

Research progress on performance optimization of organic phase change materials

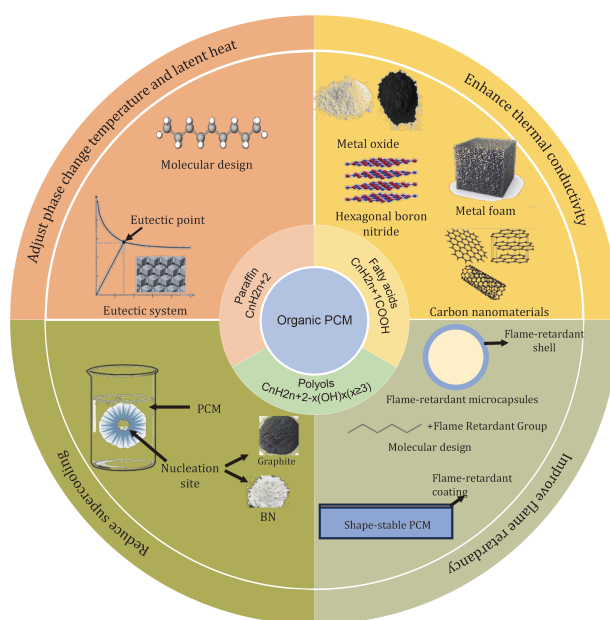
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Abstract: Organic phase change materials (PCMs), known for their excellent energy storage properties, have been widely applied in thermal management and energy storage fields. This review summarizes the main types and performance optimization strategies of organic PCMs. In terms of thermophysical property regulation, methods such as constructing eutectic systems, modifying molecular structures and incorporating nanoparticles to adjust phase change temperature and latent heat are introduced. For thermal conductivity enhancement, strategies involving composites with zero-, one-, two- and three-dimensional high thermal conductivity materials are reviewed. Additionally, countermeasures for common issues like supercooling and flammability of organic PCMs are analyzed, including methods to suppress supercooling and flame-retardant modification techniques. Finally, future development directions of organic PCMs are discussed, highlighting their broader application prospects in energy storage and thermal management as technology advances.

Key words: organic phase change materials; latent heat; thermal conductivity; flame retardancy



Phase change materials (PCMs) are a type of materials that store and release energy by absorbing or releasing heat when a substance undergoes a phase change at a specific temperature. Unlike sensible heat storage materials that rely on temperature gradients for energy accumulation, PCMs facilitate high-density energy management through nearly isothermal processes, thereby enabling more efficient thermal buffering and regulation^[1-2]. Generally, PCMs are categorized into organic and inorganic variants. Compared to the inorganic counterparts, organic PCMs possess exceptional chemical inertness, non-corrosivity and long-term cycling robustness^[3]. Crucially, organic PCMs exhibit

minimal subcooling and are inherently free from phase segregation—common bottlenecks for inorganic systems. Furthermore, organic PCMs align with green chemistry principles due to their low toxicity and biodegradability, while offering modular tunability of phase transition temperatures across a broad spectrum through eutectic or mixture engineering^[4].

This review provides a comprehensive synthesis of the classifications, fundamental properties, and key performance metrics of organic PCMs, with a particular focus on recent paradigms for their performance optimization.

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1 Types of organic PCMs

Organic PCMs exhibit extensive application potential in thermal energy storage and temperature regulation due to their diverse compositions, tunable phase change temperature ranges and high latent heat. However, the significant differences in molecular structure, phase change behavior and thermophysical properties among different organic PCMs have greatly influenced their performance and applicability. Therefore, it is essential to systematically categorize these materials and outline their characteristics. Based on their chemical compositions, organic PCMs are classified into three categories: paraffins, polyols and fatty acids. The fundamental features of each category are summarized.

1.1 Paraffins

Based on their chemical composition, paraffin PCMs can be divided into two categories: pure linear alkanes with the general formula C_nH_{2n+2} and hydrocarbon mixtures^[5]. The latter is more cost-effective than pure alkanes and is widely utilized for energy storage, as its melting point can be tailored by blending alkanes with varying chain lengths^[6]. As the number of carbon atoms increases, the melting temperature of paraffin exhibits an upward trend, providing a highly flexible phase change temperature range^[7]. Furthermore, paraffins with self-nucleating behavior can be resistant to severe supercooling or phase separation, making them as ideal PCMs. However, their intrinsically low thermal conductivity severely restricts the rates of thermal energy absorption and release^[8–10]. The performance parameters of representative linear alkane PCMs are summarized in Table 1^[11].

Paraffin-based PCMs play a crucial role across various sectors due to their efficient heat storage capabilities. In the construction industry, they are integrated into building materials to regulate indoor temperatures and reduce heating energy consumption. In solar energy utilization, they serve as heat storage media in solar water heating or space heating systems, storing surplus solar energy during the day for the utilization at night or on cloudy days, thereby enhancing overall solar utilization efficiency^[12]. Regarding the thermal management of electronic devices, paraffins can rapidly absorb transient peak heat generated by electronic components (e.g., CPUs, batteries), effectively preventing thermal runaway^[13]. Additionally, paraffin PCMs are employed in cold chain logistics to maintain stable low-temperature environments in cold storage or insulated containers^[14]. Kahwaji et al.^[6] evaluated six paraffin PCMs with melting points ranging from 30 to 60 °C highlighting their potential for building applications, solar thermal storage, and passive cooling-by investigating their thermophysical properties, thermal stability and chemical compatibility. The results demonstrated high latent heats of fusion across all materials, ranging from 150–200 J/g for commercial paraffins and exceeding 200 J/g for pure alkanes. Long-term cycling tests revealed that both pure paraffins and mixtures maintained excellent thermal stability after 3000 melt-freeze cycles, with no significant degradation in melting point or latent heat. Chemical compatibility with nine metal alloys and seven plastics was assessed *via* corrosion immersion tests. While all tested metals exhibited good compatibility with paraffin, Cu 101 and Mg AZ91D samples displayed oxidation after 12 weeks of immersion at 75 °C. Certain plastics (e.g., polycarbonate, Type I PVC, and ABS) demonstrated good compatibility at 75 °C, whereas silicone rubber and polypropylene were deemed unsuitable due to excessive PCM absorption.

Table 1 Properties of *n*-alkane PCMs with different chain lengths

Formula	Melting temperature/°C	Melting enthalpy/(J·g ⁻¹)	Thermal conductivity/(W·m ⁻¹ ·K ⁻¹)
C ₁₄ H ₃₀	5.5	228	0.129
C ₁₆ H ₃₄	16.7	237	0.130
C ₁₈ H ₃₈	28.0	244	0.138
C ₂₀ H ₄₂	36.7	246	0.137
C ₂₂ H ₄₆	44	249	0.164
C ₂₄ H ₅₀	50.6	255	0.156
C ₂₆ H ₅₄	56.3	256	0.146
C ₂₈ H ₅₈	61.6	253	0.147

1.2 Polyols

Polyols are compounds containing two or more hydroxyl groups per molecule. Due to the unique molecular structures, polyols exhibit excellent thermal stability, environmental friendliness, and high phase change heat capacity. Common polyol PCMs include polyethylene glycol (PEG), neopentyl glycol (NPG), pentaerythritol (PE),

tris(hydroxymethyl)aminomethane (TAM) and sugar alcohols (SAs).

As an organic PCM, PEG holds the characterization of high phase change enthalpy, chemical and thermal stability, biodegradability, non-toxicity, non-corrosiveness and low vapor pressure. Notably, its properties can be precisely tailored by controlling its molecular weight^[15]. Xu et al.^[16] prepared bio-based composite PCMs (CPCMs) by using

diatomite as the supporting matrix, expanded graphite (EG) as an additive and PEG as the phase change medium. With an additive mass fraction (w) of 10%, the composite exhibited a melting enthalpy of 116.81 J/g and a crystallization enthalpy of 118.44 J/g. Gao et al.^[17] fabricated shape-stabilized PCMs *via* vacuum impregnation, using PEG as the energy storage material to yield sunflower stem carbon/PEG (SS-PEG) and sunflower receptacle carbon/PEG (SR-PEG) composites. The melting enthalpies of SS-PEG and SR-PEG were 153.4 and 171.5 J/g, with initial thermal decomposition temperatures of 344 and 368 °C, respectively.

NPG, PE and TAM are typically preferred as solid-solid PCMs. During the solid-solid phase change, they exhibit high enthalpies of transition and possess inherent advantages, including minimal volume variation, absence of liquid or gas generation, zero leakage risk and the ability to be molded into desired shapes^[18]. Wang et al.^[19] fabricated composite PCMs by integrating NPG with compressed expanded natural graphite (CENG) of varying densities. DSC results indicated that the latent heat of the NPG/CENG composites increased proportionally with the NPG mass ratio, achieving values between 75 and 106 J/g. Wang et al.^[20] pioneered the encapsulation of PE by using silicon dioxide (SiO₂) as a shell material *via* sol-gel technology to form a spherical core-shell structure. The PE-SiO₂ composite presented an average size of 0.3 μm, an encapsulation efficiency exceeding 77.8% and a latent heat reaching 255 kJ/kg. After 100 heating-cooling cycles, the latent heat degraded by less than 5%.

The history of sugar alcohols as PCMs is relatively brief,

with the first application in thermal energy storage dating back to 1981^[21]. Compared to conventional PCMs (e.g., paraffins), sugar alcohols possess higher volumetric latent heat. Derived from natural sources, they are non-flammable, non-toxic, non-corrosive, and cost-effective. As a new generation of PCMs^[22], they demonstrate great potential. Despite their favorable thermal properties, certain sugar alcohols exhibit severe supercooling during crystallization. Shao et al.^[23] utilized DSC to measure the degree of supercooling for erythritol, *d*-mannitol, *d*-dulcitol and myo-inositol, recording values of 89, 48, 66 and 38 °C, respectively. This pronounced supercooling phenomenon prevents the timely release of stored latent heat, causing temperature mismatch during the charge-discharge cycles and subsequently reducing thermal energy utilization efficiency^[24]. Regarding thermal stability, Kaizawa et al.^[25] employed thermogravimetric analysis to determine the thermal decomposition points of erythritol, mannitol, xylitol, and sorbitol, which were 160, 280, 200 and 240 °C, respectively. Tomassetti et al.^[26] reviewed the thermophysical properties and thermal stability of six SAs (xylitol, sorbitol, erythritol, mannitol, inositol, and dulcitol). Literature provides several thermal stability analyses for erythritol and mannitol, revealing that erythritol exhibits the highest thermal stability, whereas mannitol remains unstable even under an inert atmosphere. The evaluation of other SAs remains highly limited, and existing literature reports inconsistent findings regarding their thermal stability. The phase change parameters of selected sugar alcohols are detailed in Table 2^[27].

Table 2 Performance parameters of sugar alcohols as PCMs

Classification	Formula	Melting temperature/°C	Melting enthalpy/(J·g ⁻¹)	Volumetric energy density/(kW·h·m ⁻³)
Erythritol	C ₄ H ₁₀ O ₄	118.9	332.3	133.9
Xylitol	C ₅ H ₁₂ O ₅	93.4	237.5	100.2
Inositol	C ₆ H ₁₂ O ₆	224.3	256.3	124.6
Mannitol	C ₆ H ₁₄ O ₆	166.1	281.1	124.5
Sorbitol	C ₆ H ₁₄ O ₆	99.4	184.4	34.9
Galactitol	C ₆ H ₁₄ O ₆	186-187	330.0	117.5

1.3 Fatty acids

Fatty acids, with the general formula C_{*n*}H_{2*n*+1}COOH, can be extracted from biomass or synthesized chemically. Their amphiphilic molecular structure facilitates excellent interfacial interactions with most functional fillers. They possess moderate latent heats of fusion, and their phase change temperatures can be systematically adjusted by altering the fatty acid composition. The presence of polar functional groups in fatty acid PCMs promotes hydrogen

bond formation, enhancing their compatibility with other materials and thereby improving their performance in composite systems^[28]. Common PCMs within the fatty acid family include capric acid, myristic acid, lauric acid, palmitic acid and stearic acid. Their primary advantages include high latent heat, robust thermal stability, zero supercooling, and low cost^[29]. Xie et al.^[30] measured the phase change parameters, thermal stability, and thermal conductivity of a series of fatty acids (octanoic, decanoic, dodecanoic, tetradecanoic, hexadecanoic, and octadecanoic acids). The corresponding results are presented in Table 3.

Table 3 Performance parameters of fatty acids as PCMs

Formula	Melting temperature/°C	Melting enthalpy/(J·g ⁻¹)	Thermal conductivity/(W·m ⁻¹ ·K ⁻¹)	Maximum operational temperature/°C
C ₈ H ₁₆ O ₂	16.2	154.1	0.1872	153.6
C ₁₀ H ₂₀ O ₂	28.8	172.9	0.2133	163.7
C ₁₂ H ₂₄ O ₂	43.1	192.9	0.2388	180.7
C ₁₄ H ₂₈ O ₂	53.6	213.3	0.2662	203.5
C ₁₆ H ₃₂ O ₂	61.4	222.9	0.2881	208.2
C ₁₈ H ₃₆ O ₂	68.2	229.4	0.3079	224.2

2 Performance optimization of organic PCMs

Organic PCMs (including paraffins, polyols and fatty acids) with distinct characteristics in chemical composition, phase change mechanisms, and thermophysical properties enable them to fulfill diverse application requirements ranging from low-temperature energy storage to medium-temperature thermal management. However, the practical implementation is hindered by several limitations, such as inflexible phase change temperatures, insufficient latent heat, inherently low thermal conductivity, pronounced supercooling and safety risks associated with flammability. Consequently, utilizing these materials in their pristine state is insufficient for engineering applications. It is imperative to systematically enhance their performance through multi-scale material design strategies. This section reviews current primary performance optimization strategies for organic PCMs, focusing on phase change behavior regulation, thermal conductive network construction, nucleation site introduction and flame-retardant technology optimization.

2.1 Phase transition temperature and latent heat regulation

The most critical parameters of a PCM are phase change temperature and latent heat. The phase change temperature dictates the applicable scenarios for the PCMs, such as cold storage (7–15 °C), space heating equipment (40–50 °C), and solar cooling and heating (80–90 °C)^[31]. High latent heat means that the material can store or release substantial amounts of thermal energy without undergoing significant temperature fluctuations^[32]. Modulating the phase change temperature and latent heat is essential to adapt PCMs for diverse practical applications.

2.1.1 Eutectic PCMs

Eutectic PCMs are mixtures of two or more component PCMs that melt at a single, congruent temperature lower than the melting point of any individual constituent. This unique characteristic makes eutectic PCMs exceptionally suitable for targeting specific phase change temperature ranges that are unattainable by single-component PCMs^[33]. By fabricating various eutectic systems of organic PCMs, both the phase change temperature and latent heat can be

easily manipulated. The phase transition process of a eutectic PCM complies with the second law of thermodynamics and phase equilibrium theory. The liquidus line of a binary PCM system can be described by the following equation^[34]:

$$-\frac{H_A}{T_A}(T_m - T_A) + RT_m \ln(X_A) + G_{A,ex} = 0 \quad (1)$$

$$-\frac{H_B}{T_B}(T_m - T_B) + RT_m \ln(X_B) + G_{B,ex} = 0 \quad (2)$$

where H_A and H_B represent the enthalpies of components A and B (kJ/kg), respectively; T_A , T_B and T_m denote the melting points of components A, B and the mixture (K); X_A and X_B are the mole fractions of components A and B; $G_{A,ex}$ and $G_{B,ex}$ are the excess enthalpies of components A and B (kJ/kg); R is the universal gas constant (J/(mol·K)).

For organic compounds such as alkanes or alkanolic acids, it can be reasonably assumed that ideal mixing occurs, wherein $G_{A,ex} = G_{B,ex} = 0$. The aforementioned equation can thus be simplified to^[35–36]:

$$T_m = \left(\frac{1}{T_m} - \frac{R \ln X_i}{H_i} \right)^{-1} \quad (i = A, B) \quad (3)$$

By utilizing this simplified equation, the phase diagram can be plotted, and the eutectic point of the binary system can be mathematically determined. The latent heat of the binary system can be calculated using the following theoretical formulation:

$$H_m = T_m \left(\frac{X_A H_A}{T_A} + \frac{X_B H_B}{T_B} \right) \quad (4)$$

By solving these thermodynamic equations simultaneously, the eutectic point and the theoretical latent heat of the eutectic system can be determined.

Sari et al.^[37] prepared eutectic PCMs using *n*-alkanes including heptadecane (C₁₇), octadecane (C₁₈), nonadecane (C₁₉) and tetracosane (C₂₄). The formulated eutectic PCMs, with compositions and mass percentages of C₁₇–C₂₄ (90%–10%), C₁₉–C₁₈ (5%–95%), C₁₉–C₂₄ (95%–5%) and C₂₀–C₂₄ (90%–10%), exhibited DSC-measured melting points of 19.24, 25.15, 32.20 and 35.80 °C, respectively, alongside latent heats of fusion of 171.14, 180.25, 187.90 and 265.60 J/g. Duraipandi et al.^[38] developed a novel eutectic PCM composed of palmitic acid and sebacic acid (PA-SA). DSC analysis determined the melting point and latent heat of the PA-SA eutectic PCM to be 61.10 °C and 161.14 J/g, respectively. Baran et al.^[39] synthesized a binary

eutectic system by blending palmitic acid and stearic acid at a mass ratio of 64.2:35.8. The resultant eutectic mixture demonstrated a phase change temperature of 52.3 °C and a latent heat of 181.7 J/g.

2.1.2 Molecular structure regulation

Modifying materials at the molecular level by chemical methodologies is an effective strategy to intrinsically control phase change temperature and latent heat. During the phase transition process, parameters such as molecular weight and the dynamics of hydrogen bond formation and dissociation are paramount; they dictate intermolecular forces, which subsequently govern the thermal properties^[40]. By systematically tuning chain lengths, molecular weights, and the types of grafted moieties, the phase change parameters of PCMs can be precisely engineered to meet specific application demands. For instance, the phase change performance of a PCM can be tailored by grafting alkyl chains or utilizing PEG of varying molecular weights^[41].

Liu et al.^[42] synthesized a series of polypropylene-g-maleic anhydride-g- $C_nH_{2n+1}OH$ (PMC n , $n = 18, 22, 26$) comb-like polymeric solid-solid phase change materials (CPCMs) *via* a solvent-free esterification reaction between polypropylene-grafted maleic anhydride (PP-g-MAH) and normal alkanols with side chain lengths of $n=18, 22$ and 26 . The synthesis process of the CPCMs is illustrated in Figure 1. The pendant alkyl side chains distributed along the PP-g-MAH backbone exhibited distinct side-chain crystallization behavior. As the side-chain length increased, the phase change temperature of PMC n escalated from 60.0 to 82.2 °C, while the phase change enthalpy increased from 53.6 to 118.4 J/g.

Kong et al.^[43] successfully grafted acrylic acid onto starch to fabricate a superabsorbent resin (SAR). At a starch addition level of 10% (w/w), the latent heat reached 330.4 J/g, approximately 1.1 times higher than that of the pristine sample without starch. Liu et al.^[44] synthesized two azo compounds (T-Azo and F-Azo) appended with different alkyl chains. Following trans-to-cis isomerization, the short-chain T-Azo exhibited a drastically reduced crystallization temperature of -6.3 °C, compared to 8.2 °C for F-Azo. Furthermore, the combined photothermal energy release and phase change enthalpy of F-Azo achieved a high energy

density of 198.4 J/g, outperforming T-Azo (170.5 J/g) by 16%.

2.1.3 Adding nanoparticles

Recent studies have revealed that incorporating specific quantities of nanoparticles (such as nanocarbon materials or metal oxide nanoparticles) into PCMs can paradoxically induce an elevation in the phase change enthalpy of the resulting composites. This influence of nanoparticles on phase change enthalpy can be fundamentally understood as a competitive interplay between enhanced interfacial interactions and the adverse effects of mass substitution and crystallization disruption. The introduction of nanoparticles alters the interfacial molecular interaction network within the PCM, thereby impacting its solid-liquid phase change enthalpy. Upon nanoparticle incorporation, novel interfacial interactions emerge between the nanoparticle surface and the PCM molecular chains. Under conditions of strong interfacial affinity and excellent particle dispersion, the system must absorb supplementary energy during the melting process to overcome these interfacial constraints. When this surplus energy exceeds the enthalpy contribution lost by the fraction of PCM replaced by the nanomaterials, the overall system manifests an enhanced phase change enthalpy^[45–46].

Emeema et al.^[9] uniformly dispersed carbon quantum dots (CQDs) into paraffin to formulate composite PCMs. Compared to pristine paraffin, the latent heat of the composite PCMs was consistently augmented. Notably, at a CQD mass fraction of 1%, the latent heat experienced a remarkable 65.1% enhancement. Sahan et al.^[47] fabricated paraffin nanocomposites by embedding Fe_3O_4 nanoparticles into a paraffin matrix. At a Fe_3O_4 loading of $w = 10\%$, the latent heat of the paraffin increased by 27%. This enhancement was primarily attributed to the intensified intermolecular forces induced by the high specific surface area of the nanomaterials. Furthermore, X-ray diffraction analysis indicated the presence of defect sites on the Fe_3O_4 surface, which likely generated supplementary interfacial interactions between the nanoparticles and the paraffin, thereby elevating the overall enthalpy. Jacob et al.^[48] dispersed TiO_2 into a eutectic system composed of paraffin and palmitic acid to fabricate nano-enhanced eutectic PCMs (NeUPCMs). Compared to the pure matrix, the latent heat of the NeUPCMs improved by 17%.

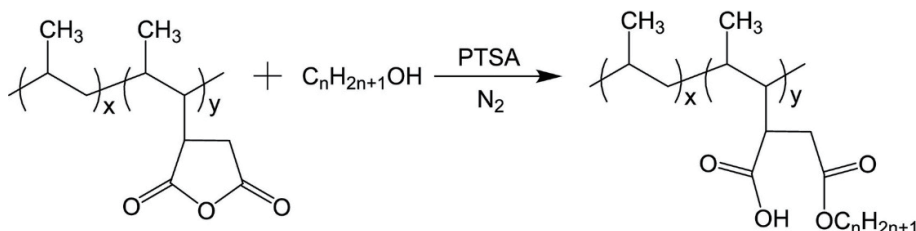


Fig. 1 Synthesis process of the CPCMs^[42]
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2.2 Thermal conductivity

For non-metallic PCMs, thermal conduction predominantly relies on lattice vibrations, specifically *via* phonons. As illustrated in Figure 2, the material lattice can be conceptualized as a system of atoms interconnected by spring-like interaction forces, where the interatomic forces are approximated by elastic potential energy. Upon heating, the atoms undergo thermal vibrations, deviating from their equilibrium positions. Phonons, functioning as the quantized carriers of lattice vibrations, transport thermal energy from the heated surface atoms to adjacent atoms,

thereby facilitating thermal diffusion through the lattice and establishing a temperature gradient^[49]. In an ideal and entirely defect-free crystal lattice, phonon propagation occurs without scattering limits, theoretically yielding infinite thermal conductivity. However, in practical materials, phonons inevitably encounter various scattering mechanisms. These scattering events perturb the phonon propagation paths, altering their momentum and energy, which significantly curtails their mean free path and consequently degrades the macroscopic thermal conduction efficiency^[50].

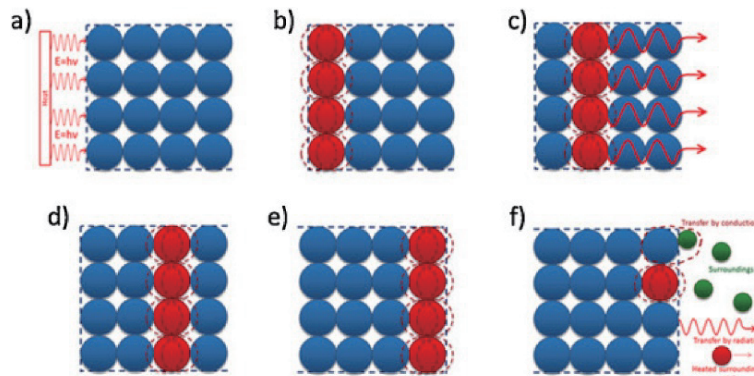


Fig. 2 Heat transfer mechanism in crystalline materials^[49]
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When nanofillers are introduced into a PCM, beyond providing supplementary thermal conduction pathways, interfacial phonon transport emerges as the decisive factor dictating the overall thermal performance. The heterogeneous interface formed between the PCM and the nanofillers induces acoustic impedance mismatch and severe scattering effects. This causes a fraction of high-frequency phonons to be reflected or randomly scattered, manifesting as substantial interfacial thermal resistance (Kapitza resistance)^[51]. This bottleneck is particularly pronounced in low-dimensional nanofiller systems (e.g., 0D or 1D fillers). The construction of a continuous thermally conductive pathway often necessitates heat to traverse the filler/matrix interface repeatedly. The cumulative superimposition of this interfacial thermal resistance severely restricts the effective enhancement of thermal conductivity^[52]. Conversely, as the dimensionality of the nano-additives increases, the fillers more readily form direct contacts, overlaps, or interconnected networks. This architectural shift enables thermal energy to propagate continuously over extended distances within the highly conductive fillers, drastically minimizing cross-interfacial phonon transport and thereby mitigating interfacial thermal resistance^[53]. The heat transfer mechanisms between PCMs and nano-additives of varying dimensions are schematically depicted in Figure 3. As the dimensionality of the nano-additives increases, the total interfacial area diminishes, resulting in a heat transfer efficiency that follows the trend: $3D > 2D > 1D > 0D$ ^[54]. While excessive filler loadings

inherently compromise the energy density of the PCM, insufficient loadings fail to establish continuous phonon transmission pathways. Therefore, engineering highly efficient, network-type (e.g., 3D interconnected) nano-additive architectures remains the ultimate objective for achieving superior thermal conductivity enhancement in PCMs^[55].

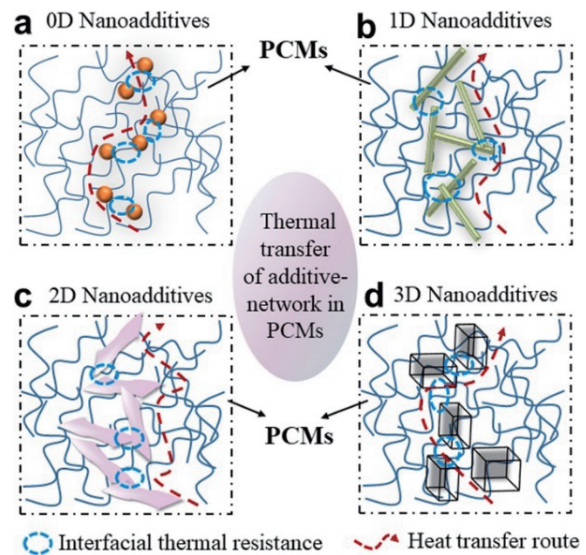


Fig. 3 Schematic diagram of the fundamental heat transfer mechanisms in a phase change material system incorporating nanoadditives of different dimensions^[54]
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2.2.1 Compositing with zero-dimensional (0D) materials

0D structural particles possessing high intrinsic thermal conductivity play a pivotal role in augmenting the thermal transport of PCMs. They primarily enhance heat flux by uniformly dispersing within the PCM to form local thermal conduction chains, which incrementally elevate the phonon group velocity and mean free path^[56]. However, during solution processing, nanoparticles are highly susceptible to sedimentation and agglomeration. This aggregation obstructs phonon transmission, detrimentally impacting the heat transfer characteristics of the phase change composite. Consequently, achieving adequate dispersion often necessitates the assistance of barrier or functionalization materials^[54]. Commonly employed 0D nanomaterials predominantly include metallic nanoparticles, metal oxides, and specific nanocarbon derivatives.

Liu et al.^[57] engineered a highly thermally conductive nano-ZnO functionalized paraffin/diatomite PCM (PA/F-DE/ZnO) via a facile melt-blending and physical adsorption approach. Compared to PA/F-DE, the integration of ZnO ($w = 10\%$) boosted the thermal conductivity of PA/F-DE/ZnO by 87.73%. Ezhumalai et al.^[58] utilized CuO nanoparticles to amplify the thermal conductivity of palmitic acid. Compared to pure palmitic acid, composite PCMs loaded with CuO mass fractions of 0.1%, 0.2% and 0.3% exhibited thermal conductivity enhancements of 19.87%, 45.61% and 66.08%, respectively^[59]. When the TiO₂ nanoparticles were dispersed into palmitic acid to formulate nano-enhanced PCMs (PA/TiO₂) with TiO₂ mass fractions of 0.5%, 1%, 3% and 5%, the thermal conductivity of the palmitic acid was elevated by 12.7%, 20.6%, 46.6% and 80%, respectively.

2.2.2 Compositing with one-dimensional (1D) materials

1D materials, such as nanowires, carbon fibers and carbon nanotubes (CNTs), possess distinct advantages including inherently high thermal conductivity, low density, and robust chemical stability, rendering them highly effective for phonon transport. When these 1D materials reach a critical percolation threshold and are uniformly dispersed within the PCM matrix, their large aspect ratios facilitate mutual contact and overlapping. This spatial arrangement naturally constructs an interconnected thermally conductive network, drastically enhancing the thermal transport capabilities of the composite^[55].

Nomura et al.^[60] fabricated composite PCMs (PCCs) utilizing erythritol as the phase change matrix and carbon fibers as the highly conductive filler, employing both melt-dispersion and hot-pressing techniques. As illustrated in Figure 4, compared to pristine erythritol (thermal conductivity of 0.76 W/(m·K)), both preparation methods

successfully elevated the effective thermal conductivity of the PCCs. Notably, the hot-pressing method yielded a significantly more pronounced enhancement than melt-dispersion. In the hot-pressed PCCs, the carbon fibers preferentially aligned within the plane perpendicular to the applied pressing stress. Given the highly anisotropic thermal conductivity of carbon fibers, this highly oriented architecture critically accelerated the dramatic improvement in the effective thermal conductivity of the PCM.

Zhang et al.^[61] prepared phase change composites (PCCs) utilizing an erythritol matrix filled with short carbon fibers (SCFs) of two distinct lengths. The SEM images of the SCFs and PCCs, shown in Figure 5, confirm the uniform dispersion of the SCFs within the phase change matrix. The thermal conductivity of the PCCs escalated concurrently with the SCF loading. The composite containing 10% of the longer SCFs achieved the highest thermal conductivity at 3.92 W/(m·K), representing a staggering 407.8% enhancement over pure erythritol (0.77 W/(m·K)).

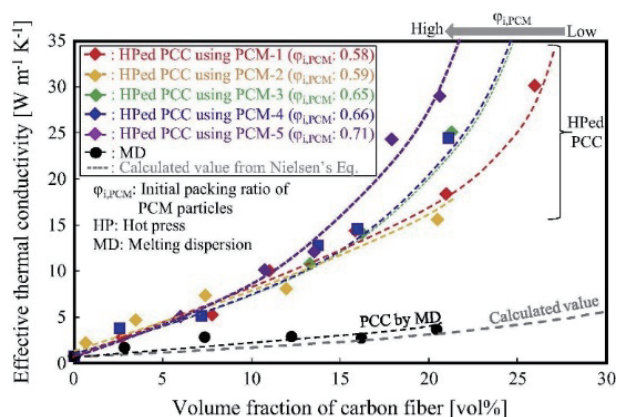


Fig. 4 Effective thermal conductivity of phase change composites via hot-pressing and melt-blending^[60] (with permission from Elsevier)

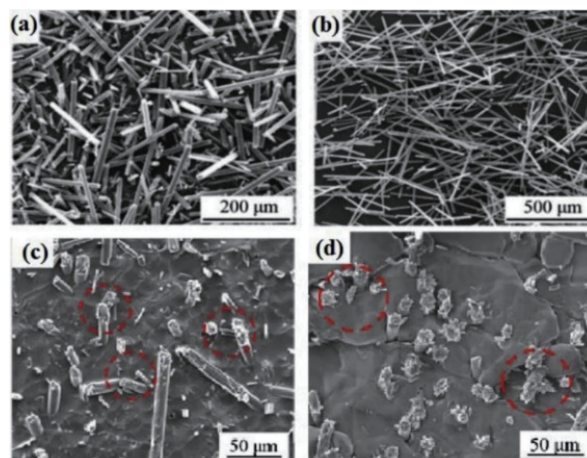


Fig. 5 SEM images of SCFs and PCCs: (a), (b) C₅ and C₂₅ at different magnifications; (c), (d) PCCs containing 10% C₅ and 10% C₂₅^[61] (with permission from Elsevier)

Salyan et al.^[62] dispersed multi-walled carbon nanotubes (MWCNTs) into D-mannitol to prepare thermally enhanced nanocomposite PCMs at weight fractions of 0.1% and 0.5%, which generated maximum thermal conductivity enhancements of 7% and 32%, respectively.

2.2.3 Compositing with two-dimensional (2D) materials

2D materials, prominently represented by graphene, are atomically thin carbon monolayers. Owing to the robust sp^2 covalent bonds between carbon atoms and its planar architecture, graphene exhibits extraordinary thermal conductivity, permitting near-ballistic phonon transport with minimal scattering and yielding an intrinsic thermal conductivity of up to $5000 \text{ W/(m}\cdot\text{K)}$ ^[63]. 2D graphene is frequently combined with 1D fillers to introduce supplementary thermal conduction pathways, thereby amplifying the overall thermal conductivity of the PCM. The 2D structure efficiently establishes thermal percolating networks within the PCM matrix and acts as a bridging element between localized clusters of 1D fillers, synergistically enhancing heat transfer^[55,64].

Zhong et al.^[65] incorporated four types of graphene nanoplatelets (GNPs) (Figure 6) with varying lateral dimensions and thicknesses into *n*-eicosane to formulate nano-enhanced PCMs (NePCMs). Compared to pristine *n*-eicosane, all GNP-loaded samples exhibited augmented thermal conductivity, with a maximum enhancement of 2.1-fold. Experimental results confirmed that the aspect ratio of GNPs significantly dictates the thermophysical properties of NePCMs; a smaller aspect ratio facilitates the more seamless establishment of a thermally conductive network, thereby optimizing thermal conductivity.

As shown in Figure 7(a), Zhang et al.^[66] utilized hyperbolic graphene aerogels (HGAs) (Figure 7(c)) as highly conductive scaffolds to encapsulate paraffin,

fabricating paraffin-graphene composites (PGCs) (Figure 7(d)) specifically tailored for lithium-ion battery (LIB) thermal management. At a filler mass fraction of 12.5%, the PGC retained a high latent heat of 200 J/g , only a 9.1% reduction compared to pure paraffin, while achieving an exceptional thermal conductivity of $30.75 \text{ W/(m}\cdot\text{K)}$. Regarding LIB thermal management, the PGC effectively suppressed thermal runaway. Even after 10000 rapid charge-discharge cycles, the system strictly maintained the battery temperature at 42°C , ensuring stable capacity and Coulombic efficiency.

Badakhsh et al.^[67] employed a synergistic mixture of CNTs and GNPs to enhance the thermal performance of paraffin. While all composite PCMs exhibited higher thermal conductivities than pure paraffin, an increasing ratio of CNTs to GNPs resulted in a declining trend in thermal conductivity. The findings indicated that the planar 2D structure of GNPs more effectively bridges the isolated CNT clusters, as depicted in Figure 8, thereby forging superior thermal conduction pathways throughout the matrix.

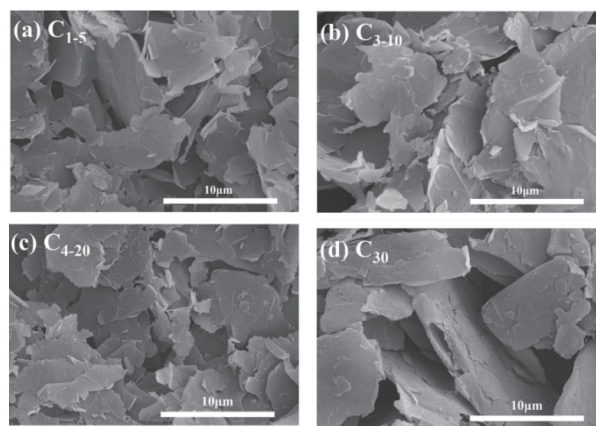


Fig. 6 SEM image of GNPs dispersed in ethanol^[65] (with permission from Elsevier)

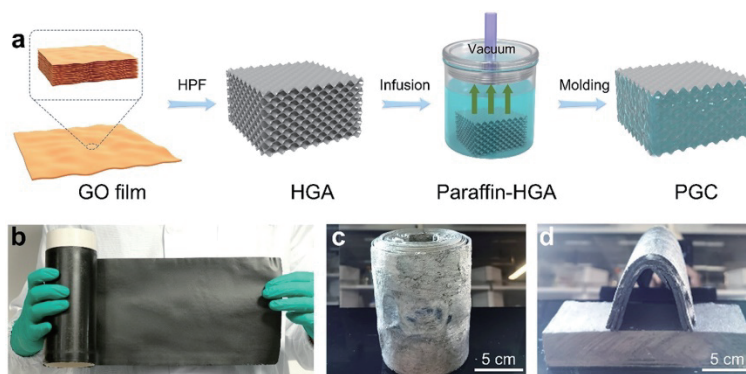


Fig. 7 Fabrication and properties of PGC material^[66]
(a) Schematic of PGC preparation; (b) GO film; (c) HGA film; (d) PGC monolith
(with permission from Elsevier)

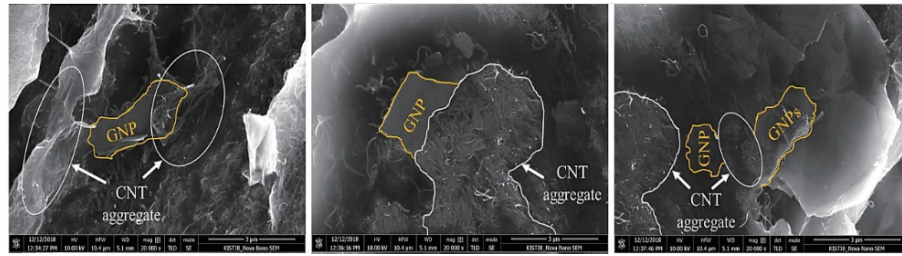


Fig. 8 Bridging effect of the GNPs^[67]
(with permission from Elsevier)

2.2.4 Compositing with three-dimensional (3D) materials

3D structural additives inherently form interconnected porous networks, which provide continuous, long-range thermal conduction pathways within the PCM matrix. A highly porous network can accommodate and retain a maximal volume of the PCM^[68]. Furthermore, the robust capillary forces and surface tension provided by the micropores effectively confine the liquid PCM, preventing leakage during the solid-liquid phase transition^[69]. Commonly utilized porous materials include metal foams,

porous carbons, and inorganic minerals^[70].

Xiao et al.^[71] prepared paraffin/nickel foam and paraffin/copper foam composite PCMs *via* vacuum impregnation. Figure 9(a) and (c) display the morphologies of the pristine nickel and copper foams with distinct pore sizes, while Figure 9(b) and (d) present the resultant paraffin/nickel and paraffin/copper foam composites. It is evident that pure paraffin uniformly infiltrates the metal foam pores, demonstrating excellent interfacial compatibility. Experimental measurements revealed that the thermal conductivity of the paraffin/nickel foam composite was three times that of pure paraffin, whereas the paraffin/copper foam composite achieved a 15-fold enhancement.

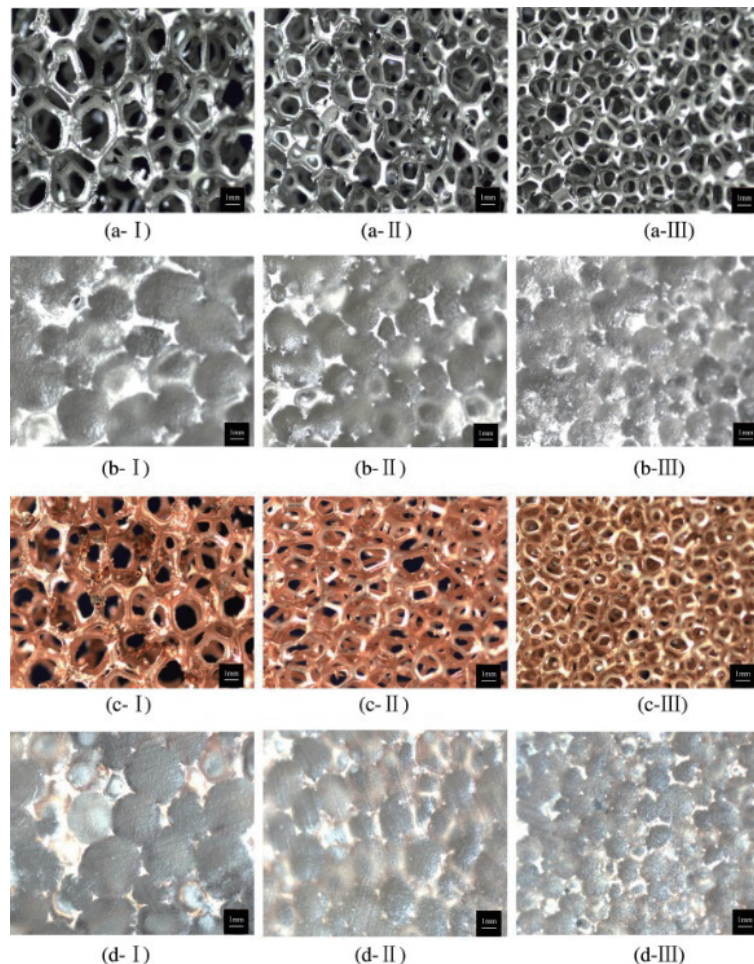


Fig. 9 Images of metallic foams with different pore sizes and their corresponding paraffin/metal foam composite PCMs^[71]
(a) Nickel foam; (b) Paraffin/nickel foam composite PCM; (c) Copper foam; (d) Paraffin/copper foam composite PCM
(with permission from Elsevier)

Yuan et al.^[72] developed a shape-stabilized erythritol/EG composite PCM for medium-temperature thermal energy storage *via* a facile “impregnation-compression-sintering” three-step strategy, with SEM images shown in Figure 10 (a)–(i). The evolution of the latent heat and thermal conductivity as a function of EG content is plotted in Figure 10(j). As the EG mass fraction increased, the latent heat decreased while the thermal conductivity exhibited an almost linear escalation. Balancing thermal storage capacity and conductivity, the composite with an EG mass fraction of 10% demonstrated optimal performance, yielding a latent heat of 212.5 J/g and a thermal conductivity of 12.51 W/(m·K), which is 17.38 times higher than that of pristine erythritol.

Qian et al.^[73] fabricated a composite PCM by infiltrating paraffin into a hexagonal boron nitride (*h*-BN) porous scaffold featuring continuous thermal conductive pathways. The resulting PCM, containing an *h*-BN mass fraction of 18%, exhibited a melting latent heat of 165.4 and an impressive thermal conductivity of 0.85 W/(m·K), representing a ~600% enhancement over pure paraffin.

2.3 Supercooling

Supercooling refers to the phenomenon where a material remains in a liquid state below its thermodynamic melting point. In short-term energy storage applications, supercooling can severely degrade the thermal efficiency of a PCM system. For instance, in building applications, solidification typically occurs within a limited nocturnal timeframe. If the PCM experiences severe supercooling instead of timely crystallization, it fails to release its stored thermal energy^[74]. Whether a PCM smoothly transitions into the solidification phase during cooling is intricately linked to the dominant nucleation mechanism within the system. If the system relies on homogeneous nucleation, the crystal nucleus must form spontaneously within a perfectly uniform liquid phase. Because this process must overcome a

substantial interfacial energy barrier, requiring significant energy fluctuations, it generally necessitates deep supercooling to achieve stable nuclei formation. High-purity PCMs devoid of solid interfaces or nucleation sites typically exhibit this mechanism, making them prone to pronounced supercooling^[75].

Conversely, heterogeneous nucleation occurs at solid-liquid interfaces, such as container walls, trace impurities, dispersed nanoparticles, porous scaffolds, or rough topological surfaces. The presence of these interfaces drastically reduces the effective interfacial energy required for nucleus formation, lowering the critical nucleation energy barrier and enabling the system to initiate crystallization at minimal supercooling degrees^[74,76]. For organic PCMs plagued by high supercooling, such as sugar alcohols, the introduction of nucleating agents, porous frameworks, or encapsulation shell materials serves as a highly effective mitigation strategy.

Cheng et al.^[77] introduced sodium carboxymethyl cellulose (CMC-Na) and hexagonal boron nitride (*h*-BN) to regulate the supercooling and thermal properties of erythritol. The supercooling degree and solidification stability of erythritol were suppressed by 93% and 84%, respectively, while the melting latent heat experienced a controlled reduction from 335.1 to 294.1 J/g. Shao et al.^[24] uniformly dispersed polyvinylpyrrolidone (PVP) in erythritol to mitigate supercooling. The results demonstrated that a PVP addition ($w = 1.0\%$) plummeted the supercooling degree from over 64 °C down to approximately 21 °C. The composite retained a melting latent heat of 301 J/g, merely a 9.3% decrease compared to pure erythritol. Yuan et al.^[78] investigated the supercooling behavior of pure erythritol versus an erythritol/EG composite ($w = 15\%$). Their findings indicated that the incorporation of EG suppressed the supercooling degree by 25 °C and tightened the solidification stability margin to 10 °C.

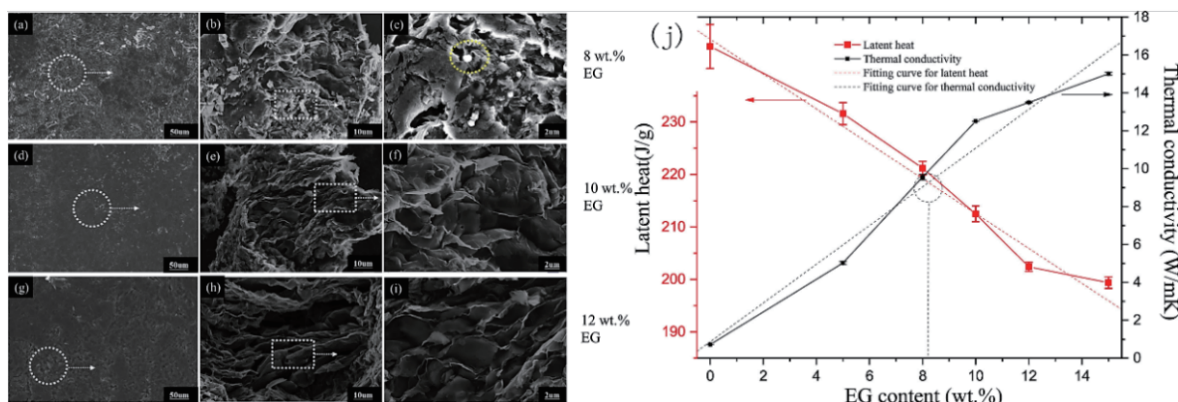


Fig. 10 (a)–(i): SEM images of the EG/erythritol composite; (j) Latent heat and thermal conductivity of the composite PCM as a function of the EG mass fraction^[72] (with permission from Elsevier)

2.4 Flame retardancy

Organic PCMs, composed primarily of carbon, hydrogen and oxygen, are highly susceptible to ignition upon exposure to high temperatures or direct flames, posing a severe threat to life and property. Resolving the inherent flammability of organic PCMs is of paramount importance for advancing their practical engineering applications and commercialization. Xiao et al.^[79] comprehensively enumerated various classes of flame retardants, elucidating their underlying flame-retardant mechanisms alongside their respective advantages and limitations, as summarized in Table 4. Current flame-retardant strategies for organic PCMs primarily encompass physical blending, chemical modification, microencapsulation and surface coating.

2.4.1 Physical blending of flame retardants

Dispersing flame retardants into PCMs *via* physical blending is highly favored for its simplicity and operational ease. However, this approach presents distinct drawbacks. Poor interfacial compatibility between the flame retardant and the PCM often compromises the mechanical properties and long-term stability of the composite. Furthermore, achieving satisfactory flame retardancy typically necessitates high loading concentrations, which detrimentally dilutes the thermal storage density of the

PCM.

Li et al.^[80] fabricated a composite PCM using *n*-octadecane, rice husk char (RHC), ammonium polyphosphate (APP), epoxy resin (EP) and EG. The composite containing an APP mass fraction of 15% exhibited exceptional flame retardancy, achieving a UL-94 V-0 rating. The limiting oxygen index (LOI) increased from 18.3% to 23.7% and the total heat release (THR) measured *via* cone calorimetry decreased by 20%. DSC analysis confirmed a melting peak temperature of 27.0 °C and a latent heat of 128 J/g. Li et al.^[81] introduced a novel triazine charring foaming agent (CFA) and APP into a shape-stabilized PCM, utilizing paraffin as the thermal storage medium and polypropylene (PP) as the supporting matrix. At an APP/CFA loading of $w = 30\%$, the composite passed the UL-94 V-0 rating and achieved an LOI value of 32.8%. Fang et al.^[82] prepared a flame-retardant *n*-hexadecane/silica (SiO₂) composite for thermal energy storage, further augmenting its flame retardancy by incorporating EG. At an *n*-hexadecane mass fraction of 73.3%, the melting and solidification latent heats were 147.58 and 145.10 kJ/kg, respectively. Microstructural analysis of the post-combustion char residues revealed that a uniform, continuous, and dense char layer effectively barricaded heat and mass transfer, thereby enhancing the composite's flame resistance.

Table 4 Types, mechanisms, advantages and disadvantages of flame retardants

Classification	Example	Mechanism	Advantage	Disadvantage
Halogenated flame retardants	Chlorinated paraffins (CP), polybrominated diphenyl ethers (PBDEs), hexabromocyclododecane (HBCD)	Halogen free radicals are generated during combustion, which can rapidly capture $\cdot\text{H}$ and $\cdot\text{OH}$	Efficiency, good thermal stability, and good compatibility	Produces corrosive and toxic fumes
Nitrogen flame retardants	Triazine compounds melamine, dicyandiamide, guanidine salts and melamine cyanurate	Releasing non-flammable gases, absorbing heat, diluting oxygen	Safe, non-toxic, and broadly applicable	Inefficiency when used alone, synergistic with N-/P-based flame retardants
Silicon flame retardant	SiO ₂ , Halloysite nanotubes (HNT), Oxidized montmorillonite (OMT), silicone resin (SR) and polysiloxane (PSO)	Promoting polymer degradation forms a protective layer based on Si–O and Si–C bonds, suppressing the diffusion of heat and oxygen	Processability and environment-friendly	Poor dispersion, high cost, susceptible to moisture and heat
Phosphorus flame retardant	Red phosphorus, phosphorus oxychloride (POCl ₃) and phosphorus black (PB)	$\cdot\text{PO}$ and $\cdot\text{PO}_2$ radicals were generated during combustion, and $\cdot\text{H}$ and $\cdot\text{OH}$ were captured	Low smoke, high process stability and good thermal stability	High cost, low degradation rate and environmentally unfriendly
Metal hydroxide	Mg(OH) ₂ , Al(OH) ₃	It decomposes to absorb heat and releases water vapor, forming a metal oxide protective layer that reduces heat transfer	Lower price, non-toxic and environmental protection	Poor compatibility and inefficiency
Intumescent flame retardant	Ammonium polyphosphate (APP), pentaerythritol (PER) and melamine	Form foam and Carbonization layer on the surface to prevent the underlying material from further exposure to ignition	Low smoke, non-toxic and efficient	Poor compatibility, affecting mechanical properties

2.4.2 Chemical modification

Covalently bonding flame-retardant functional groups to the molecular structure of PCMs ensures uniform dispersion at the molecular level, fundamentally eradicating the poor compatibility issues inherent to physical blending. This chemical modification strategy not only significantly suppresses the migration and phase separation of flame retardants but also bolsters the flame-retardant efficacy while preserving the PCM's thermal storage capacity.

Qian et al.^[83] synthesized a novel halogen-free, flame-retardant hybrid phase change composite *via* an *in situ* sol-gel method, utilizing paraffin as the core medium and tris(triethoxysilylpropyl) phosphoramidate as the sol-gel precursor. The synergistic interaction between the paraffin and the hybrid silsesquioxane network substantially enhanced the char-forming capability of the paraffin. Compared to pure paraffin, the peak heat release rate (PHRR) was drastically reduced by 62.1%.

Jiang et al.^[84] engineered a series of novel shape-stabilized PCMs enriched with phosphorus and silica *via* the transesterification of diethyl phosphite with 1-tetradecanol, 1-hexadecanol, and 1-octadecanol, followed by a ring-opening reaction and the hydrolytic condensation of 2,3-epoxypropoxy propyltrimethoxysilane (silane coupler KH-560). The synthesis route is detailed in Figure 11. SEM imaging and EDS spectroscopy corroborated that the dual incorporation of phosphorus and silicon accelerated protective char formation, thereby elevating both thermal stability and fire resistance.

Zhang et al.^[85] synthesized boron-modified octadecanol PCMs with varying molecular architectures by systematically adjusting the stoichiometric ratio of boric acid to octadecanol (Figure 12). The boron modification remarkably elevated the thermal stability and flame retardancy of octadecanol, yielding a 70.9% reduction in the heat release rate (HRR) and a 77.8% decrease in the smoke production rate (SPR).

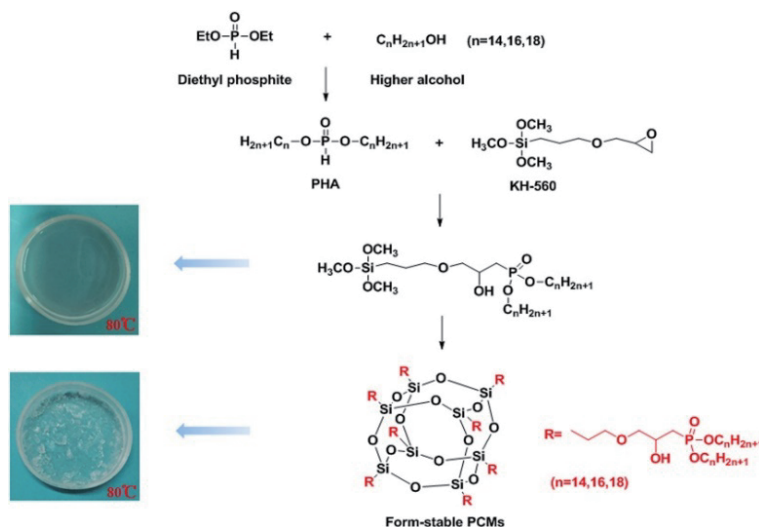


Fig. 11 Synthetic process of shape-stabilized PCMs^[84]
(with permission from Elsevier)

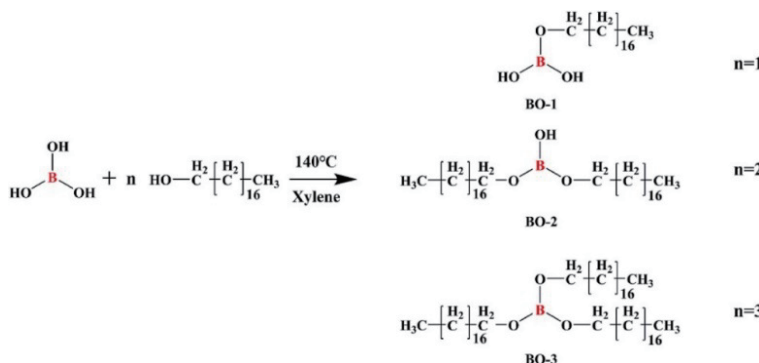


Fig. 12 Chemical reaction of boron-modified octadecane^[85]
(with permission from Elsevier)

2.4.3 Flame-retardant microencapsulation

Phase change microencapsulation involves enveloping

PCMs within a protective microscopic shell. This architecture prevents direct material contact and leakage during the phase transition while simultaneously enhancing

heat transfer area^[86]. Flame retardancy in microcapsules is typically achieved *via* two routes: directly encapsulating the flame retardant within the core alongside the PCM, or chemically/physically modifying the polymer shell to be intrinsically flame-retardant.

Hu et al.^[87] formulated silica-shelled microcapsules (SiO₂-MCs) and poly(urea-formaldehyde)-shelled microcapsules (PUF-MCs), utilizing a hybrid core of octadecane and tributyl phosphate (TBP). The pHRR of the SiO₂-MCs was suppressed by 26.7%, significantly outperforming the 11.5% reduction observed in PUF-MCs. The rigid SiO₂ shell provided superior thermal stability and a robust flame-retardant barrier, slowing the combustion rate and shortening the burning duration. The flame-retardant mechanism of these microcapsules is schematically illustrated in Figure 13.

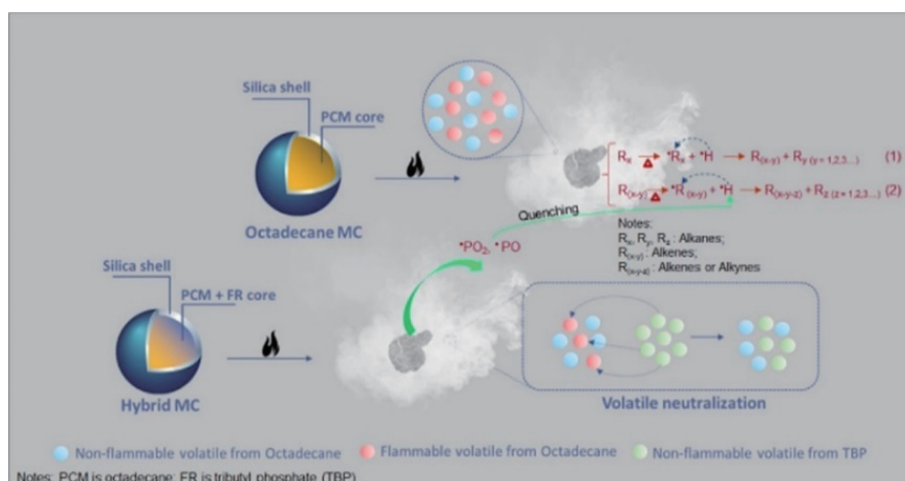


Fig. 13 Schematic diagram of the flame retardant behavior of octadecane/TBP hybrid microencapsulated PCMs (ME-PCMs)^[87] (with permission from ACS Publications)

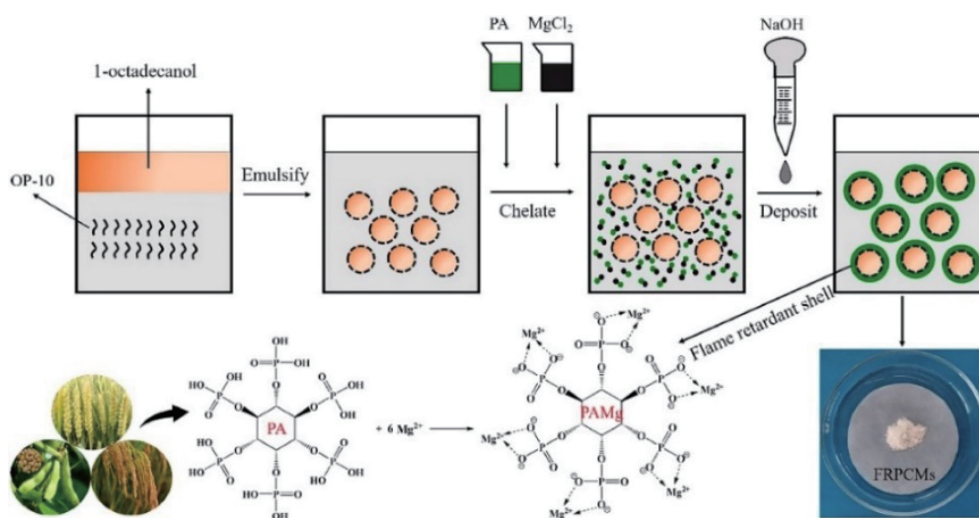


Fig. 14 Schematic diagram of the FRPCMs preparation process^[88] (with permission from Elsevier)

2.4.4 Surface coating

The coating method involves applying a flame-retardant

Liao et al.^[88] fabricated flame-retardant PCMs (FRPCMs) utilizing octadecane as the core and bio-based magnesium phytate (PAMg) as the intrinsically flame-retardant shell (Figure 14). DSC measurements indicated a melting latent heat of 117–158 J/g. LOI and cone calorimetry assessments demonstrated an LOI increase from 18.5% to 21.6% and a 39.5% reduction in total heat release (THR).

Du et al.^[89] synthesized nanoencapsulated PCMs (NanoPCMs) featuring an n-octadecane core and a poly(methyl methacrylate) (PMMA) shell, prepared *via* the condensation reaction between diethyl bis(2-hydroxyethyl) aminomethylphosphonate (DEAMP) and acryloyl chloride. The incorporation of the phosphorus-containing DEAMP into the NanoPCMs significantly stifled heat and smoke release in EP/NanoPCM composites, concurrently enhancing their residual char mass and LOI values.

layer directly onto the surface of macroscopic, shape-stabilized PCMs to fortify fire resistance. Because the flame-retardant barrier is confined to the exterior, the phase

change dynamics and intrinsic thermal storage capacity of the internal PCM are largely preserved without disruptive chemical or physical interference.

Xu et al.^[90] fabricated shape-stabilized PCMs (SSPCMs) comprising styrene-butadiene-styrene block copolymer (SBS)/HDPE/paraffin/OMMT. Three distinct intumescent coatings—acrylic resin/EG, alkyd resin/EG and epoxy resin/EG—were applied to the SSPCM surfaces. Once the EG content surpassed a critical threshold, all surface-coated SSPCMs exhibited outstanding flame retardancy. The horizontal burning time was substantially prolonged, achieving a UL-94 V-0 rating, while the LOI values surged from 17.4% to over 30%. Additionally, the PHRR plummeted by 62%–84%, the overall heat release process

was remarkably delayed, and total smoke production was minimized. The flame-retardant mechanism of the coated SSPCMs is depicted in Figure 15.

Niu et al.^[91] compounded ammonium polyphosphate (APP) and dipentaerythritol (DEPR) as flame retardants (APP:DEPR = 3:1) with polyurethane (PU) to formulate an intumescent flame-retardant coating. Initially, a composite PCM (CPCM) was prepared using paraffin and silica aerogel (SA). By applying the flame-retardant coating onto the CPCM, an FR-CPCM was obtained. The FR-CPCM achieved an exceptional LOI of 56.31%, and due to the formation of a dense, intumescent carbonized layer at elevated temperatures, its flame-retardant rating escalated to V-0 in the UL-94 vertical burning test.

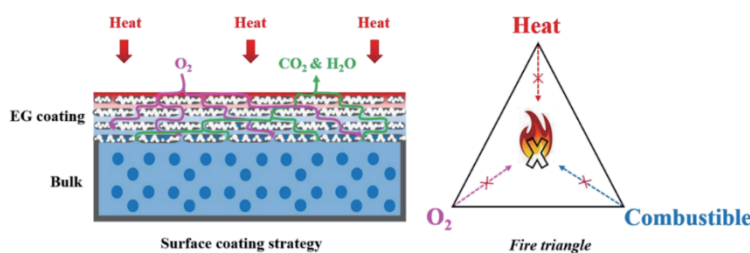


Fig. 15 Flame retardant mechanism of surface-coated SSPCMs^[90]
(with permission from MDPI)

3 Conclusion and outlook

Organic PCMs possess distinct advantages such as high phase change enthalpy, tunable phase change temperatures, excellent chemical stability, minimal supercooling, relative cost-effectiveness, and abundant raw material sources. Therefore, they have emerged as indispensable candidates in the domains of thermal energy storage and thermal management. This review has comprehensively summarized several mainstream organic PCMs, detailing their phase transition characteristics (temperature, enthalpy), intrinsic merits, and limitations. Furthermore, grounded in the physicochemical properties of these materials, recent advancements in performance optimization strategies for organic PCMs have been systematically evaluated.

Despite the broad application prospects of organic PCMs, the large-scale engineering deployment remains bottlenecked by critical challenges, including inherently low thermal conductivity, compromised long-term cycling stability, and elevated overall cost. To propel this field forward, future research should strategically pivot towards the following pivotal directions.

(1) **Intrinsic Enhancement of Material Properties:** Future studies should focus on the precise matching of phase change temperatures and latent heats through molecular architectural design, the construction of tailored eutectic systems, and targeted functionalization. Concurrently, it is crucial to fundamentally elucidate the mechanisms governing intermolecular forces, crystal growth behavior, and nucleation kinetics following the introduction of nanoparticles, thereby enabling the predictive modulation of

latent heat variations and the targeted suppression of supercooling.

(2) **Efficient Heat Transfer and Interfacial Engineering:** By rationally designing the dimensionality, spatial distribution and interfacial coupling of thermally conductive fillers—while stringently preserving high latent heat density—researchers must construct composite PCM architectures characterized by minimized interfacial thermal resistance and continuous phonon transmission pathways. This is essential to decisively overcome the limited thermal response rates of organic PCMs.

(3) **Structuralization and Multifunctional Integration:** The continuous development of spatial confinement technologies—such as porous framework infiltration, micro- and nano-encapsulation and 3D skeleton supporting structures—is imperative for elevating the morphological stability and cyclic reliability of PCMs. Moreover, the seamless integration of functional units capable of photothermal, electrothermal, or electromagnetic-responsive conversion into the phase change matrix will yield multifunctional phase change materials, satisfying the stringent demands of complex operational environments.

(4) **Engineering Application and Sustainable Development:** Finally, the trajectory of organic PCM research must shift towards cost reduction, green chemistry, and scalable manufacturing. Enhanced efforts should be directed towards developing bio-based, industrial by-product-derived and renewable resource-based PCMs to simultaneously mitigate material costs and alleviate the environmental footprint.

Declaration of Competing Interest

The authors declare no conflict of interest.

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有机相变储能材料性能优化研究进展

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摘要: 有机相变材料(PCM)因其优异的能量存储特性, 在热管理和能源存储领域具有广泛应用。本工作综述了有机PCM的主要类型及其性能优化策略。在热物性调控方面, 介绍了通过构建共晶体系、调控分子结构及添加纳米颗粒等方式调节相变温度与潜热。在导热增强方面, 总结了与零维、一维、二维及三维结构高导热材料复合的策略。还针对有机PCM存在的过冷现象和易燃性问题, 分析了相应的抑制过冷方法与阻燃改性技术。展望了有机相变材料的发展方向, 随着技术的突破, 有机相变材料将在能源存储和热管理等领域得到更广泛应用。

关键词: 有机相变材料; 潜热; 导热性能; 阻燃性

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