

## BEYOND THERMAL CONDUCTIVITY: A HOLISTIC APPROACH TO PERFORMANCE OPTIMIZATION OF PHASE CHANGE MATERIALS FOR HEAT STORAGE – A CRITICAL REVIEW

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### ABSTRACT

Phase change materials (PCMs) have emerged as promising candidates for thermal energy storage due to their high latent heat storage capacity and nearly isothermal phase transition behavior. However, their widespread deployment in practical heat storage systems remains constrained by several intrinsic limitations, including low thermal conductivity, leakage during phase transition, phase segregation, supercooling, and long-term thermal instability. While substantial research efforts have primarily focused on enhancing thermal conductivity, growing evidence suggests that conductivity improvement alone is insufficient to achieve optimal system-level performance. This critical review therefore adopts a holistic perspective to evaluate performance optimization strategies for PCMs beyond thermal conductivity enhancement. Key approaches reviewed

include composite formulation, encapsulation and shape-stabilization techniques, geometry-driven heat transfer enhancement, long-term durability, and sustainability considerations. Emphasis is placed on the complex trade-offs between thermal conductivity, latent heat retention, structural stability, charging–discharging rates, and long-term reliability. Emerging trends such as hybrid nanocomposites, additive manufacturing-enabled structures, and

sustainability-driven PCM design are also discussed. Finally, current research gaps and future directions are identified to bridge the disconnect between laboratory-scale material development and scalable, application-ready thermal energy storage systems.

**KEYWORDS:** Phase change materials, Thermal energy storage, Supercooling, Thermal conductivity enhancement, Phase transition, Hybrid nanocomposites, Encapsulation.

## 1.0 Introduction

The accelerating transition toward renewable and sustainable energy systems has intensified the need for efficient thermal energy storage (TES) technologies capable of mitigating temporal mismatches between energy generation and demand. Among the available TES approaches, latent heat thermal energy storage based on phase change materials (PCMs) has gained considerable attention due to its high energy storage density, narrow operating temperature range, and ability to enhance thermal management in applications such as solar thermal systems, buildings, waste heat recovery, and electronics cooling.<sup>[1-3]</sup>

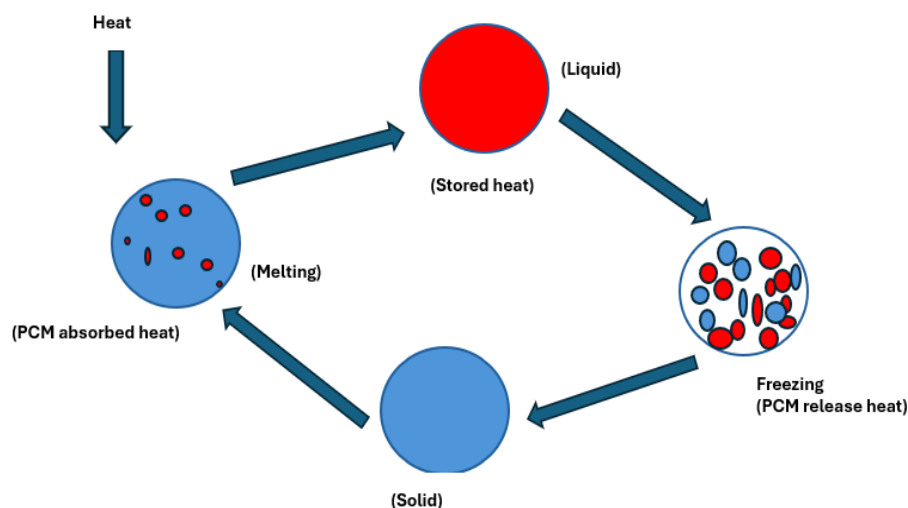
PCMs store and release thermal energy through reversible phase transitions, most commonly solid–liquid transformations. Figure 1 shows the transition phases of PCM during heating and cooling activity. Despite their advantages, the practical deployment of PCMs in large-scale or long-term heat storage systems remains limited by several inherent challenges. These include low intrinsic thermal conductivity, leakage during melting, phase segregation, supercooling, and deterioration of thermophysical properties under repeated thermal cycling.<sup>[4,5]</sup> Among these issues, low thermal conductivity, typically below  $0.3 \text{ W} \cdot \text{m}^{-1} \cdot \text{K}^{-1}$  for organic PCMs, has been widely reported as a major constraint that restricts heat transfer rates and reduces overall system efficiency.<sup>[1]</sup>

To address this limitation, extensive research efforts over the past decade have focused on enhancing the thermal conductivity of PCMs through the incorporation of conductive fillers, including carbon-based nanomaterials, ceramic particles, and metallic additives.<sup>[6]</sup> While these approaches have demonstrated measurable improvements in heat transfer performance, recent studies increasingly report that thermal conductivity enhancement alone does not ensure superior thermal energy storage performance. In many cases, the addition of conductive fillers leads to reduced latent heat capacity, increased material cost, agglomeration-induced instability, or compromised long-term reliability under cyclic

operation.<sup>[5,7]</sup> These findings highlight the limitations of conductivity-centered optimization strategies.

As a result, recent research trends have shifted toward phase change composites and integrated optimization approaches that consider not only thermal conductivity but also structural stability, leakage prevention, phase transition reliability, charging–discharging efficiency, and durability.<sup>[3,8]</sup> Strategies such as shape stabilization, encapsulation, hybrid filler systems, heat transfer surface enhancement, and optimized system geometry have been explored to improve overall performance at both material and system levels. However, many existing reviews continue to examine these approaches independently, without adequately addressing their interdependencies or performance trade-offs, limiting their usefulness for practical design and scale-up.

Therefore, this review adopts a holistic perspective to critically evaluate performance optimization strategies for phase change materials beyond thermal conductivity enhancement. The review systematically examines material modification techniques, composite architectures, encapsulation methods, and system-level design strategies, with emphasis on balancing thermal conductivity, latent heat retention, structural integrity, charging–discharging behavior, and long-term cycling stability. Emerging research directions, including hybrid nanocomposites, multifunctional PCMs, additive manufacturing-enabled thermal storage structures, and sustainability-oriented material design, are also discussed. By identifying key research gaps and proposing future directions, this review aims to support the rational development of application-ready PCM-based heat storage systems.



**Fig. 1: Phase Transition of PCM.**

## 2.0 Fundamentals of Phase Change Materials for Heat Storage

### 2.1 Classification of Phase Change Materials

Phase change materials used for latent heat thermal energy storage are commonly classified into organic, inorganic, and eutectic PCMs based on their chemical composition and phase transition characteristics. This classification is essential for understanding their thermophysical behavior, compatibility, and suitability for specific heat storage applications. Fig.2 illustrates classification of Phase Change Materials.

#### 2.1.1. Organic Phase Change Materials (PCMs)

Organic PCMs, including paraffin waxes and fatty acids, are widely utilized due to their chemical stability, congruent melting behavior, and non-corrosive nature. Their long-chain hydrocarbon structures allow them to undergo repeated melting and solidification without decomposition, making them highly suitable for long-term thermal energy storage applications. Organic PCMs typically exhibit melting temperatures between 20 and 80 °C, depending on molecular chain length, enabling their use in low- to medium-temperature systems such as building envelopes, solar air heaters, and passive cooling devices.<sup>[6,7,9]</sup>

A key thermophysical advantage of organic PCMs is their high latent heat of fusion, generally ranging from 150 to 250 kJ/kg. This high energy storage density allows them to absorb and release significant amounts of heat during phase transitions, providing effective thermal buffering. Their specific heat capacity is moderate in both solid and liquid phases, meaning that sensible heat contributes less to total storage compared to latent heat. Nevertheless, the combination of high latent heat and stable melting behavior ensures predictable thermal performance across a wide range of applications.<sup>[10]</sup>

Despite these advantages, organic PCMs suffer from inherently low thermal conductivity, typically around 0.2 W/m·K.<sup>[3,4,7]</sup> This limits heat transfer rates and can result in slow charging and discharging processes. Numerous enhancement strategies have been proposed, including embedding metal foams, adding nanoparticles, and incorporating extended surfaces such as fins. These methods significantly improve thermal conductivity but may reduce effective latent heat due to added mass or reduced PCM volume.<sup>[11]</sup>

Organic PCMs exhibit minimal supercooling and excellent cycling stability, which distinguishes them from many inorganic PCMs. Their non-corrosive nature also allows compatibility with common metals and polymers used in thermal storage containers. These

characteristics collectively make organic PCMs ideal for applications requiring long-term reliability, safety, and predictable thermal behavior.<sup>[12]</sup>

Recent advancements have shown that hybrid organic PCM systems integrated with porous supports, encapsulation technologies, and nanostructured additives have demonstrated enhanced thermal conductivity, reduced leakage, and improved cycling stability under repeated thermal operation.<sup>[13,14]</sup> Consequently, while organic PCMs remain highly promising for TES applications, future research must focus on balancing thermal conductivity enhancement, safety, durability, and sustainability to achieve scalable and high-performance heat storage systems.

### ***2.1.2. Inorganic Phase Change Materials (PCMs)***

Inorganic PCMs, including salt hydrates and metallic alloys, are characterized by high thermal conductivity, high density, and high volumetric energy storage capacity. Salt hydrates typically exhibit thermal conductivities between 0.5 and 2.0 W/m·K, enabling faster heat transfer and more efficient charging/discharging cycles compared to organic PCMs. Their melting points span a wide range (30–120 °C), making them suitable for medium- and high-temperature applications such as industrial waste heat recovery and domestic hot water systems.<sup>[14]</sup>

Inorganic PCMs also possess high latent heat values, often comparable to or exceeding those of organic PCMs. Their high density further increases volumetric storage capacity, allowing compact thermal storage designs. However, many inorganic PCMs suffer from incongruent melting, where phase separation occurs during melting and solidification. This leads to sedimentation, reduced latent heat over cycles, and long-term instability unless stabilizing additives or encapsulation methods are used.<sup>[10]</sup>

Supercooling is a major challenge associated with inorganic PCMs, particularly salt hydrates. Supercooling delays solidification and prevents timely heat release, reducing system efficiency. Nucleating agents such as borax or metal powders are often added to promote crystallization at the desired temperature, but these interventions do not always eliminate the problem entirely.<sup>[13]</sup>

Another critical limitation is corrosiveness. Many inorganic PCMs react with metals commonly used in storage containers, such as aluminum, copper, and steel. This necessitates

the use of corrosion-resistant materials or protective coatings, increasing system cost and complexity. Metallic PCMs, while offering extremely high thermal conductivity, are expensive and require specialized containment. Despite these drawbacks, inorganic PCMs remain attractive for applications requiring rapid heat transfer and high volumetric energy density.<sup>[15]</sup> Additionally, phase segregation caused by density differences among components can result in irreversible material separation and gradual loss of thermal performance during repeated melting–solidification cycles.<sup>[16]</sup> To address these limitations, recent research has increasingly focused on stabilization strategies including nucleating agents, thickeners, porous support structures, eutectic formulation, and encapsulation techniques to improve thermal reliability and structural stability.<sup>[14,17,18]</sup> Considerable research efforts have been directed toward mitigating these drawbacks through stabilization strategies including encapsulation, additives, and composite formulations.

### ***2.1.3. Eutectic Phase Change Materials (PCMs)***

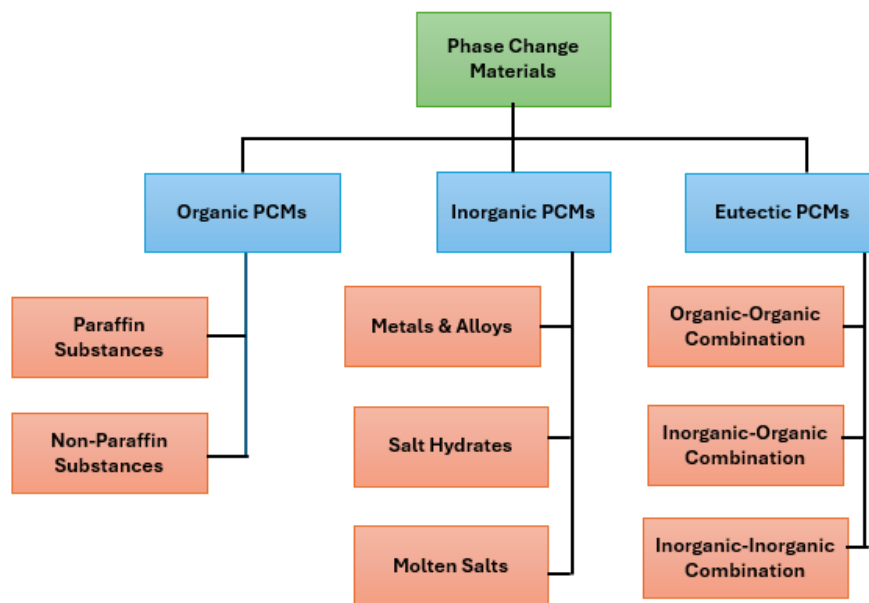
Eutectic PCMs are engineered mixtures of two or more components. These PCMs are organic–organic, inorganic–inorganic, or organic–inorganic that melt and solidify congruently at a single, sharp temperature. This unique behavior arises from the eutectic composition, where the mixture behaves as a single thermodynamic phase during melting. The ability to precisely tune the melting point by adjusting component ratios makes eutectics highly versatile for targeted thermal applications.<sup>[19]</sup> Their melting temperatures can be engineered across a wide range, enabling use in solar thermal systems, electronics cooling, and thermal regulation in buildings.

Thermophysically, eutectic PCMs exhibit balanced latent heat and thermal conductivity, often falling between the values of their parent components. Organic–inorganic eutectics, for example, combine the stability of organic PCMs with the higher conductivity of inorganic salts. This synergy can reduce supercooling, improve heat transfer, and enhance overall thermal performance.<sup>[20]</sup> Their latent heat typically ranges from 120–220 kJ/kg, depending on composition, making them competitive with both organic and inorganic PCMs.

Eutectic PCMs also demonstrate **congruent melting**, which eliminates the phase segregation issues commonly observed in salt hydrates. This ensures consistent thermal performance over repeated cycles and reduces the need for stabilizing additives. However, some eutectic systems—particularly inorganic–inorganic mixtures—may still exhibit supercooling or

require careful encapsulation to prevent moisture absorption or chemical degradation.<sup>[21]</sup> Their long-term stability depends heavily on the compatibility of the constituent materials.

Despite their advantages, eutectic PCMs face challenges related to cost, material compatibility, and manufacturing complexity. Organic–inorganic eutectics may require precise mixing and controlled solidification processes to maintain eutectic composition. Additionally, some eutectic systems may be corrosive or hygroscopic, requiring protective containment. Nevertheless, their tunability, predictable melting behavior, and balanced thermophysical properties make eutectic PCMs a promising class for advanced thermal energy storage applications. However, despite these benefits, eutectic PCMs may still suffer from challenges such as low thermal conductivity, phase segregation in certain formulations, and supercooling, particularly in inorganic eutectic systems.<sup>[7,20,21]</sup>



**Fig. 2: Classification of Phase Change Materials.**

**Table 1: Comparative Summary of PCMs.**

| Property                     | Organic PCM   | Inorganic PCM     | Eutectic PCM |
|------------------------------|---------------|-------------------|--------------|
| Latent Heat (Kj/Kg)          | 150–250       | 150–300           | 120–220      |
| Thermal Conductivity (W/M·K) | 0.2–0.4       | 0.5–2.0           | 0.3–1.0      |
| Supercooling                 | Minimal       | Pronounced        | Moderate     |
| Corrosiveness                | Non-corrosive | Corrosive         | Low          |
| Melting Behavior             | Congruent     | Often incongruent | Congruent    |
| Stability                    | Excellent     | Moderate          | Good         |

## 2.2 Thermophysical Properties Governing PCM Performance

Figure 2 illustrates the major thermophysical properties governing the performance of phase change materials (PCMs) for latent heat thermal energy storage. These properties are broadly classified into thermal properties, physical properties, and stability properties, each contributing uniquely to the efficiency, durability, and practical applicability of PCM-based heat storage systems. Thermal properties primarily determine the amount of energy that can be stored and released, physical properties influence material integration and structural behavior during phase transition, while stability properties ensure long-term operational reliability under repeated thermal cycling. An optimal PCM should possess favorable characteristics in all three categories to achieve high energy efficiency, rapid heat transfer, minimal degradation, and prolonged service life.

### 2.2.1 Thermal Properties

Thermal properties are the most fundamental characteristics governing the heat storage capability of phase change materials. These properties determine the amount of energy stored, the speed of heat absorption and release, and the suitability of a PCM for a specific operating temperature range. Because latent heat storage relies on repeated melting and solidification, thermal properties directly influence system efficiency, charging and discharging rates, and overall thermal performance.

#### 2.2.1.1 Latent Heat (Energy Storage Capacity)

Latent heat is the amount of energy absorbed or released during the phase transition without any significant temperature change. It is regarded as the most important property of PCMs because it determines the total thermal energy storage capacity per unit mass. A PCM with high latent heat can store a large amount of energy while occupying relatively little space, thereby reducing system size and improving storage efficiency. Organic PCMs such as paraffin wax generally exhibit latent heat values between 150 and 250 kJ/kg, whereas salt hydrates and eutectic mixtures may exceed these values depending on composition. High latent heat is particularly desirable for solar thermal collectors, building energy systems, and industrial waste heat recovery because it maximizes energy density while minimizing material usage.<sup>[22,23]</sup>

However, latent heat alone cannot guarantee superior performance. A PCM possessing high latent heat, but poor heat transfer characteristics may require extended charging and discharging times, thereby reducing overall system efficiency. Consequently, latent heat must

be evaluated together with thermal conductivity and diffusivity to optimize PCM performance.

#### **2.2.1.2 Thermal Conductivity (Heat Transfer Rate)**

Thermal conductivity determines how rapid heat can be transferred through a PCM during melting and solidification. Most commercial organic PCMs possess inherently low thermal conductivity, typically between 0.2 and 0.4 W/m·K, resulting in slow heat transfer and long charging/discharging periods. This limitation significantly reduces the effectiveness of thermal energy storage systems, especially under fluctuating solar irradiation or intermittent waste heat sources.

To overcome this drawback, researchers have incorporated high-conductivity additives such as aluminum oxide (Al<sub>2</sub>O<sub>3</sub>), CuO, expanded graphite (EG), graphene nanoplatelets, carbon nanotubes (CNTs), and metallic foams into PCM matrices. These additives establish conductive pathways that accelerate heat diffusion while preserving most of the latent heat capacity. Numerous studies have reported improvements in thermal conductivity using optimized nanoparticle concentrations, thereby shortening melting times and increasing heat recovery efficiency.<sup>[24,25]</sup>

#### **2.2.1.3 Specific Heat Capacity (Sensible Heat Storage)**

Specific heat capacity represents the amount of sensible heat required to raise the temperature of a PCM before melting or after solidification. Although latent heat contributes most of the stored energy, sensible heat storage also plays an important role during the heating and cooling stages surrounding the phase transition.

Materials possessing higher specific heat capacities absorb more thermal energy prior to melting and release more energy after complete solidification, thereby increasing the total energy storage capability. This property is particularly important when the operating temperature fluctuates over a wide range. The combined contribution of sensible and latent heat often determines the overall thermal storage capacity of practical PCM systems.<sup>[25]</sup>

#### **2.2.1.4 Thermal Diffusivity (Heat Transfer Rate)**

Thermal diffusivity measures how rapidly temperature changes propagate through a material and is defined as:

$$\alpha = \frac{k}{\rho C_p}$$

where  $k$  is thermal conductivity,  $\rho$  is density, and  $C_p$  is specific heat capacity.

High thermal diffusivity allows heat to penetrate rapidly throughout PCM, producing uniform melting and reducing localized overheating. Materials with low diffusivity often exhibit significant temperature gradients that delay complete phase transition and reduce thermal storage efficiency. Therefore, increasing thermal conductivity while maintaining moderate density and specific heat can substantially improve thermal diffusivity and accelerate charging/discharging processes.<sup>[26]</sup>

#### 2.2.1.5 Phase Transition Temperature (Application Match)

The phase transition temperature determines the operating temperature at which a PCM absorbs or releases latent heat. An effective PCM should possess a melting temperature closely matching the intended application. For instance, building thermal regulation generally requires melting temperatures between 20 and 30°C, whereas concentrated solar thermal systems may require temperatures exceeding 150°C.

Selecting an inappropriate melting temperature results in incomplete melting or delayed solidification, significantly reducing stored energy utilization. Consequently, carefully matching PCM transition temperature with system operating conditions remains one of the primary design considerations for thermal energy storage applications.<sup>[27,28]</sup>

#### 2.2.1.6 Phase Behavior (Supercooling and Segregation)

Phase behavior refers to the melting and solidification characteristics exhibited during repeated thermal cycling. Two major concerns are supercooling and phase segregation.

Supercooling occurs when a liquid PCM cools below its freezing temperature without solidifying, delaying heat release and lowering storage efficiency. Salt hydrates are particularly susceptible to this phenomenon. Phase segregation, on the other hand, results from density differences between constituent components during repeated melting, causing irreversible separation and loss of latent heat capacity. Various nucleating agents, thickening agents, and encapsulation techniques have been developed to minimize these effects and improve cycling stability.<sup>[23,29,30]</sup>

### 2.2.2 Physical Properties

Physical properties determine how well a PCM can be integrated into a thermal energy storage system and how it behaves mechanically during repeated melting and solidification. Unlike thermal properties, which control energy storage and heat transfer, physical properties influence material compatibility, structural integrity, and manufacturing feasibility. Parameters such as density, volume change, viscosity, and wettability directly affect encapsulation, composite fabrication, heat exchanger design, and long-term operational safety.

#### 2.2.2.1 Density (Volumetric Energy Storage)

Density determines the amount of thermal energy that can be stored within a given volume. While latent heat is usually expressed on a mass basis (kJ/kg), practical engineering systems are constrained by available volume. Therefore, volumetric energy density, calculated as the product of density and latent heat, provides a more realistic measure of storage capacity. High-density PCMs enable compact thermal storage units, making them attractive for applications with limited installation space, such as electric vehicles, electronics cooling, and compact solar thermal collectors. Density also influences buoyancy-driven convection during melting, affecting heat transfer and melting uniformity.<sup>[25,26]</sup>

#### 2.2.2.2 Volume Change (Expansion and Mechanical Stress)

Most PCMs undergo volume expansion during melting and contraction during solidification. Excessive volume change can induce mechanical stress on encapsulation shells, heat exchanger tubes, and storage containers, leading to deformation, leakage, or fatigue failure after repeated thermal cycles. Organic paraffins generally exhibit moderate expansion (typically 5–15%), whereas some inorganic PCMs experience larger dimensional changes. To accommodate expansion, engineers often incorporate expansion chambers, flexible encapsulation materials, or optimized container geometries. Minimizing volumetric change is therefore essential for maintaining structural integrity and extending system lifespan.<sup>[27,28,31]</sup>

#### 2.2.2.3 Viscosity and Wettability (Composite Formation)

Viscosity governs the flow behavior of molten PCMs, while wettability describes the ability of the liquid PCM to spread and adhere to supporting matrices or heat transfer surfaces. Low-viscosity PCMs can more easily infiltrate porous materials such as expanded graphite, metal foams, or polymer networks, facilitating the fabrication of shape-stabilized composites. Good wettability promotes intimate thermal contact between the PCM and conductive additives,

reducing interfacial thermal resistance and enhancing heat transfer. Conversely, high viscosity or poor wettability can hinder composite preparation, reduce nanoparticle dispersion, and impair long-term thermal performance. Surface modification of fillers and the use of coupling agents are common strategies to improve compatibility and achieve homogeneous composite structures.<sup>[25,31]</sup>

### 2.2.3 Stability Properties

Stability properties determine the long-term reliability and durability of phase change materials during prolonged operation. Since PCM-based thermal energy storage systems often undergo thousands of heating and cooling cycles over their service life, maintaining consistent thermophysical performance is critical. Stability encompasses thermal stability, chemical stability, and cyclic stability, each of which contributes to sustained energy storage capacity, operational safety, and economic viability.

#### 2.2.3.1 Thermal Stability (Resistance to Decomposition)

Thermal stability refers to the ability of a PCM to withstand repeated heating without undergoing thermal decomposition or significant changes in composition. A thermally stable PCM retains its latent heat, melting temperature, and chemical structure when exposed to operating temperatures over extended periods. Poor thermal stability may result in decomposition, volatilization, or oxidation, leading to reduced storage capacity and shortened service life. Thermogravimetric analysis (TGA) is commonly employed to evaluate decomposition temperatures and determine the safe operating range of PCMs. Organic paraffins generally exhibit good thermal stability below approximately 200°C, whereas many inorganic PCMs can tolerate substantially higher temperatures depending on composition.<sup>[22,32]</sup>

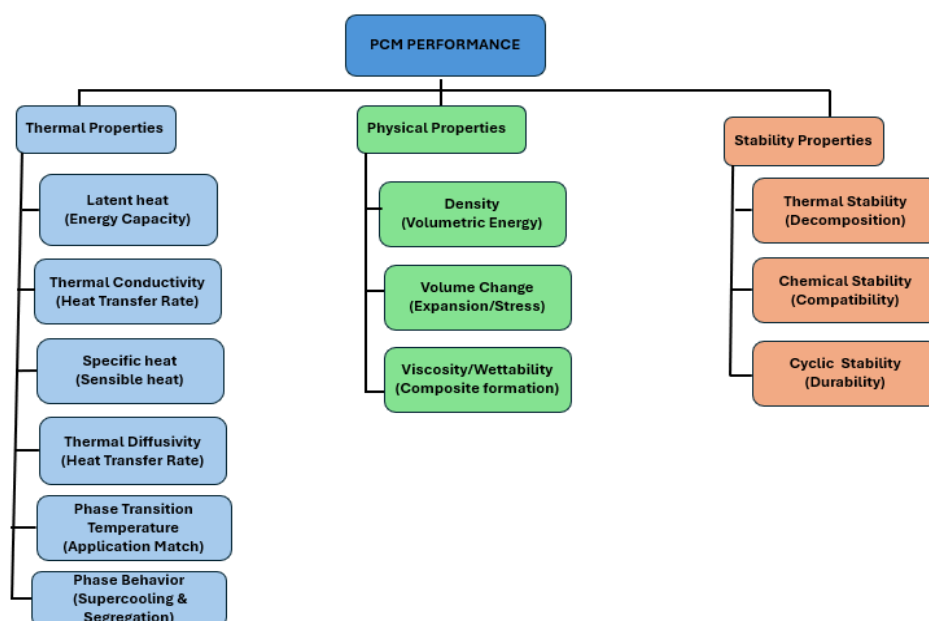
#### 2.2.3.2 Chemical Stability (Material Compatibility)

Chemical stability describes the resistance of a PCM to undesirable chemical reactions with container materials, encapsulation shells, conductive additives, or the surrounding environment. Chemically unstable PCMs may corrode metallic containers, react with fillers, or absorb moisture, resulting in degradation of thermophysical properties and reduced operational reliability. Compatibility testing with common construction materials such as aluminum, copper, stainless steel, and polymers is therefore essential during PCM selection. Protective coatings, corrosion inhibitors, and inert encapsulation materials are often employed to minimize chemical interactions and preserve long-term performance.<sup>[27]</sup>

### 2.2.3.3 Cyclic Stability (Long-Term Durability)

Cyclic stability is the ability of a PCM to maintain its thermophysical properties after repeated melting and solidification cycles. During long-term service, some PCMs may exhibit degradation through phase separation, supercooling, leakage, chemical decomposition, or loss of latent heat. A high-quality PCM should preserve its latent heat capacity, phase transition temperature, and thermal conductivity over hundreds or thousands of cycles with minimal performance loss. Differential scanning calorimetry (DSC) is frequently used to evaluate changes in melting behavior after accelerated cycling tests. Recent studies indicate that properly stabilized composite PCMs containing conductive nanofillers and supporting matrices can retain more than 90% of their original latent heat after thousands of thermal cycles, demonstrating excellent durability for practical thermal energy storage systems.<sup>[32,33,34]</sup>

Overall, stability properties are essential for ensuring that PCM-based heat storage systems deliver consistent performance throughout their operational lifetime. Even materials with excellent thermal and physical properties may prove unsuitable if they undergo rapid degradation during repeated thermal cycling. Consequently, thermal, chemical, and cyclic stability should be evaluated alongside thermal and physical characteristics when selecting or designing PCMs for long-term heat storage applications.

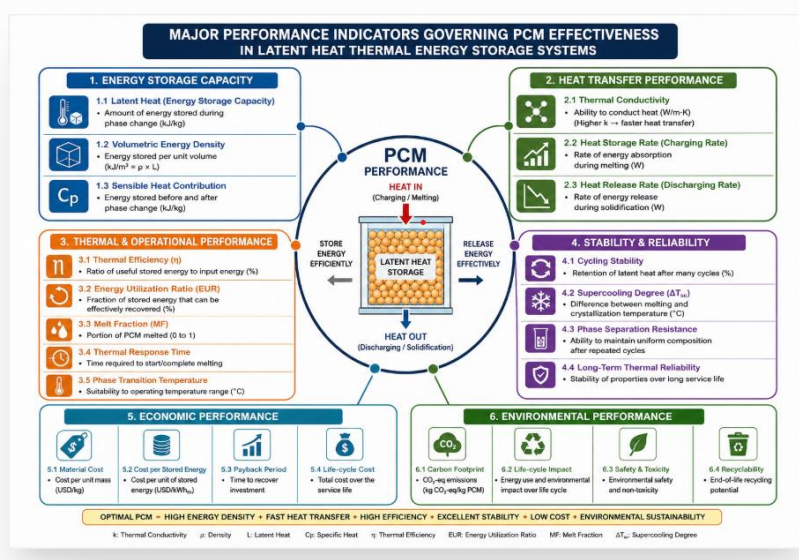


**Fig. 3: Schematic representation of thermophysical properties governing phase change material (PCM) performance.**

### 2.3 Performance Metrics for PCM-Based Heat Storage Systems

The effectiveness of a phase change material (PCM) in thermal energy storage (TES) systems is not determined by a single thermophysical property but rather by a combination of performance metrics that collectively describe its energy storage capability, heat transfer characteristics, thermal reliability, mechanical stability, and economic feasibility. While thermophysical properties such as latent heat, thermal conductivity, and melting temperature define the inherent characteristics of a PCM, performance metrics quantify how effectively these properties translate into practical system performance under real operating conditions. Consequently, selecting or designing an efficient PCM requires evaluating multiple performance indicators simultaneously rather than relying solely on latent heat capacity.<sup>[35,36]</sup>

Performance metrics are particularly important because the practical performance of PCMs is influenced by factors beyond their intrinsic material properties. For example, PCM with high latent heat but poor thermal conductivity may exhibit slow charging and discharging, while another material with moderate latent heat but superior heat transfer characteristics may deliver better overall thermal efficiency. Similarly, repeated thermal cycling can gradually degrade PCM properties, making long-term stability an equally important evaluation criterion. Therefore, researchers increasingly adopt comprehensive performance assessments incorporating thermal, physical, chemical, economic, and environmental indicators to optimize PCM selection for applications such as solar thermal collectors, building energy systems, industrial waste heat recovery, and electronic cooling.<sup>[25,28,37]</sup>



**Figure 4:** summarizes major performance indicators governing PCM effectiveness in latent heat thermal energy storage systems.

### 2.3.1 Energy Storage Capacity (Latent + Sensible Heat)

Energy storage capacity is the primary performance metric for evaluating PCMs because it determines the amount of thermal energy that can be stored and subsequently released during melting and solidification. The total stored energy consists of sensible heat absorbed before melting, latent heat during phase transition, and sensible heat released after solidification.

The total stored energy can be expressed as

$$Q_{total} = mC_{ps}(T_m - T_i) + mL + mC_{pl}(T_f - T_m)$$

Where

- $m$  = PCM mass (kg)
- $C_{ps}$  = specific heat of solid PCM
- $C_{pl}$  = specific heat of liquid PCM
- $L$  = latent heat (kJ/kg)
- $T_i$  = initial temperature
- $T_m$  = melting temperature
- $T_f$  = final temperature

For most latent heat storage applications, the latent heat component contributes between 70% and 90% of the total stored energy, making it the dominant energy storage mechanism. Consequently, PCMs possessing high latent heat values provide higher energy density and reduce the required storage volume. Organic paraffins typically exhibit latent heat values between 150 and 250 kJ/kg, whereas some salt hydrates exceed 300 kJ/kg.<sup>[34,35]</sup>

### 2.3.2 Volumetric Energy Density

Volumetric energy density is an important performance metric for evaluating the practical effectiveness of phase change materials (PCMs) in thermal energy storage (TES) systems. While latent heat is commonly expressed on a mass basis (kJ/kg), many engineering applications are constrained by the available installation volume rather than the total material mass. Consequently, volumetric energy density provides a more realistic assessment of a PCM's storage capability because it quantifies the amount of thermal energy that can be stored within a given volume. This parameter is particularly significant in applications where

compactness is essential, such as electric vehicles, aerospace thermal management, portable thermal storage devices, and building-integrated energy systems. A PCM with high volumetric energy density can store more energy within a smaller space, thereby reducing the overall size and weight of the storage unit while improving system efficiency.<sup>[27,35]</sup>

Volumetric energy density is determined by the product of the PCM density and its latent heat capacity, and it can be expressed as:

$$E_v = \rho L$$

Where:

- $E_v$  = volumetric energy density (kJ/m<sup>3</sup>),
- $\rho$  = density of the PCM (kg/m<sup>3</sup>), and
- $L$  = latent heat of fusion (kJ/kg)

This relationship demonstrates that both density and latent heat contribute directly to the amount of energy stored per unit volume. Although some organic PCMs exhibit relatively high latent heat values, their comparatively low densities may reduce their volumetric energy storage capacity. Conversely, many inorganic PCMs, such as salt hydrates, possess higher densities, enabling them to achieve superior volumetric energy densities despite having similar or slightly lower latent heat values. Therefore, volumetric energy density should be considered alongside gravimetric latent heat when selecting PCMs for practical heat storage applications, particularly where space limitations dictate system design.<sup>[34,36]</sup>

### 2.3.3 Thermal Conductivity Enhancement Ratio

Thermal conductivity largely determines the rate at which heat is absorbed and released.

The enhancement ratio is defined as

$$\eta_k = \frac{k_{composite}}{k_{PCM}}$$

where

- $k_{composite}$  is the thermal conductivity of the modified PCM.
- $k_{PCM}$  is the conductivity of pure PCM.

Nano-enhanced PCMs containing graphene, carbon nanotubes, expanded graphite, or metallic nanoparticles frequently exhibit thermal conductivity improvements that significantly reduce charging and discharging times.<sup>[24,25]</sup>

### 2.3.4 Charging Rate

Charging rate represents the speed at which the PCM stores thermal energy during melting.

It is commonly evaluated by

- charging time,
- melting completion time,
- heat storage rate (W),
- percentage melt fraction.

A shorter charging time indicates faster heat absorption and improved system responsiveness.

The charging rate depends primarily on thermal conductivity, airflow rate, heat transfer coefficient, and PCM geometry. For solar thermal collectors, rapid charging enables efficient utilization of intermittent solar radiation.

### 2.3.5 Discharging Rate

Discharging rate measures the speed at which stored latent heat is released during solidification.

It is commonly expressed using

- Cooling time,
- Heat release rate,
- Outlet air temperature,
- Discharge efficiency.

Rapid heat release improves the ability of PCM systems to provide continuous heating after the external heat source becomes unavailable.

### 2.3.6 Thermal Efficiency

Thermal efficiency evaluates how effectively incoming heat is converted into useful stored thermal energy.

It is calculated as

$$\eta = \frac{Q_{stored}}{Q_{input}} \times 100$$

Where:

- $Q_{stored}$  is useful stored energy.
- $Q_{input}$  is supplied thermal energy.

Thermal efficiency is one of the most widely reported performance indicators in solar collectors because it directly reflects system effectiveness.

### 2.3.7 Heat Storage Rate

The instantaneous heat storage rate describes the rate of thermal energy accumulation.

$$\dot{Q} = mC_p \frac{dT}{dt}$$

Or during phase transition

$$\dot{Q} = mL \frac{df}{dt}$$

Where:

$f$  is the melt fraction.

Higher heat storage rates indicate faster thermal charging.

### 2.3.8 Energy Utilization Ratio (EUR)

Energy utilization ratio quantifies the fraction of stored energy that can actually be recovered.

$$EUR = \frac{Q_{released}}{Q_{stored}}$$

Values approaching unity indicate minimal thermal losses and efficient utilization.

### 2.3.9 Melt Fraction

Melt fraction describes the proportion of PCM that has melted.

$$MF = \frac{m_{melted}}{m_{total}}$$

It ranges from 0 to 1.

Monitoring melt fraction helps evaluate charging completeness and optimize collector operation.

### 2.3.10 Cycling Stability

Cycling stability measures the ability of a PCM to retain its thermophysical properties after repeated melting and solidification. It is commonly evaluated through

- Latent heat retention,
- Melting temperature variation,
- Thermal conductivity retention,
- Mass loss,
- Decomposition temperature.

High-performance PCMs should maintain more than 90–95% of their original latent heat after several hundred or several thousand thermal cycles.<sup>[36,37]</sup>

### 2.3.11 Supercooling Degree

Supercooling is the difference between melting and crystallization.

$$\Delta T_{SC} = T_m - T_c$$

Where:

- $T_m$  = melting temperature
- $T_c$  = crystallization temperature

Lower supercooling promotes faster and more predictable heat release

### 2.3.12 Phase Separation Index

For salt hydrates and eutectic PCMs, phase separation significantly reduces long-term performance.

It is quantified by monitoring

- Latent heat degradation,
- Composition uniformity,
- Density variation after repeated cycles.

Shape stabilization and encapsulation techniques help minimize segregation.

### 2.3.13 Long-Term Thermal Reliability

Long-term reliability combines several degradation indicators including

- latent heat retention,
- melting point stability,
- thermal conductivity retention,
- chemical stability,
- cyclic stability.

A reliable PCM should maintain nearly constant performance throughout its intended service life. This metric is especially important for building-integrated thermal storage and industrial applications where systems are expected to operate for periods with minimal maintenance.<sup>[28]</sup>

### 2.3.14 Economic Performance

Beyond technical performance, economic viability is essential for large-scale deployment of PCM technologies. Common economic metrics include:

- Cost per unit mass (USD/kg)
- Cost per unit of stored energy (USD/kWh)
- Payback period
- Life-cycle cost
- Levelized cost of thermal energy storage (LCOTES)

Materials with slightly lower latent heat but significantly lower cost often provide better overall economic performance than expensive high-performance alternatives.

### 2.3.15 Environmental Performance

Environmental sustainability is increasingly incorporated into PCM evaluation. Key indicators include:

- Carbon footprint (kg CO<sub>2</sub>-eq/kg PCM)
- Life-cycle energy consumption
- Recyclability
- Toxicity
- Biodegradability
- Environmental impact over the system life cycle

Life cycle assessment (LCA) has become an important tool for comparing organic, inorganic, bio-based, and composite PCMs from a sustainability perspective.

## 3.0 Limitations of Conventional Phase Change Materials

Despite the significant potential of phase change materials (PCMs) for latent heat thermal energy storage, their practical application in real-world heat storage systems remain constrained by several intrinsic limitations. These challenges originate from both material-level deficiencies and system-level interactions, which collectively hinder heat transfer efficiency, operational reliability, and long-term performance. Understanding these limitations is essential for contextualizing the need for holistic performance optimization strategies

### 3.1 Low Intrinsic Thermal Conductivity

Conventional PCMs, especially organic paraffins and many salt hydrates, possess inherently low thermal conductivity, typically in the range of  $0.2\text{--}0.4\text{ W/m}\cdot\text{K}$  for organics and only moderately higher for some inorganics. This low conductivity severely restricts heat transfer during both melting and solidification, leading to slow charging and discharging rates and pronounced temperature gradients within the PCM domain.<sup>[37-39]</sup> As a result, only regions close to the heat transfer surface may fully participate in phase change, while interior zones remain partially melted or solid, reducing effective latent heat utilization.

The impact of low thermal conductivity becomes more critical in large-scale or compact TES configurations, such as packed bed storage, shell-and-tube exchangers, or building envelope integration. Numerical and experimental studies consistently show that conduction-dominated melting leads to long melting times and incomplete phase change, even under favorable boundary conditions.<sup>[38,40]</sup> Natural convection in the liquid phase can partially alleviate this limitation, but its effect is highly geometry-dependent and often insufficient to ensure uniform melting.

To mitigate low conductivity, numerous enhancement strategies have been proposed, including embedding high-conductivity fins, metal foams, graphite matrices, and dispersing nanoparticles (nano-PCMs). While these approaches significantly increase effective thermal conductivity and reduce melting/solidification times, they introduce new challenges: increased cost, potential sedimentation of fillers, reduced PCM fraction, and possible changes in rheology or stability.<sup>[39,41]</sup> Thus, low thermal conductivity remains a fundamental limitation that requires careful system-level design and material engineering.

### 3.2 Leakage during Phase Transition

Leakage of liquid PCM during melting is a critical limitation, particularly for organic PCMs such as paraffins and fatty acids. When the PCM transitions to the liquid state, capillary forces, gravity, and thermal expansion can drive the liquid out of its containment if encapsulation is inadequate or damaged. This leads to loss of active PCM mass, contamination of surrounding components, and, in the case of flammable organics, potential fire hazards.<sup>[42,43]</sup> Leakage is especially problematic in building applications, where escaped PCM can damage finishes or insulation.

Microencapsulation, macro-encapsulation, and composite PCM structures have been widely investigated to address leakage. Polymer shells, metal capsules, and porous matrices (e.g., expanded graphite, metal foams) can physically confine the PCM and prevent liquid escape.<sup>[44,45]</sup> However, these solutions increase manufacturing complexity and cost, and encapsulation shells themselves may suffer from fatigue, cracking, or chemical degradation under repeated thermal cycling, eventually reintroducing leakage risks.

Leakage also affects thermal performance by altering the effective PCM distribution and contact with heat transfer surfaces. Once PCM migrates away from the designed interface, heat transfer deteriorates and the system's storage capacity declines. Long-term studies of encapsulated PCM systems show that even minor leakage over many cycles can significantly reduce effective latent heat and compromise reliability.<sup>[43,46]</sup> Therefore, leakage during phase transition remains a major practical limitation, demanding robust encapsulation design and quality control

### 3.3 Phase Segregation and Supercooling

Phase segregation is a prominent limitation in salt hydrate PCMs and some eutectic systems. During melting, water of crystallization may separate from the salt, leading to incongruent melting and formation of distinct phases with different compositions. Over repeated cycles, heavier salt phases can settle, while lighter liquid phases migrate, causing stratification and loss of latent heat capacity.<sup>[47,48]</sup> This segregation reduces the effective enthalpy of fusion and leads to non-uniform melting/solidification behavior, severely degrading long-term performance.

Supercooling delayed solidification below the equilibrium freezing point is another critical issue, particularly in inorganic PCMs. Due to nucleation barriers, the liquid phase may remain metastable over a wide temperature range, preventing timely crystallization and heat release.<sup>[49]</sup> This results in extended solidification times, reduced discharge efficiency, and thermal hysteresis. In practical systems, supercooling can cause PCM to fail to solidify during the available cooling period (e.g., overnight), leaving residual heat and reducing subsequent storage capacity.

Various mitigation strategies have been explored, including the addition of nucleating agents, thickening additives, and formulation of stabilized salt hydrates or eutectics. While these measures can reduce supercooling and phase segregation, they rarely eliminate them entirely

and may introduce new issues such as increased viscosity, sedimentation of additives, or changes in melting point.<sup>[47,50]</sup> Consequently, phase segregation and supercooling remain major limitations for many inorganic and eutectic PCMs, particularly in long-term, cyclic applications.

### 3.4 Poor Structural and Mechanical Stability

Conventional PCMs often exhibit poor structural and mechanical stability, especially when used in bulk form or within rigid containers. Volume change during phase transition—expansion upon melting and contraction upon solidification—can induce mechanical stresses on encapsulation shells, container walls, and surrounding structures.<sup>[51]</sup> Over repeated cycles, these stresses can lead to cracking, delamination, or loss of contact between PCM and heat transfer surfaces, reducing heat transfer efficiency and potentially causing leakage.

Inorganic PCMs, particularly salt hydrates, may form brittle crystalline structures that fracture under thermal cycling or mechanical loads. Organic PCMs, while softer, can still cause deformation of polymeric encapsulation or composite matrices due to repeated expansion and contraction. Studies on PCM-enhanced building materials and composite panels report issues such as microcracking, debonding, and loss of structural integrity after extended cycling.<sup>[52,53]</sup>

Mechanical instability is further exacerbated when PCMs are combined with porous matrices or reinforcement structures. Differential thermal expansion between PCM and matrix can generate internal stresses, leading to pore collapse, matrix fracture, or loss of PCM confinement.<sup>[54]</sup> Recent work on flexible and shape-stable PCMs aims to address these issues by incorporating elastomeric binders or fibrous reinforcements, but such materials are still emerging and often more expensive. Overall, poor structural and mechanical stability remains a significant barrier to long-term, reliable PCM integration in structural and compact TES applications.

### 3.5 Long-Term Degradation under Thermal Cycling

Thermal cycling stability is a critical performance requirement for PCMs intended for long-term heat storage applications. Many conventional PCMs experience gradual degradation of latent heat capacity, shifts in phase transition temperature, or chemical instability after hundreds or thousands of thermal cycles.<sup>[47,55]</sup> For inorganic PCMs, repeated cycling can exacerbate segregation and supercooling, leading to progressive loss of performance. For

organic PCMs, oxidation, polymerization, or contamination may occur, especially at elevated temperatures.

Encapsulated PCMs are particularly susceptible to shell fatigue and microcracking. As PCM expands and contracts during phase change, encapsulation shells experience cyclic mechanical loading. Over hundreds or thousands of cycles, this can lead to crack formation, shell rupture, and eventual leakage.<sup>[45,56]</sup> Once encapsulation fails, the PCM may escape, oxidize, or react with surrounding materials, further accelerating degradation.

Long-term studies of PCM-enhanced building components, solar collectors, and domestic hot water systems consistently report performance decline over extended operation, even when initial performance is promising.<sup>[52,55]</sup> This degradation manifests as reduced energy storage capacity, altered phase change temperature, increased supercooling, and mechanical damage. While improved formulations, stabilizers, and advanced encapsulation can extend service life, long-term degradation under thermal cycling remains a fundamental challenge that must be carefully evaluated in system design and material selection.

### 3.6 Flammability and Corrosiveness

Organic PCMs such as paraffins and some fatty acids are flammable, posing safety risks in high-temperature or industrial environments. Fire hazards necessitate strict thermal control, fire-resistant encapsulation, and additional safety systems, complicating system design and increasing cost.<sup>[42,58]</sup> This limitation is particularly relevant in applications with high heat fluxes or potential ignition sources.

Many inorganic PCMs, particularly salt hydrates, are corrosive to common metals such as steel, copper, and aluminum. Corrosion of containment materials leads to structural failure, contamination of PCM, and reduced thermal conductivity at the interface.<sup>[47,57]</sup> To mitigate this, corrosion-resistant alloys, protective coatings, or polymeric containers are used, but these increase cost and may introduce new compatibility issues.

### 3.7 Environmental and Economic constraints

Advanced PCM systems incorporating nanoparticles, metal foams, or complex encapsulation are often significantly more expensive than conventional materials. Economic analyses show that high material and manufacturing costs can limit the feasibility of PCM-based TES in large-scale applications.<sup>[41,59]</sup> Additionally, environmental concerns related to nanoparticle

release, polymer waste, and end-of-life disposal are increasingly recognized, prompting the search for more sustainable PCM formulations.

### 3.8 Implications for Performance Optimization

The limitations discussed above highlight that conventional PCMs, when used without modification, are insufficient to meet the performance demands of advanced thermal energy storage systems. Importantly, these challenges are interrelated: efforts to mitigate one limitation often exacerbate another. For example, increasing thermal conductivity through filler addition may reduce latent heat capacity, while encapsulation may introduce thermal resistance.

These trade-offs underscore the inadequacy of single-parameter optimization strategies and reinforce the need for a holistic approach that integrates material modification, structural stabilization, and system-level design. The subsequent sections of this review critically examine such strategies, with emphasis on balancing competing performance requirements to achieve application-ready PCM-based heat storage solutions.

## 4.0 Thermal Conductivity Enhancement Techniques

Low thermal conductivity remains one of the most significant barriers to the efficient utilization of phase change materials (PCMs) in heat storage systems. To overcome this limitation, extensive research has been devoted to enhancing heat transfer within PCMs through material modification strategies. These techniques primarily involve the incorporation of thermally conductive additives and the development of composite structures. While notable improvements have been reported, conductivity enhancement is often accompanied by trade-offs that necessitate critical evaluation.

### 4.1 Carbon-Based Conductive Additives

Carbon-based materials are among the most effective additives for enhancing the thermal conductivity of PCMs due to their intrinsically high thermal conductivity and low density. Commonly investigated carbon-based fillers include carbon nanotubes, graphene, graphene nanoplatelets, and expanded graphite. These materials improve heat transfer by forming conductive pathways within the PCM matrix, facilitating phonon transport during phase transition.<sup>[60]</sup>

Graphite-enhanced PCMs also exhibit improved structural stability, reduced leakage, and enhanced cycling durability. EG is particularly effective in stabilizing salt hydrates by reducing phase segregation and supercooling. Composite PCMs based on EG have been successfully applied in solar collectors, building materials, and industrial heat recovery systems.<sup>[5,24,33]</sup> Compared to nanoparticles, EG is cheaper, easier to disperse, and less prone to agglomeration, making it one of the most practical enhancement strategies. However, excessive filler content can disrupt the phase change process, reduce latent heat storage capacity, and increase material cost. In the case of carbon nanotubes and graphene-based additives, dispersion challenges and agglomeration remain major obstacles, often requiring surface functionalization or advanced mixing techniques to achieve stable composites.<sup>[33]</sup>

#### 4.2 Incorporation of High-Conductivity Nanoparticles

One of the most widely studied methods for improving PCM thermal conductivity is the dispersion of high-conductivity nanoparticles such as CuO, SiC, TiO<sub>2</sub>, graphene, and carbon nanotubes (CNTs). Nanoparticles increase the effective thermal conductivity of the PCM by forming conductive pathways within the matrix. Numerous studies have demonstrated that adding 1–5 wt% nanoparticles can increase PCM conductivity by 20–60%, significantly reducing melting and solidification times.<sup>[11]</sup> However, their high density can lead to sedimentation and phase separation in liquid PCMs, particularly during prolonged thermal cycling. Additionally, metallic additives may introduce corrosion concerns and significantly increase composite weight, limiting their suitability for lightweight or portable thermal storage systems.<sup>[26]</sup>

In addition, nanoparticle addition also introduces challenges. High concentrations may increase viscosity, reduce latent heat due to dilution, and cause sedimentation during cycling. Graphene and CNTs, while highly conductive, are expensive and may agglomerate without proper surfactants or functionalization. Despite these limitations, nanoparticle-enhanced PCMs (nano-PCMs) remain one of the most promising approaches due to their tunability and compatibility with both organic and inorganic PCMs.<sup>[54]</sup>

#### 4.3 Hybrid and Synergistic Conductivity Enhancement Strategies

To overcome the limitations associated with single-filler systems, recent studies have explored hybrid conductivity enhancement strategies that combine two or more additives with complementary properties. Hybrid systems, such as carbon–ceramic, carbon–metal

composites, and graphene aerogels provide lightweight, high-conductivity networks that significantly improve PCM performance.<sup>[32]</sup>

Synergistic effects observed in hybrid composites often result in improved thermal conductivity at lower filler loadings compared to mono-filler systems. This approach helps mitigate common drawbacks such as excessive latent heat reduction and poor mechanical integrity. Nevertheless, the complexity of composite design and the lack of standardized optimization methodologies remain significant challenges for reproducibility and scale-up.

#### **4.4 Incorporation of Fins, Extended Surfaces, and heat Exchanger Optimization.**

Geometric enhancement—using fins, extended surfaces, and optimized heat exchanger designs—is another widely adopted method for improving PCM thermal conductivity. Fins increase the effective heat transfer area and reduce conduction path length within the PCM. Studies show that finned PCM systems melt significantly faster than unfinned systems, even without material-level conductivity enhancement.

Advanced fin designs such as longitudinal fins, radial fins, dendritic fins, and fractal structures have been explored to maximize heat transfer. Shell-and-tube heat exchangers with optimized fin geometry are commonly used in latent heat storage systems for solar thermal applications. Although fins reduce PCM volume and may increase system weight, they remain one of the most reliable and cost-effective enhancement techniques.<sup>[61]</sup>

#### **4.5 Microencapsulation and Shape-Stabilized PCMS**

Microencapsulation involves coating PCM droplets with polymeric or inorganic shells, which improves heat transfer by increasing surface area and reducing conduction path length. Microcapsules also prevent leakage and improve cycling stability. Studies show that microencapsulated PCMs exhibit faster melting and solidification compared to bulk PCMs.<sup>[45]</sup>

Shape-stabilized PCMs—where PCM is embedded in a polymer matrix—also exhibit improved conductivity when conductive fillers are added. These materials are widely used in building applications due to their structural stability and ease of integration.

#### **4.6 Critical Assessment of Conductivity Enhancement Approaches**

Despite the demonstrated effectiveness of conductivity enhancement techniques, it is increasingly evident that thermal conductivity improvement alone does not guarantee

superior overall heat storage performance. Excessive additive loading can significantly reduce the effective latent heat capacity, alter phase transition behavior, and increase material viscosity, negatively affecting system efficiency.<sup>[62]</sup> Furthermore, most conductivity enhancement studies are conducted at the material level, with limited consideration of system-level performance metrics such as charging–discharging efficiency, pressure drop penalties, and long-term durability.

These limitations highlight the need for balanced optimization strategies that integrate conductivity enhancement with structural stabilization, encapsulation, and system-level design. Conductivity enhancement should therefore be regarded as one component of a broader performance optimization framework rather than an isolated objective.

## **5. Structural Stabilization and Leakage Prevention Strategies**

Leakage during the solid–liquid phase transition remains one of the most critical challenges limiting the practical deployment of phase change materials (PCMs) in heat storage systems. Structural instability not only leads to material loss but also degrades thermal performance, contaminates surrounding components, and compromises long-term reliability. To address these issues, extensive research has focused on structural stabilization strategies that preserve the shape and integrity of PCMs during repeated thermal cycling.

### **5.1 Shape-Stabilized Phase Change Materials**

Shape-stabilized phase change materials (SS-PCMs) are developed by embedding PCMs within solid supporting matrices that retain structural integrity even when the PCM melts. Common supporting materials include porous carbon structures, polymer frameworks, metal foams, and inorganic porous materials such as silica and clay-based matrices. These frameworks physically confine the PCM through capillary forces and surface tension, effectively preventing leakage while maintaining latent heat storage capability.<sup>[43,45]</sup>

Expanded graphite and carbon foam matrices have attracted particular attention due to their dual functionality: providing structural support while simultaneously enhancing thermal conductivity. However, excessive matrix content can reduce the effective PCM fraction, leading to diminished energy storage density. Polymer-based support offers mechanical flexibility and ease of processing but may introduce additional thermal resistance and long-term degradation concerns at elevated temperatures.<sup>[63]</sup>

## 5.2 Encapsulation Techniques

Encapsulation represents another widely adopted approach to mitigating leakage by enclosing PCMs within protective shells. Depending on capsule size and fabrication method, encapsulation strategies are typically categorized into macro-, micro-, and nano encapsulation. Macro-encapsulation involves packaging PCMs in containers such as tubes, spheres, or panels, offering high PCM loading and ease of system integration. However, heat transfer limitations and mechanical stresses during thermal cycling can reduce performance over time.<sup>[38]</sup>

Micro- and nano-encapsulation provide significantly higher surface area-to-volume ratios, improving heat transfer rates and enhancing thermal response. Shell materials such as polymers, silica, and hybrid organic–inorganic coatings are commonly employed to enhance chemical stability and mechanical strength. Despite these advantages, encapsulation often reduces the overall latent heat capacity due to the presence of inactive shell material and increases fabrication complexity and cost.<sup>[64-66]</sup>

## 5.3 Composite Architecture and Structural Integrity

Beyond simple confinement, recent research has emphasized the role of composite architecture in maintaining structural integrity under repeated thermal cycling. Hierarchical and multi-scale composite designs aim to optimize the distribution of PCM, conductive fillers, and structural supports to balance leakage prevention with thermal performance. Such architectures are particularly important for hybrid nanocomposites, where filler agglomeration or phase separation can undermine both stability and heat transfer efficiency.<sup>[7,62]</sup>

Mechanical robustness and dimensional stability are increasingly recognized as critical performance criteria, especially for compact and modular thermal energy storage systems. Failure to account for volume expansion, mechanical stress, and interface compatibility can result in microcracking, delamination, and progressive performance degradation during long-term operation.<sup>[67,68]</sup>

## 5.4 Critical Evaluation of Stabilization Strategies

While structural stabilization techniques have proven effective in preventing leakage, they introduce new trade-offs that must be critically evaluated. Shape stabilization and encapsulation inevitably reduce the effective PCM content, thereby lowering latent heat

storage density. Additionally, stabilizing matrices and encapsulation shells may act as thermal barriers, partially offsetting conductivity enhancement gains. The complexity of fabrication processes and scalability challenges further limit the widespread adoption of certain stabilization strategies.

These considerations reinforce the importance of adopting a holistic optimization framework in which leakage prevention is integrated with thermal conductivity enhancement, mechanical stability, and system-level design. Structural stabilization should therefore be viewed not as an isolated solution but as a complementary strategy within a broader performance optimization paradigm.

## **6. Hybrid and Multifunctional Phase Change Composites**

The growing complexity of performance requirements for phase change material (PCM)-based heat storage systems has driven a shift from single-additive modification toward hybrid and multifunctional composite designs. Hybrid phase change composites aim to simultaneously address multiple limitations, such as low thermal conductivity, leakage, mechanical instability, and durability through the synergistic combination of fillers, matrices, and structural supports. This approach represents a significant evolution beyond conventional, single-parameter optimization strategies.

### **6.1 Hybrid Filler Systems and Synergistic Effects**

Hybrid phase change composites typically incorporate two or more fillers with complementary physical and chemical properties. Common examples include carbon–ceramic (e.g., carbon nanotubes combined with aluminum oxide or boron nitride), carbon–carbon (e.g., graphene with expanded graphite), and carbon–metal oxide systems. These hybrid configurations are designed to lower the percolation threshold for thermal conductivity enhancement while minimizing adverse effects on latent heat capacity and phase transition behavior.<sup>[69,70]</sup>

Synergistic effects in hybrid composites arise from improved filler dispersion, enhanced interfacial heat transfer, and the formation of interconnected thermal networks at relatively low filler loadings. Compared to mono-filler systems, hybrid composites often achieve comparable or superior thermal conductivity enhancement with reduced material cost and improved structural stability. Nevertheless, the effectiveness of these systems is highly

sensitive to filler ratio, particle morphology, and processing conditions, underscoring the need for systematic optimization methodologies.

## 6.2 Multifunctional Performance Enhancement

Beyond thermal conductivity, hybrid phase change composites are increasingly engineered to exhibit multifunctional properties that enhance overall system performance. These properties may include improved mechanical strength, flame retardancy, electrical insulation, corrosion resistance, and resistance to phase segregation. For example, the incorporation of carbon fillers not only enhances thermal conductivity but also provides electrical insulation and thermal stability, making it suitable for applications involving electronic thermal management.<sup>[2,71.72]</sup>

In addition, multifunctional composites have been developed to address environmental and safety concerns, such as reducing flammability in organic PCMs or improving chemical compatibility with containment materials. However, the integration of multiple functionalities often introduces complex trade-offs, including increased composite heterogeneity, processing difficulty, and uncertainty in long-term reliability.

## 6.3 Hierarchical and Multi-Scale Composite Architectures

Recent studies have highlighted the importance of hierarchical and multi-scale architectures in achieving stable and efficient hybrid PCM composites. In such systems, micro- or nano-scale fillers enhance thermal conductivity, while macro-scale structures—such as porous frameworks or encapsulation shells, provide mechanical support and leakage prevention. This layered approach enables more precise control over composite behavior during phase transition and thermal cycling.<sup>[6]</sup>

Despite their promise, hierarchical composites remain challenging to manufacture at scale. Achieving uniform dispersion across multiple length scales requires advanced fabrication techniques and rigorous quality control, which can limit industrial adoption. Furthermore, standardized testing protocols for evaluating multifunctional performance remain largely absent.

## 6.4 Critical Assessment of Hybrid and Multifunctional Composites

While hybrid and multifunctional phase change composites represent a promising pathway toward high-performance heat storage systems, their practical implementation faces several

unresolved challenges. The lack of universally accepted design guidelines complicates material selection and performance prediction. In addition, many reported studies emphasize initial performance metrics without adequately addressing long-term thermal cycling stability, aging effects, or environmental impact.<sup>[7]</sup>

These limitations reinforce the need for holistic evaluation frameworks that integrate material performance, processing scalability, system-level efficiency, and sustainability considerations. Hybrid composite design should therefore be guided by application-specific requirements rather than isolated material enhancements. The next section builds on this perspective by examining how geometry and system-level design can further enhance heat transfer and overall thermal energy storage performance.

## **7. Heat Transfer Enhancement through Geometry and System Design**

While material-level modifications play a crucial role in improving the performance of phase change materials (PCMs), growing evidence indicates that system-level design and geometric optimization can deliver equal or greater performance gains, often without sacrificing latent heat capacity. Geometry-driven heat transfer enhancement addresses fundamental limitations associated with slow melting and solidification by improving thermal contact, increasing effective heat transfer area, and promoting more uniform temperature distribution within PCM-based thermal energy storage (TES) systems.

### **7.1 Extended Heat Transfer Surfaces**

The incorporation of extended heat transfer surfaces, such as fins, metal inserts, and porous structures, is one of the most widely adopted strategies for accelerating heat transfer in PCM systems. Fins—typically fabricated from high-conductivity metals such as aluminum or copper, enhance heat transfer by increasing surface area and reducing thermal resistance between the heat transfer fluid and the PCM. Various fin configurations, including longitudinal, radial, pin, and tree-like designs, have been investigated to optimize melting and solidification rates.<sup>[73,74]</sup>

Metal foams and porous inserts offer additional advantages by providing three-dimensional conductive networks that enhance heat transfer throughout the PCM volume. However, these structures often reduce the effective PCM fraction and may introduce pressure drop penalties in flow-assisted systems. The trade-off between enhanced heat transfer and reduced energy storage density remains a central design consideration.<sup>[73-75]</sup>

## 7.2 Container Geometry and PCM Configuration

Container geometry strongly influences heat transfer behavior, phase front evolution, and overall TES performance. Common configurations include shell-and-tube, cylindrical capsules, flat plates, and coil-based systems. Tube-based configurations are particularly attractive for solar thermal and air heater applications due to their modularity and ease of integration. Studies comparing horizontal and vertical tube arrangements have shown that orientation affects natural convection patterns during melting, leading to differences in charging rates and temperature uniformity.<sup>[76,77]</sup>

Helical and multi-pass coil geometries further enhance heat transfer by increasing contact length and inducing secondary flow effects in the heat transfer fluid. However, increased geometric complexity can complicate fabrication and maintenance, emphasizing the need for application-specific optimization rather than universal design solutions.<sup>[26]</sup>

## 7.3 Influence of Operating Conditions

Operating conditions, including heat transfer fluid velocity, flow regime, and thermal boundary conditions, play a decisive role in determining the effectiveness of geometric enhancements. Higher flow rates generally improve heat transfer coefficients but may lead to increased pumping power and reduced overall system efficiency. Conversely, low flow rates can result in incomplete PCM melting or solidification, particularly in systems with poor internal heat distribution.<sup>[38]</sup>

The interaction between forced and natural convection within the PCM during phase transition further complicates system behavior. In many cases, optimal performance is achieved through dynamic or adaptive control strategies that adjust operating conditions in response to the PCM state, rather than fixed operating parameters.

## 7.4 System-Level Integration and Design Trade-Offs

System-level integration of PCM-based TES units requires careful consideration of geometric design, operating conditions, and application-specific constraints. Enhancements that significantly improve heat transfer may simultaneously increase system weight, volume, or cost, potentially offsetting performance gains. Additionally, many geometric optimization studies prioritize short-term thermal response without adequately addressing long-term durability, mechanical stress, or compatibility with real operating environments.<sup>[38,76]</sup>

These considerations reinforce the importance of holistic design approaches that integrate geometry, material properties, and operating strategies. Geometry-driven heat transfer enhancement should therefore be evaluated not only in terms of melting and solidification rates but also with respect to overall system efficiency, scalability, and reliability. The following section extends this discussion by examining long-term reliability and thermal cycling performance of PCM-based systems.

## **8. Long-Term Reliability, Durability, and Thermal Cycling Performance**

Long-term reliability is a decisive factor governing the practical viability of phase change materials (PCMs) for heat storage applications. While many PCM systems demonstrate promising initial thermal performance, their widespread adoption has been limited by performance degradation under prolonged thermal cycling, chemical instability, and mechanical failure. These durability concerns are particularly critical for applications such as solar thermal energy storage and building-integrated systems, where PCMs are expected to operate reliably over thousands of charges–discharge cycles. This section examines the major dimensions of long-term PCM reliability, including thermal cycling stability, chemical compatibility, structural fatigue, evaluation methodologies, and broader implications for holistic optimization.

### **8.1 Thermal Cycling Stability**

Thermal cycling stability refers to the ability of a PCM to maintain its thermophysical properties such as latent heat, melting point, thermal conductivity, and phase transition behavior—over hundreds or thousands of melt–freeze cycles. Repeated cycling can induce changes in PCM composition, microstructure, and phase behavior, ultimately reducing storage capacity and system efficiency. Recent studies show that organic PCMs such as paraffins generally exhibit good cycling stability but may still experience gradual reductions in latent heat due to oxidation or minor structural rearrangements.<sup>[32]</sup> In contrast, inorganic PCMs, particularly salt hydrates, are more prone to degradation due to phase segregation, supercooling, and incongruent melting.

Long-term cycling tests reveal that even small deviations in melting temperature or enthalpy can accumulate over time, leading to significant performance losses in TES systems. For example,<sup>[78]</sup> demonstrated that salt hydrate PCMs can lose up to 15–20% of their latent heat after extended cycling due to water migration and crystallization inconsistencies. Composite

PCMs incorporating stabilizers or porous matrices show improved cycling stability, but their performance still depends heavily on encapsulation quality and operating conditions.

Thermal cycling stability is also influenced by system-level factors such as heat transfer geometry, charging/discharging rates, and thermal gradients. High cycling frequencies, rapid temperature swings, and incomplete solidification can accelerate degradation. Therefore, evaluating PCM cycling stability under realistic operating conditions is essential for predicting long-term performance in solar thermal systems, building envelopes, and industrial heat recovery applications.<sup>[45]</sup>

## 8.2 Chemical Compatibility and Material Degradation

Chemical compatibility between PCM and containment materials is a major determinant of long-term reliability. Inorganic PCMs, especially salt hydrates, can be highly corrosive to metals such as aluminum, copper, and steel. Corrosion not only weakens containment structures but also contaminates PCM, altering its melting behavior and reducing thermal conductivity. Recent investigations show that corrosion rates can increase significantly under cyclic thermal loading due to repeated expansion, contraction, and moisture migration.<sup>[79]</sup>

Chemical degradation can also occur within the PCM itself. Organic PCMs may undergo oxidation, polymerization, or thermal decomposition when exposed to elevated temperatures or impurities. These reactions can shift melting points, reduce latent heat, and alter viscosity. Salt hydrates may lose water of crystallization, leading to irreversible changes in phase composition and reduced energy storage capacity. Studies by<sup>[61]</sup> highlight that even minor moisture loss in salt hydrates can cause substantial reductions in latent heat and increased supercooling.

Encapsulation material polymers, metals, ceramics also face chemical degradation challenges. Polymer shells may soften, crack, or react with PCM components, while metal containers may corrode or form surface oxides. Composite PCMs incorporating nanoparticles or carbon materials may experience chemical interactions that alter dispersion stability or thermal conductivity.<sup>[80]</sup> Thus, chemical compatibility must be evaluated holistically, considering PCM chemistry, containment materials, additives, and operating environment.

### 8.3 Mechanical Integrity and Structural Fatigue

Mechanical integrity is essential for maintaining PCM containment, heat transfer efficiency, and structural stability. During phase change, PCMs undergo volumetric expansion and contraction, generating cyclic mechanical stresses on encapsulation shells, heat exchanger surfaces, and composite matrices. Over time, these stresses can lead to fatigue, cracking, delamination, and structural failure. Recent studies show that microencapsulated PCMs often experience shell fatigue after several hundred cycles, resulting in leakage and performance degradation.<sup>[45,73,78]</sup>

Bulk PCMs embedded in building materials or composite structures may also suffer from mechanical instability. Salt hydrates can form brittle crystalline phases that fracture under thermal cycling, while organic PCMs may soften or deform polymeric matrices.<sup>[79]</sup> reported that PCM-enhanced gypsum boards exhibited microcracking and loss of structural cohesion after extended cycling, reducing both thermal performance and mechanical strength.

Mechanical fatigue is exacerbated by thermal gradients, rapid cycling, and poor encapsulation design. Systems with rigid containment structures are particularly vulnerable, as they cannot accommodate PCM expansion without generating high stresses. Flexible or shape-stabilized PCMs offer improved mechanical resilience but may require additional reinforcement or conductive fillers to maintain thermal performance.<sup>[23]</sup> Therefore, mechanical integrity must be considered alongside thermal and chemical stability when designing PCM systems.

### 8.4 Evaluation Methods and Standardization Challenges

Despite extensive research, standardized methods for evaluating PCM reliability remain limited. Current testing protocols vary widely in cycling duration, temperature range, heating/cooling rates, and performance metrics, making it difficult to compare results across studies. Many researchers conduct cycling tests under idealized laboratory conditions that do not reflect real-world thermal loads, leading to discrepancies between predicted and actual performance.<sup>[32]</sup>

Standardization challenges also arise from the diversity of PCM types—organic, inorganic, eutectic, composites and their unique degradation mechanisms. For example, salt hydrates require tests for phase segregation and supercooling, while organic PCMs require oxidation and flammability assessments. Encapsulated PCMs require additional evaluation of shell

integrity, leakage resistance, and mechanical fatigue. Recent efforts by international research groups aim to establish unified testing protocols, but consensus remains limited due to the complexity of PCM behavior.<sup>[79]</sup>

Advanced characterization techniques such as differential scanning calorimetry (DSC), thermogravimetric analysis (TGA), X-ray diffraction (XRD), and micro-CT imaging have improved understanding of PCM degradation, but their use is not yet standardized. Long-term field testing in buildings, solar collectors, and industrial systems is increasingly recognized as essential for validating laboratory results.<sup>[45]</sup> Thus, establishing robust, standardized evaluation frameworks remains a critical challenge for PCM research and commercialization. Lack of standardized durability metrics complicates material selection and system design, underscoring the need for harmonized testing methodologies that incorporate thermal, chemical, and mechanical degradation indicators. Establishing such standards would significantly enhance the comparability and reliability of reported PCM performance data.

### 8.5 Implications for Holistic Performance Optimization

The long-term reliability challenges discussed above highlight the need for holistic optimization strategies that integrate material selection, encapsulation design, system architecture, and operating conditions. Enhancing PCM durability requires balancing thermal performance with chemical stability, mechanical resilience, and cost. Composite PCMs incorporating expanded graphite, metal foams, or nanoparticles offer improved conductivity and stability, but may reduce latent heat or increase cost.<sup>[81]</sup>

System-level optimization such as fin design, heat exchanger geometry, and controlled charging/discharging profiles can mitigate thermal gradients and reduce mechanical fatigue. Chemical stabilization strategies, including nucleating agents, thickening additives, and corrosion inhibitors, can improve long-term stability of salt hydrates. Encapsulation innovations such as flexible shells, hybrid composites, and multi-layer barriers can enhance mechanical and chemical resilience.<sup>[57]</sup>

Ultimately, achieving long-term PCM reliability requires integrated approaches that consider the interplay between thermal, chemical, mechanical, and operational factors. As recent studies emphasize, PCM systems must be optimized not only for initial performance but also for sustained operation over thousands of cycles in real-world environments.<sup>[23]</sup> Holistic

optimization is therefore essential for advancing PCM-based TES technologies toward widespread commercial adoption.

## 9. Sustainability and Environmental Considerations

As phase change materials (PCMs) transition from laboratory-scale development to large-scale deployment in thermal energy storage systems, sustainability and environmental impact have become increasingly important evaluation criteria. Beyond thermal performance, the environmental footprint of PCM production, modification, deployment, and end-of-life management plays a critical role in determining the long-term viability and societal acceptance of PCM-based heat storage technologies.

### 9.1 Material Availability and Resource Sustainability

The sustainability of PCM-based systems is strongly influenced by the availability and sourcing of constituent materials. Organic PCMs, particularly paraffin-based materials derived from petroleum resources, raise concerns regarding resource depletion and long-term sustainability. While these materials offer favorable thermal reliability and cost advantages, their fossil-based origin motivates the exploration of alternative bio-based or waste-derived PCMs.<sup>[26,38]</sup>

In contrast, inorganic PCMs and advanced composite additives often rely on resource-intensive extraction and processing routes. Carbon nanomaterials, metal oxides, and ceramic fillers may significantly increase the environmental burden associated with PCM production, particularly when used in high loadings. Consequently, material selection should consider not only thermal performance but also resource efficiency and supply-chain sustainability.

### 9.2 Environmental Impact of Additives and Composite Formulations

The incorporation of conductive fillers and stabilizing matrices can substantially alter the environmental profile of PCM composites. Nanomaterials such as carbon nanotubes and graphene, while highly effective for thermal conductivity enhancement, are associated with energy-intensive synthesis processes and potential environmental and health risks during manufacturing and disposal.<sup>[7,11,45]</sup>

Hybrid and multifunctional composites further increase material complexity, complicating recycling and end-of-life management. The lack of established recycling pathways for composite PCMs presents a significant sustainability challenge, particularly for systems

designed for long-term operation in building or solar thermal applications. Reducing additive loading through synergistic hybrid designs and prioritizing environmentally benign fillers are therefore essential strategies for minimizing environmental impact.

### 9.3 Life Cycle Assessment of PCM-Based Thermal Energy Storage Systems

Life cycle assessment (LCA) has emerged as a critical tool for quantifying the environmental performance of PCM-based thermal energy storage systems. Recent LCA studies indicate that while PCM integration can significantly reduce operational energy consumption and greenhouse gas emissions, the benefits may be partially offset by the environmental cost of material production and system fabrication.<sup>[82,83]</sup>

Importantly, system-level benefits are highly sensitive to PCM lifespan and thermal reliability. Short service life or premature degradation can negate environmental advantages by increasing material replacement frequency and associated resource consumption. These findings reinforce the importance of durability and long-term stability as sustainability-enabling performance metrics.

### 9.4 Recyclability and End-of-Life Considerations

End-of-life management remains a largely underexplored aspect of PCM research. Conventional PCMs are often not designed with recyclability in mind, and composite formulations may hinder material separation and recovery. Encapsulation shells, polymer matrices, and hybrid filler systems further complicate recycling processes, increasing the likelihood of landfill disposal or incineration.<sup>[3,24,53]</sup>

Design-for-recycling approaches, including modular system architectures and reversible encapsulation techniques, represent promising pathways toward more sustainable PCM deployment. However, these concepts remain at an early stage of development and require greater attention in future research.

### 9.5 Toward Sustainable PCM Design

Achieving sustainable PCM-based thermal energy storage requires an integrated approach that balances thermal performance, durability, cost, and environmental impact. Holistic optimization frameworks should therefore incorporate sustainability metrics alongside traditional thermophysical performance indicators. Emerging research directions include the

development of bio-based PCMs, low-impact fillers, and composite designs that minimize material complexity while maintaining performance.<sup>[78]</sup>

By embedding sustainability considerations into the early stages of material and system design, PCM technologies can better support long-term energy transition goals. The following section builds on this perspective by examining emerging trends and future research directions aimed at advancing high-performance, sustainable PCM-based heat storage systems.

## **10. Emerging Trends and Future Directions**

The rapid evolution of thermal energy storage requirements, driven by renewable energy integration, electrification, and sustainability goals, has stimulated the emergence of advanced strategies for phase change material (PCM) development and deployment. Recent research increasingly recognizes that incremental improvements in individual material properties are insufficient to meet future performance demands. Instead, emerging trends emphasize system integration, multifunctionality, intelligent design, and scalability.

### **10.1 Additive Manufacturing and Advanced Fabrication Techniques**

Additive manufacturing (AM) has emerged as a promising tool for the design and fabrication of PCM-based thermal energy storage systems with enhanced geometric complexity and performance control. Three-dimensional printing enables the creation of customized PCM containers, lattice structures, and graded architectures that optimize heat transfer pathways while maintaining high PCM loading. Such designs are difficult or impossible to achieve using conventional manufacturing methods.<sup>[23]</sup>

AM-enabled PCM systems allow precise control over internal geometry, facilitating tailored melting and solidification behavior and improved system-level efficiency. However, challenges related to material compatibility, printing resolution, and long-term mechanical stability remain significant barriers to large-scale adoption. Future research should focus on printable composite formulations and robust design methodologies that link geometric parameters to thermal performance.

### **10.2 Smart and Adaptive Phase Change Materials**

Smart PCM systems that respond dynamically to operating conditions represent an emerging frontier in thermal energy storage research. These systems integrate PCMs with sensing

elements, adaptive control strategies, or functional additives that enable real-time monitoring and optimization of thermal behavior. For example, thermochromic or electrically responsive additives can provide feedback on PCM state or enhance controllability of heat transfer processes.<sup>[35,84]</sup>

The integration of artificial intelligence and data-driven optimization techniques is expected to further advance smart PCM systems. Machine learning models can be employed to predict phase transition behavior, optimize composite formulations, and adapt operating conditions to maximize storage efficiency and durability. Despite their potential, smart PCM technologies remain largely at the conceptual or early experimental stage.

### 10.3 Hybrid Energy Storage and Multi-Functional Integration

Future thermal energy storage systems are increasingly envisioned as components of hybrid energy systems that integrate thermal, electrical, and mechanical storage functionalities. PCMs can be combined with thermoelectric devices, photovoltaic systems, or batteries to enhance overall energy system flexibility and efficiency. In such hybrid configurations, PCMs play a dual role by providing thermal regulation and energy buffering.<sup>[38]</sup>

Multifunctional PCM composites capable of simultaneous heat storage, thermal management, and structural support are also gaining attention. These systems challenge conventional design paradigms by requiring performance optimization across multiple domains, reinforcing the need for holistic evaluation frameworks.

### 10.4 Toward Application-Driven and Scalable PCM Design

A key future direction for PCM research lies in shifting from material-centric studies toward application-driven design. Many promising PCM formulations fail to progress beyond laboratory-scale demonstrations due to scalability, cost, or integration challenges. Future studies should prioritize system-level validation, long-term field testing, and techno-economic analysis to accelerate technology readiness.<sup>[32,84]</sup>

Standardization of performance metrics and testing protocols will be essential for enabling meaningful comparison across studies and facilitating industrial adoption. Collaborative efforts between materials scientists, thermal engineers, and system designers are therefore critical for advancing PCM technologies toward widespread implementation.

## 10.5 Outlook

Emerging trends in PCM-based heat storage increasingly emphasize integration, intelligence, and sustainability. The convergence of advanced materials, innovative fabrication techniques, and data-driven optimization offers significant opportunities for performance enhancement. However, realizing this potential will require moving beyond isolated material improvements toward comprehensive design strategies that balance performance, durability, cost, and environmental impact. These considerations form the basis for the research gaps and recommendations discussed in the next section.

## 11. Research Gaps and Recommendations

Despite significant progress in the development of phase change materials (PCMs) and phase change composites for thermal energy storage, several critical research gaps continue to limit their transition from laboratory-scale innovations to robust, application-ready technologies. These gaps span material design, performance evaluation, system integration, and sustainability considerations, underscoring the need for a coordinated and holistic research approach.

### 11.1 Overemphasis on Thermal Conductivity Enhancement

A dominant trend in PCM research remains the prioritization of thermal conductivity enhancement as the primary performance objective. While improved conductivity accelerates charging and discharging, excessive focus on this single parameter has often resulted in reduced latent heat capacity, compromised phase transition behavior, and increased material cost. Many studies report conductivity gains without adequately quantifying the associated trade-offs in energy storage density, mechanical stability, or durability.<sup>[62,85]</sup>

#### Recommendation

Future research should adopt multi-objective optimization frameworks that simultaneously evaluate thermal conductivity, latent heat retention, phase stability, and long-term cycling performance. Conductivity enhancement should be treated as one component of an integrated performance metric rather than a standalone target.

### 11.2 Lack of Standardized Testing Protocols

The absence of standardized methodologies for evaluating PCM performance remains a major barrier to meaningful comparison across studies. Variations in thermal cycling conditions, heating-cooling rates, temperature ranges, and performance metrics result in

inconsistent and sometimes contradictory conclusions. Durability assessments often rely on limited cycling tests that do not reflect realistic operational lifetimes.<sup>[3]</sup>

### **Recommendation**

There is a pressing need for standardized testing protocols that encompass thermophysical properties, thermal cycling stability, mechanical integrity, and system-level efficiency. Establishing benchmark testing conditions would significantly enhance data reliability and accelerate technology validation.

### **11.3 Insufficient Emphasis on Long-Term Reliability**

Many promising PCM formulations demonstrate excellent initial performance but suffer from gradual degradation under extended thermal cycling. Issues such as filler agglomeration, phase segregation, chemical incompatibility, and structural fatigue are often underreported or insufficiently analyzed. This shortcoming undermines confidence in long-term deployment, particularly for solar thermal and building-integrated systems expected to operate for decades.<sup>[12]</sup>

### **Recommendation**

Long-term reliability should be elevated to a core design criterion. Future studies should incorporate extended cycling tests, degradation mechanism analysis, and predictive lifetime modeling to assess performance sustainability under realistic operating conditions.

### **11.4 Limited System-Level and Application-Oriented Studies**

A significant proportion of PCM research remains confined to material-level characterization, with limited validation in full-scale or application-relevant systems. As a result, many materials optimized in isolation fail to deliver anticipated benefits when integrated into real thermal energy storage systems due to geometric, operational, or control-related constraints.<sup>[7,26]</sup>

### **Recommendation**

Future research should prioritize system-level demonstrations that integrate material optimization with geometry, heat exchanger design, and operating strategy. Application-driven design—rather than material-centric optimization should guide PCM development.

### 11.5 Sustainability and End-of-Life Considerations Remain Underexplored

Although PCMs are widely promoted as enabling technologies for energy sustainability, relatively few studies rigorously assess their environmental footprint across the full life cycle. The environmental impact of nanomaterials, composite complexity, and limited recyclability is often overlooked, potentially offsetting the sustainability benefits of PCM-based energy storage systems.<sup>[84]</sup>

#### Recommendation

Life cycle assessment (LCA) and design-for-recycling principles should be integrated into PCM research from the early stages of material selection and composite design. Sustainable PCM development should balance thermal performance with environmental responsibility and resource efficiency.

### 11.6 Need for Holistic Design Frameworks

The fragmentation of PCM research into isolated subdomains—thermal conductivity enhancement, encapsulation, geometry optimization, or sustainability—has limited the development of comprehensive design strategies. The lack of unified frameworks hinders the translation of research findings into scalable, reliable technologies.

#### Recommendation

Future efforts should focus on developing holistic design frameworks that integrate material properties, composite architecture, system geometry, operating conditions, durability, and sustainability. Such frameworks are essential for guiding the rational design of next-generation PCM-based thermal energy storage systems.

### 11.7 Outlook for Future Research

Addressing these research gaps will require interdisciplinary collaboration among materials scientists, thermal engineers, system designers, and sustainability experts. Advances in data-driven optimization, additive manufacturing, and smart thermal management offer promising tools for overcoming existing limitations. By shifting the research paradigm from isolated performance enhancement to holistic system optimization, PCM technologies can more effectively support future energy transition goals.

## 12. CONCLUSIONS

Phase change materials (PCMs) continue to play a pivotal role in advancing thermal energy storage technologies due to their high latent heat storage capacity and near-isothermal phase transition behavior. However, as demonstrated throughout this review, the performance of PCM-based heat storage systems cannot be effectively optimized through thermal conductivity enhancement alone. A narrow focus on conductivity improvement often introduces trade-offs that undermine latent heat capacity, structural stability, durability, and long-term system efficiency.

This critical review has adopted a holistic perspective to examine performance optimization strategies for PCMs, encompassing material modification, composite architecture, structural stabilization, encapsulation, geometric design, operating conditions, durability, and sustainability considerations. The analysis highlights that meaningful performance gains arise from the integration of complementary strategies rather than isolated material-level interventions. Hybrid and multifunctional phase change composites, when thoughtfully designed, offer promising pathways to balance thermal conductivity enhancement with leakage prevention, mechanical integrity, and reliable phase transition behavior.

System-level design and geometric optimization were shown to be equally influential in determining heat transfer efficiency and charging–discharging performance. The incorporation of extended heat transfer surfaces, optimized container geometries, and adaptive operating strategies can significantly enhance thermal response without sacrificing energy storage density. Moreover, long-term reliability and thermal cycling stability emerged as critical determinants of practical viability, reinforcing the need for durability-focused design and standardized testing methodologies.

Sustainability considerations further underscore the importance of holistic optimization. The environmental impact of PCM production, additive incorporation, and end-of-life management must be evaluated alongside thermal performance to ensure that PCM-based heat storage systems genuinely contribute to long-term energy transition goals. Emerging trends such as additive manufacturing, smart PCM systems, and hybrid energy storage integration offer exciting opportunities but also demand rigorous system-level validation.

In conclusion, the future development of PCM-based thermal energy storage technologies depends on shifting from isolated performance enhancement toward integrated, application-

driven design frameworks. By balancing thermal performance, durability, scalability, and sustainability, holistic optimization strategies can bridge the gap between laboratory-scale innovation and reliable, real-world deployment. This review provides a comprehensive foundation to guide future research and support the rational design of high-performance phase change materials for advanced heat storage applications.

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