly trial-and-error efforts.

Based on the systematic review of conventional organic PCMs and emerging IL-based PCMs, combined with current progress, challenges and application demands, the future research directions are refined as follows. Several key directions for future research and development in this field are identified as follows:

(1) **Deep integration of multi-scale simulation and machine learning to accelerate the rational design of high-performance IL-based PCMs.** Existing QSPR and group contribution models can predict thermo-physical properties, but dedicated tools for phase change thermal storage remain insufficient. Future work shall combine first-principles calculations and molecular dynamics simulations to clarify the correlations between hydrogen-bond networks, ionic arrangements and phase change enthalpy. High-throughput screening and advanced deep learning algorithms can rapidly screen numerous cation-anion combinations to identify high-performance IL candidates. Meanwhile, expanding IL property databases will further improve model generalization, cutting experimental costs and advancing the industrial translation of IL-based PCMs.

(2) **In-depth understanding of phase transition mechanisms and establishment of structure–property relationships based on hydrogen-bonding regulation.** Hydrogen bonding dominates the phase change and energy storage performance of FILs and DESs, but its dynamic evolution during solid-liquid transition and energy contribution mechanism are not fully understood. Combining in-situ characterization techniques with theoretical calculations, researchers can analyze how hydroxyl, carboxyl, amino and other functional groups regulate the quantity, strength and spatial configuration of hydrogen bonds, and clarify the energy distribution rules in phase transition. On this basis, quantitative structure–property relationships will be built to underpin the targeted design of high-energy-density IL-based PCMs.

(3) **Development of low-cost, biodegradable DES systems and scalable fabrication processes.** Using natural raw materials such as amino acids, organic acids and sugar alcohols can expand the DES library and prepare fully biodegradable, low-toxic and non-corrosive PCMs. Systematic assessments on cycling stability, corrosivity, supercooling, LH attenuation and volume expansion are also required. In addition, continuous synthesis and encapsulation techniques should be optimized to ensure production stability during scale-up, and unified evaluation standards need to be formulated to facilitate the commercial application of DESs in solar thermal storage, energy-saving buildings and cold chain logistics.

(4) **Development of multifunctional composite PCMs for complex application scenarios.** Combining ILs with porous carbon, MXenes, metal-organic frameworks and other substrates can fabricate composite PCMs with high thermal conductivity, anti-leakage capability, photothermal conversion, flame retardancy and shape stability. For battery thermal management and electronic cooling, composites with electrical insulation and wide-temperature adaptability are particularly needed. Moreover, further studies should mitigate the adverse effects of metal/metal oxide nanoparticles on the LH of nano-enhanced PCMs, and establish a systematic framework to match nano-additives with base organic PCMs for specific thermal storage scenarios.

(5) **Enhancing long-term reliability under real-world operating conditions and advancing system-level integration.** Most existing studies only focus on material characterization, while degradation mechanisms, material compatibility and economic analysis under real working conditions are insufficient. Standard accelerated aging tests should be established to monitor property decay, corrosion and decomposition of ILs and DESs after long-term thermal cycles. Cross-scale researches from materials to equipment will optimize heat exchangers, encapsulation strategies and thermal management systems, realizing efficient coupling between PCMs and renewable energy systems. Life cycle and techno-economic assessments are also essential to compare

the carbon footprint and cost of novel PCMs with traditional ones, supporting their large-scale promotion.

In conclusion, nanomodified organic PCMs and IL/FIL/DES-based PCMs constitute a diversified thermal energy storage material system. With in-depth mechanism exploration, material innovation, computational simulation and system optimization, IL-based PCMs will overcome current technical bottlenecks and play a key role in building low-carbon, efficient and sustainable energy storage systems.

CRediT authorship contribution statement

Abdul Wahab: Writing – original draft, Software, Methodology, Investigation, Conceptualization. **Jingwen Wang:** Writing – original draft, Project administration, Methodology, Conceptualization. **Tao Xu:** Writing – review & editing, Visualization, Resources. **Pengcheng Qi:** Writing – original draft, Resources, Methodology, Investigation. **Joao A. P. Coutinho:** Writing – review & editing, Funding acquisition, Conceptualization. **Zhiwen Qi:** Writing – review & editing, Supervision, Project administration, Funding acquisition, Conceptualization.

Declaration of competing interest

The authors declare that they have no known competing financial interests or personal relationships that could have appeared to influence the work reported in this paper.

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Nomenclature

TES	Thermal energy storage
PCMs	Phase change materials
ILs	Ionic liquids
FILs	Functionalized ionic liquids
DESs	Deep eutectic solvents
CAMD	Computer-aided molecular design
QSPR	Quantitative structure-property relationship
PEG	Polyethylene glycol
TC	Thermal conductivity
LH	Latent heat
T_m	Melting temperature
NePCMs	Nano-enhanced phase change materials
MWCNTs	Multi-walled carbon nanotubes
CQDs	Carbon quantum dots
AED	Annual energy demand
DPSAH	Double-pass solar air heater
THPP	Thermohydraulic performance parameter
DASC	Direct absorption solar collectors
RPSS	Rectangular pyramid solar still
CSDS	Conventional solar desalination system
BTMS	Battery thermal management system
HBTMS	Hybrid battery thermal management system
LIBs	Lithium-ion batteries
BNICPCM	Binary nanoparticle-infused composite PCM
SPCM	Shape stabilized phase change material
CPCMs	Composite phase change materials
HBA	Hydrogen bond acceptor
HBD	Hydrogen bond donor
EG	Expanded graphite
CAILD	Computer-aided ionic liquid design
TEA	Techno-economic analysis
LCA	Life-cycle assessment

Data availability

No data was used for the research described in the article.

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