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## Thermoresponsive hydrogels incorporating phase-change energy storage materials for advanced thermal management

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With the rising global energy demand and worsening environmental challenges, the development of efficient energy storage and conversion technologies has become increasingly critical. Combining phase-change materials with thermally responsive hydrogels integrates the high water content and biocompatibility of hydrogels with the superior thermal energy storage capacity of phase-change materials, offering a promising strategy for advanced thermal management. This review summarizes the latest advancements in phase-change hydrogels, covering synthesis, thermal responsiveness regulation, and applications. It discusses strategies for improving performance through material design and structural optimization, and explores the potential applications and challenges in areas such as building energy efficiency, personal thermal management, healthcare, and electronics. By synthesizing the latest research progress and identifying future research directions, this review aims to provide valuable insights for both academia and industry, fostering further innovation and practical implementation of phase-change energy storage hydrogels in next-generation thermal management solutions.

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### 1. Introduction

With the rapid advancement of technology, thermal management has become an essential component across various sectors. In fields such as electronic device cooling, aerospace engineering, and building energy efficiency,<sup>1</sup> effective thermal regulation is crucial for ensuring system stability, extending



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to the development of efficient means or catalysts for polyester alcoholysis and polyolefin hydrogenolysis.

equipment lifespan, and enhancing overall energy efficiency. While conventional thermal management approaches, such as air and liquid cooling, offer viable solutions, they often suffer from limitations including low efficiency, high energy consumption, and potential environmental concerns. As a result, the pursuit of high-performance, energy-efficient, and environmentally sustainable thermal management technologies has emerged as a critical research frontier, driving innovation in energy storage and conversion strategies.<sup>2</sup>

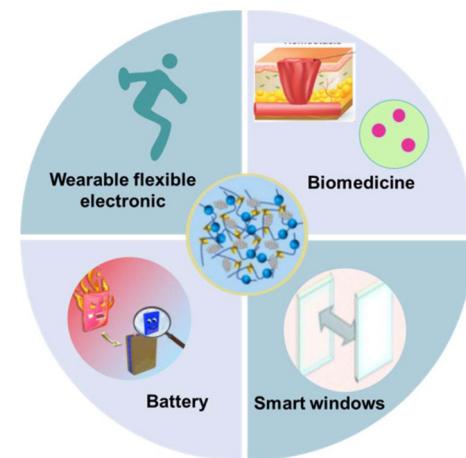
Phase-change materials (PCMs) have attracted significant attention for their ability to absorb and release latent heat during phase transitions, maintaining a stable temperature, and emerged as versatile platforms for thermal management. As the demand for clean energy and sustainable technologies grows, the development of high-performance PCMs has become increasingly critical. Phase-change hydrogels integrate the high water content, flexibility, and biocompatibility of hydrogels with the thermal energy storage and regulation capabilities of PCMs. This unique synergy enables efficient and adaptive thermal management, presenting a promising avenue for next-generation energy-efficient systems.

The three-dimensional (3D) network structure of hydrogels not only physically encapsulates PCMs, preventing leakage during phase transitions, but also enhances heat transfer efficiency through its interconnected porous architecture.<sup>3</sup> As an emerging class of intelligent materials, phase-change hydrogels have demonstrated significant potential for advanced thermal management applications (Fig. 1).<sup>4</sup> By leveraging their unique phase-change properties, these materials can efficiently absorb and release large amounts of heat at precise temperatures, offering innovative solutions for thermal regulation. However, optimizing their performance to meet diverse thermal management demands and expanding their applicability across various fields remain key research challenges, driving ongoing exploration and technological advancements.



**Jinxing Chen**

*nanostructured materials for energy, and environmental applications.*



**Fig. 1** Phase-change hydrogels for advanced thermal management.<sup>5–9</sup>

This review provides a comprehensive summary of recent advances in phase-change hydrogels for thermal management, evaluating the advantages and limitations of various materials and fabrication techniques. It explores strategies to enhance the performance of phase-change hydrogels through material design and structural optimization while addressing key technical challenges and future research directions. We systematically discuss the synthesis and preparation methods, mechanisms for tuning thermal responsiveness, and applications across diverse fields. Additionally, the existing research challenges are critically analyzed, and potential future directions are proposed to inspire innovation and guide further development in this rapidly evolving field.

## 2. Encapsulation technology of PCMs

When selecting phase change materials (PCMs) suitable for encapsulation within hydrogels, a comprehensive evaluation of their physicochemical properties, environmental adaptability, and scalability is imperative. Central to this process is the alignment of phase transition parameters, particularly latent heat and transition temperature, with application-specific thermal requirements. Solid–liquid PCMs are the most widely used in energy storage due to their high phase-change enthalpy, stable volume, diverse compositions, and broad application range.<sup>10</sup> As illustrated in Fig. 2, these PCMs can be categorized into inorganic and organic phase-change materials, each with distinct phase transition characteristics and compatibility challenges when integrated into hydrogel matrices; targeted selection can be made based on specific application requirements. For instance, refrigeration systems designed for food or pharmaceutical storage necessitate precise temperature regulation within narrow ranges, while electronics cooling demands rapid heat dissipation under high thermal thresholds. Given the intrinsic fluidity of solid–liquid PCMs, encapsulation or integration within a suitable matrix is essential to maintain long-term stability and per-

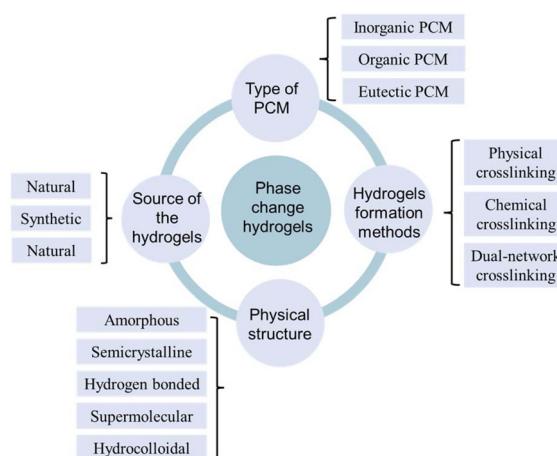


Fig. 2 Classification of phase-change hydrogels.<sup>11</sup>

formance. Hydrogels, with their tunable structures and three-dimensional network architecture, serve as ideal carriers for PCMs, effectively preventing leakage and enhancing durability. To achieve stable PCM loading within hydrogels, both material selection and structural design must be carefully optimized, ensuring efficient thermal energy storage and long-term reliability. Below, we systematically analyze their properties and encapsulation strategies.

### 2.1. Encapsulation of inorganic PCMs in hydrogels

Hydrogels, with their abundant hydrophilic groups, exhibit excellent compatibility with most inorganic PCMs, such as hydrated salts.<sup>12,13</sup> Whether the PCMs are impregnated into the hydrogel matrix or directly incorporated during hydrogel preparation,<sup>14</sup> phase-change hydrogels with relatively stable properties can be achieved. The strong polarity of hydrogels facilitates their interaction with inorganic hydrated salts. For instance, Xiao *et al.*<sup>15</sup> developed a light-to-thermal conversion phase-change hydrogel by incorporating sodium acetate trihydrate (SAT), an acrylamide-acrylic acid sodium copolymer, the gelling agent 80A-51, and CuS using a melt impregnation process. The combination of SAT and 80A-51 greatly restricted the water molecular movement when SAT was in its molten state. A common approach for preparing phase-change hydrogels involves taking advantage of the compatibility between PCMs and hydrogel monomers in a single step. For example, Yin *et al.*<sup>16</sup> proposed a simple method to construct a skin-friendly, flexible phase-change glycerol hydrogel, offering an alternative to traditional melt impregnation or microencapsulation techniques for preparing phase-change energy storage composites. They synthesized a room-temperature phase-change organohydrogel incorporating phase-change hydrated salts (disodium phosphate dodecahydrate) and polyacrylamide-glycerol hydrogels through a photoinitiated one-step *in situ* polymerization process, as shown in Fig. 3. By embedding the hydrated salt PCMs within the antidiing three-dimensional network of the organohydrogel, they overcame

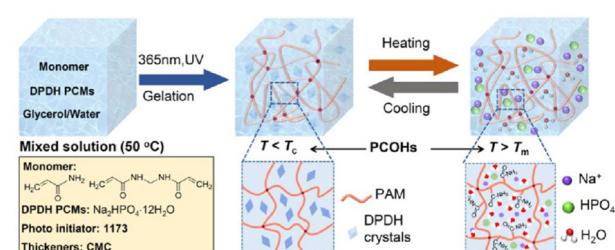


Fig. 3 Preparation process of the polyacrylamide glycerol hydrogels.<sup>16</sup> First, a homogeneous solution of raw materials was prepared by dissolving them at 50 °C, followed by gelatinization through photoinitiated polymerization under UV irradiation at 365 nm. The water within the hydrogels facilitates the melting of DPDH, while the polymer chains physically interact with and capture the phase change materials (PCMs) during the heating and melting process. Upon cooling, the solid DPDH forms, and both the polymer chains and glycerol help retain the additional water.

the rigidity of solid materials and the potential for melting leakage, thereby enabling the flexibility required for wearable thermal management devices. It is important to note that hydrogels, rich in hydrophilic groups such as carboxyl and hydroxyl moieties, undergo hydrolysis upon complexation with hydrated salts. This hydrolysis results in the generation of a significant quantity of ionic species. The interaction between these ionic species and the counterions in the hydrated salt solution induces substantial electrostatic effects, which can influence the phase transition characteristics of the hydrated salts.<sup>14,17</sup> Moreover, the hydrogen bonding interactions between the functional groups on the polymer chains and water molecules within the hydrated salts can further modulate their phase transition temperatures.

Thus, when fabricating phase-change hydrogels using inorganic hydrated salts as PCMs, it is essential to carefully consider the impact of the polymeric base of the hydrogel on the phase transition behavior of the PCMs. Such considerations are pivotal for the strategic selection of constituent materials and the optimization of their respective proportions in the composite formulation, ultimately ensuring the desired performance of the phase-change hydrogel system.

### 2.2. Encapsulation of organic PCMs in hydrogels

Currently, polyethylene glycol (PEG) is the most commonly used organic PCM in hydrogel composites, owing to its amphiphilic nature, which enhances its compatibility and support within hydrogels. For example, Mahdavian *et al.*<sup>18</sup> developed a stable, smart thermal energy storage system by impregnating PEG into the porous structure of modified resorcinol-formaldehyde hydrogels. The strong hydrogen and electrostatic interactions between the aerogel backbone and PEG not only facilitate the impregnation process but also help minimize leakage. In another approach, Yin *et al.*<sup>19</sup> fabricated high-performance, form-stable composite PCMs by incorporating thermally conductive, three-dimensional hierarchical porous carbon carriers. They synthesized polyacrylonitrile (PAN) based hydrogels

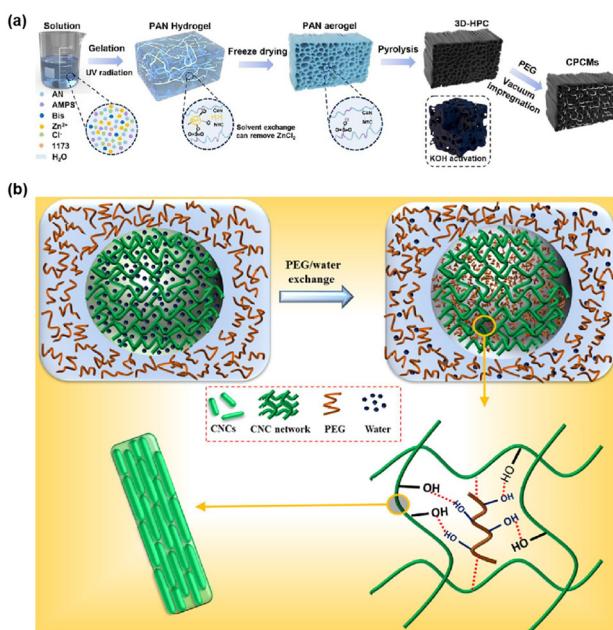
in zinc chloride solution through a one-step photoinitiated polymerization method, followed by the creation of 3D hierarchical porous (3D-HPC) carbon carriers *via* pyrolysis of the freeze-dried PAN hydrogel, as shown in Fig. 4a. This innovative approach enhances both the stability and thermal conductivity of the composite PCMs, showcasing the potential of combining hydrogels with advanced carbon-based materials for superior thermal management applications. The 3D network of the freeze-dried PAN gel creates layered macroporous cavities in the carbonized 3D-HPC, while the hierarchical pore structures (micropores, mesopores, and macropores) formed during the pyrolysis process enhance the PCM storage capacity. These pore structures also improve capillary adsorption and chemical interactions, effectively preventing PCM leakage during melting. The PEG/3D-HPC composite PCMs, with high energy storage density, thermal conductivity, and shape stability, were then prepared *via* vacuum melting impregnation of PEG.

Beyond vacuum impregnation, phase-change hydrogels can also be prepared by directly mixing the PCM with hydrogel monomer components in a one-step synthesis.<sup>21</sup> Another reliable method involves using PEG as both the solvent and phase-change material. For example, Qi *et al.*<sup>22</sup> developed a double-solvent phase-change hydrogel by random copolymerization of acrylic acid and hydroxyethyl methacrylate with water

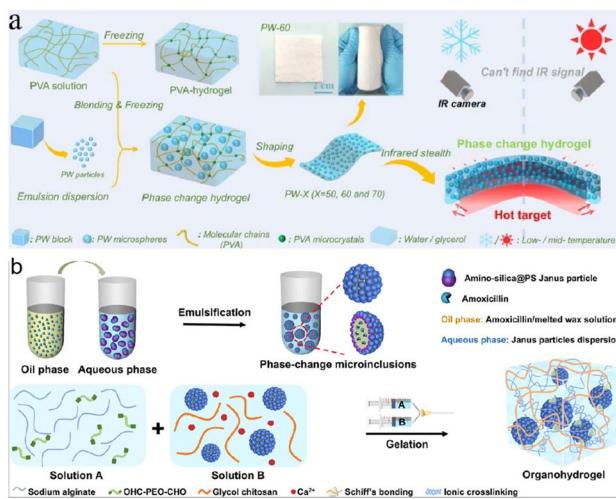
and PEG as solvents. The resulting hydrogel exhibited a high phase-change material loading rate, excellent compression resistance, and superior mechanical properties compared to existing materials. Due to PEG's solid–liquid phase transition behavior, the hydrogel displayed reversible adhesion, transparency, and shape memory, with low thermal conductivity and high energy storage efficiency, making it ideal for infrared stealth applications. Similarly, Zhou *et al.*<sup>20</sup> prepared a cellulose nanocrystal (CNC) hydrogel through UV-induced thiolene click chemistry, followed by the direct impregnation of PEG PCMs using a solvent-exchange strategy. The hybrid PCMs were created by immersing CNC hydrogels in molten PEG until fully saturated, as shown in Fig. 4b. The strong hydrogen bonding between the CNC nanofiber network and PEG chains restricted PEG flow while imparting excellent mechanical strength, further enhancing the performance of the phase-change hydrogel system.

Organic non-polar PCMs, such as paraffin and fatty acids, are often incompatible with typical hydrogels and prone to phase separation, making it challenging to achieve stable phase-change hydrogels. To address this, strategies like introducing surfactants to improve compatibility and structural stability are commonly employed.<sup>23</sup> Alternatively, non-polar PCMs can be encapsulated in microcapsules with appropriate wall materials before being incorporated into hydrogels.<sup>24</sup> Zhou *et al.*<sup>25</sup> developed flexible phase-change hydrogels by integrating phase-change microspheres into a hydrogel substrate. Using the poly(vinyl alcohol) (PVA) hydrogel as the base and paraffin (PW) microspheres as the PCM, they successfully created composite PCMs (Fig. 5a). Paraffin microspheres, fabricated in large quantities *via* the emulsion dispersion method, offer excellent flexibility and adaptability, allowing the phase-change hydrogel to be easily coated on fabrics for wearable infrared stealth applications. Gui *et al.*<sup>26</sup> fabricated multifunctional organohydrogels by incorporating phase-change microinclusions within a double-network hydrogel, stabilized by Janus particles. The silica@polystyrene Janus particles were modified with  $\gamma$ -aminopropyltriethoxysilane to stabilize a wax-in-water emulsion (Fig. 5b). The phase-change microinclusions provided latent heat storage and release capabilities, enabling the hydrogel to regulate skin temperature on wounds through the solid–liquid phase transition of the wax. Moreover, by incorporating medicine into the microcapsules, the hydrogel can release the medication upon reaching the melting point of the wax, making it a promising material for biomedical applications.

Utilizing hydrogels for encapsulating PCMs is an effective strategy for containment, preventing leakage and enhancing cycling stability. PCMs can be integrated into hydrogels through different methods, as listed in Table 1. However, the porosity of the hydrogel matrix plays a critical role in influencing the phase transition behavior of encapsulated PCMs. The surface free energy of the hydrogel matrix, which is positive and dynamic, contrasts with the bulk free energy, creating a balance that significantly affects the thermodynamic properties of the confined crystalline phases. Additionally, factors such as the functional groups on the pore walls, the geometry



**Fig. 4** (a) Schematic illustration of the preparation process of the PAN hydrogel, 3D-HPC and the PEG/3D-HPC CPCMs.<sup>19</sup> PAN hydrogels were synthesized by photo-initiated polymerization from ZnCl<sub>2</sub> aqueous solution of AN and a small amount of hydrophilic monomer 2-acrylamido-2-methyl-1-propanesulfonic acid (AMPS) with the crosslinking agent *N,N'*-methylene bis(acrylamide) (Bis). (b) Schematic diagram of the solvent exchange and intermolecular interaction between PEG and CNC gel.<sup>20</sup> Immersion of CNC hydrogel in molten PEG induces solvent exchange (water replacement) and robust hydrogen-bonding interactions between the CNC network and PEG chains.



**Fig. 5** (a) Schematic illustration of the fabrication of PVA-based flexible phase change hydrogels.<sup>25</sup> The PVA solution and PW microspheres were thoroughly stirred and mixed. The phase change hydrogel was then prepared using the freeze–thaw method, with a control group also prepared by the same method but without PW microspheres. The resulting phase change hydrogel demonstrated infrared stealth properties. (b) Schematic preparation of organohydrogels containing Janus particle stabilized phase-change microinclusions.<sup>26</sup> The aqueous dispersion of amino-modified Janus particle (aminosilica@PS) microspheres was mixed with the oil phase and emulsified to form phase-change microinclusions. These microinclusions were then dispersed into a mixture of GC and CaCl<sub>2</sub> aqueous solution (Solution A). Solution A and Solution B (an aqueous solution of alginate and OHC-PEGCHO) were simultaneously extruded from a mold using a double-tube syringe to fabricate the organic hydrogel.

of the pores, and their aperture sizes can influence the interactions between the PCMs and the pore surfaces, thereby altering the melting point. Modifying the hydrogel structure allows for more precise control over these interactions. In practical applications, carefully selecting or tailoring the porous matrix to meet specific thermal requirements can enable the fine-tuned regulation of the phase transition temperatures of the encapsulated PCMs.

### 3. Regulation of thermal response performance

The thermal response performance of phase-change hydrogels is primarily governed by factors such as thermal conductivity,

phase-change behavior, thermal stability, and thermal responsiveness. To optimize the performance of phase change hydrogels, researchers are focusing on improving their energy storage density, thermal conductivity, and cycling stability by altering the composition, structure, and preparation processes. Heat transfer efficiency depends not only on the thermal conductivity of the hydrogel matrix and phase-change units but also on the interactions between them, which influence interfacial thermal resistance and, consequently, overall heat transfer performance. The phase-change behavior of hydrogels is characterized by their phase transition temperature and phase-change enthalpy. The transition temperature determines their suitability for specific applications, while the enthalpy dictates their energy storage capacity. Thermal stability can be enhanced through structural modifications and various physical treatments, including thermal processing, mechanical stretching, and freeze-drying. Additionally, both thermal conductivity and phase-change behavior significantly impact the hydrogel's thermal responsiveness. The following discussion provides a comprehensive overview of the heat transfer properties, phase-change behavior, and strategies for enhancing the thermal stability of phase-change hydrogels.

Various techniques, such as magnetic and electric field alignment, directional freezing, and mechanical stretching, enable the fabrication of anisotropic hydrogels. Moreover, these structured designs play a crucial role in forming thermally conductive pathways within the hydrogel framework, thereby enhancing thermal transfer efficiency. This improved thermal conductivity is particularly beneficial for applications in thermal regulation and management.

#### 3.1. Methods for improving heat transfer efficiency

Improving the heat transfer efficiency of phase-change hydrogels is crucial for optimizing their applications in the field of thermal management. However, as a polymer matrix, hydrogels inherently possess low thermal conductivity.<sup>51,52</sup> To enhance the thermal transfer properties of phase change hydrogels, the polymer network structure can be strategically engineered. Adjusting the degree of crosslinking and molecular architecture can strengthen interactions between polymer chains, thereby facilitating more efficient thermal energy transfer. Additionally, controlling the pore size and porosity can modulate heat transfer pathways, further optimizing the thermal performance. A widely adopted approach for enhancing thermal conductivity involves incorporating thermally conductive fillers, particularly nanoscale materials,

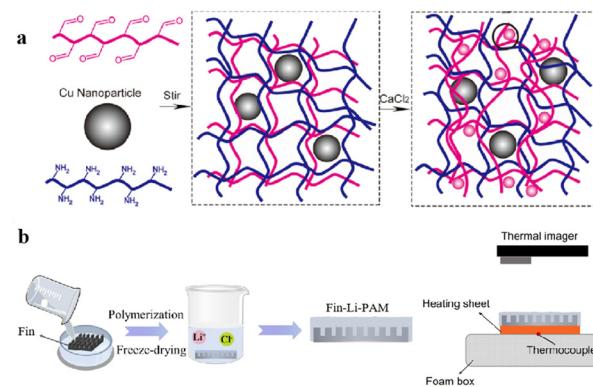
**Table 1** Comparative analysis of fabrication methods for phase-change hydrogels

Method	Advantage	Limitation	Type	Ref.
Solution mixing	Simple process	Limited filler dispersion	Inorganic PCMs	4, 15, 16 and 27–30
Melt impregnation	Uniform network	Complex setup	Organic PCMs	20–22, 25 and 31
Template-assisted 3D-print	Precise pore structure Custom architectures, multi-material	Template removal required Post-processing	Inorganic PCMs	15
			Organic PCMs	18 and 32
				33–43
				44–50

which offer a high surface area-to-volume ratio and effectively improve heat transfer and enable rapid thermal response, thus promoting heat dissipation.<sup>31</sup> These fillers can be broadly categorized into metal, inorganic, and carbon-based materials, each contributing distinct thermal transfer mechanisms, advantages and challenges, as listed in Table 2.<sup>53–55</sup>

Metal fillers possess excellent thermal conductivity, significantly enhancing the heat transfer performance of materials,<sup>71–74</sup> such as silver, copper, and zinc oxide. Sheng *et al.*<sup>56</sup> combined a solid–liquid interpenetrating polymer network with copper nanoparticles to enhance thermal management performance; the distribution schematic of each component can be seen in Fig. 6a. Copper nanoparticles were added to create a thermally conductive pathway, while the interpenetrating polymer network effectively minimized nanoparticle aggregation. Beyond utilizing a single filler type, incorporating multi-dimensional fillers to construct thermal conduction pathways is a widely adopted strategy for further improving thermal performance.<sup>59</sup> For example, Liu *et al.*<sup>58</sup> employed a combination of one-dimensional and two-dimensional geometrical stacking to facilitate thermal energy transfer within a three-dimensional hydrogel matrix. In their design, an ion-crosslinked sodium alginate (SA) hydrogel served as a super-hydrophilic matrix supported by a porous polyurethane (PU) sponge. Graphene oxide (rGO) and silver (Ag) nanowires functioned as photothermal conversion and heat conduction enhancement media. The excellent photothermal and thermal conductivity properties of rGO and Ag nanowires, along with the high aspect ratio of Ag nanowires, facilitated the rapid transfer of thermal energy within the hydrogel.

Integrating metallic fin-based heat transfer structures is another effective approach for enhancing thermal performance.<sup>76,77</sup> Zou *et al.*<sup>75</sup> developed a composite structure consisting of metal fins and hygroscopic salt hydrogel to enhance the passive cooling of solar photovoltaic panels based on evaporative cooling; the preparation process can be seen in Fig. 6b. This modified structure, termed the Fin-LiCl-Polyacrylamide (Fin-Li-PAM) system, was designed to enhance heat transfer within the hydrogel and promote water evap-



**Fig. 6** (a) Schematic preparation of the copper based PNIPAM/Alginate- $\text{Ca}^{2+}$  hydrogel.<sup>56</sup> Copper powder was thoroughly mixed with *N*-isopropylacrylamide, *N,N'*-methylenebisacrylamide, and sodium alginate (SA), followed by polymerization under controlled conditions to obtain a copper-based PNIPAM/alginate- $\text{Ca}^{2+}$  hydrogel. The resulting hydrogel was then immersed in  $\text{CaCl}_2$  aqueous solution to form the copper based PNIPAM/alginate- $\text{Ca}^{2+}$  hydrogel. (b) Schematic illustration of the fabrication of the Li-PAM hydrogel.<sup>75</sup> The PAM hydrogel was immersed in a LiCl solution to form the Li-PAM hydrogel. The Fin-LiPAM structure was prepared by combining a fin heat sink with the hydrogel.

ation. By filling the hydrogel into the spaces between the fins, the heat exchange area was significantly expanded, further boosting heat transfer efficiency. While the incorporation of metal fillers into hydrogels greatly enhances thermal performance due to their superior conductivity, certain challenges must be addressed. Metals have a higher density than hydrogel systems, necessitating measures to prevent sedimentation and aggregation during mixing. Furthermore, their relatively high mass poses limitations in applications where light weight materials are essential.

Inorganic fillers such as alumina, boron nitride (BN), silicon dioxide, and silicon carbide have emerged as crucial ceramic reinforcements for enhancing hydrogel performance.<sup>78–80</sup> A notable advancement was demonstrated by Ji *et al.*<sup>62</sup> through their development of a structural composite hydrogel. This innovative material integrates aligned

**Table 2** Performance contributions of fillers in hydrogel-PCM composites

Filler type	Advantage	Challenge	Representative system
Metal	Ultra-high intrinsic conductivity (Ag: $429 \text{ W m}^{-1} \text{ K}^{-1}$ )	High density, oxidation risk	Cu-PNIPAM/alginate <sup>56</sup> Cu-C/PVA <sup>57</sup> Ag/rGO/SA <sup>58</sup> Ag/Au/trimethyl chitosan <sup>59</sup> Ag NPs/P(AM- <i>co</i> -AA) <sup>60</sup> BNNS/PVA <sup>61</sup> $\text{Al}_2\text{O}_3$ /SA-PAM <sup>62</sup> BN/agarose <sup>63</sup> BN-PDA/PVA/carboxymethylated lignin <sup>64</sup> BN/SA <sup>65</sup> BN/melamine-boric acid <sup>66</sup> Graphene-PAA <sup>67</sup> GO/PAM/hydroxyethyl cellulose <sup>68</sup> CNT/PVA <sup>69</sup> CNT/PNIPAM <sup>70</sup>
Inorganic fillers	High thermal stability	Poor dispersibility	
Carbon-based materials	Light weight, multifunctional	Interfacial phonon scattering	

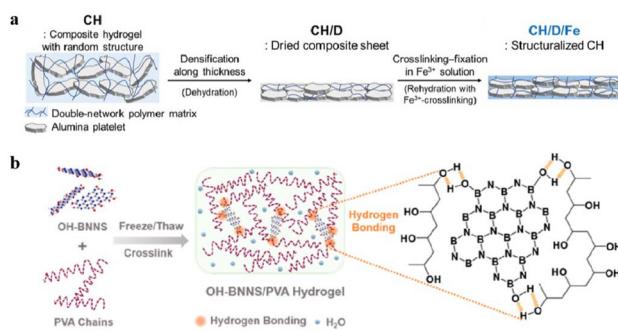


Fig. 7 (a) Design and fabrication of thermal interface hydrogel.<sup>62</sup> The initial composite hydrogel (CH) was first dehydrated and densified, and then dried along the thickness direction. Subsequent rehydration, coupled with  $\text{Fe}^{3+}$  crosslinking, yielded a structurally reinforced composite hydrogel (CH/D/Fe). (b) OH-BNNS/PVA interpenetrating hydrogels were fabricated by a cyclic freeze/thaw process, leveraging hydrogen bonding interactions between the OH-BNNS and PVA chains.<sup>61</sup>

alumina microplatelets within a densely packed alginate-polyacrylamide (SA-PAM) double-network matrix (Fig. 7a), effectively addressing the inherent limitations of conventional hydrogels. The conventional hydrogels typically consist of randomly and sparsely crosslinked polymer networks with significant water content, resulting in weak mechanical properties and limited heat conduction capability. A closer contact and packing of polymer chains would enhance heat conduction through the hydrogel. Moreover, such a configuration can ensure that the hydrogel maintains mechanical robustness when subjected to external loads, such as those imposed by a rigid solid PTM device. Boron nitride, owing to its two-dimensional layered structure akin to graphene and its exceptional thermal conductivity,<sup>81</sup> has earned the moniker “white graphene”, and is consequently frequently utilized as a thermally conductive filler. Jing *et al.*<sup>61</sup> reported hydroxylated boron nitride nanosheets (OH-BNNS)/PVA interpenetrating hydrogels by cyclically freezing/thawing the aqueous mixture of PVA and highly hydrophilic OH-BNNS, as shown in Fig. 7b. Attributed to the synergistic effect of the intrinsic superior properties of the as-prepared OH-BNNS and strong hydrogen bonding interactions between the OH-BNNS and PVA chains, improvements in the thermal diffusivity can be achieved. In fact, the exceptional mechanical properties of BNNS make it a promising candidate to improve the mechanical of hydrogels. It should be noted that the high hydrophobicity and the extremely poor dispersibility of BNNS in aqueous media make it difficult to be homogeneously incorporated into the hydrogel network; therefore, it needs to be modified, such as introducing hydrophilic groups like hydroxyl and amino functional groups. In addition, the general modification is connected through non-covalent bonds, so attention should also be paid to the stability of the hydrophilic groups on the filler.

The primary carbon materials used in thermal management applications include graphene oxide (GO), graphene, carbon nanotubes (CNTs), carbon fiber, *etc.*<sup>82–84</sup> These materials not only exhibit exceptionally high thermal conduction

but also possess low density, making them particularly well-suited for use in electronic devices and related fields.<sup>85</sup> Song *et al.*<sup>86</sup> synthesized a photothermal phase change hydrogel using sodium acetate trihydrate as the PCM, graphene oxide as the photothermal agent, and acrylamide and konjac glucomannan as the support structure. The photothermal conversion efficiency of the hydrogel was enhanced due to the high light absorption capacity of graphene oxide. Moreover, the three-dimensional network structure of the hydrogel facilitated the formation of a continuous heat transfer pathway, further improving its thermal conductivity. However, graphene oxide contains a large number of oxygen-containing groups and its contribution to heat conduction is limited. It is usually necessary to restore it to obtain higher heat transfer efficiency. Park *et al.*<sup>87</sup> fabricated a self-assembled granular hydrogel (GH) using GO-coated agarose microbeads (GAMs) as precursor materials. They further thermally reorganized the GH into a highly conductive hydrogel, forming a thermally annealed graphene-channelled agarose hydrogel (TAGAH) with a fully connected three-dimensional reduced GO network, as shown in Fig. 8a. This continuous graphene pathway significantly improves heat transfer efficiency within the hydrogel, thereby enhancing its thermal conductivity. It proved that by strategically arranging a minimal amount of thermally conductive carbon fillers to form an interconnected thermal-conducting network, it is possible to enhance both thermal and electrical conductivity.<sup>88</sup> Yang *et al.*<sup>67</sup> obtained a novel thermal interface

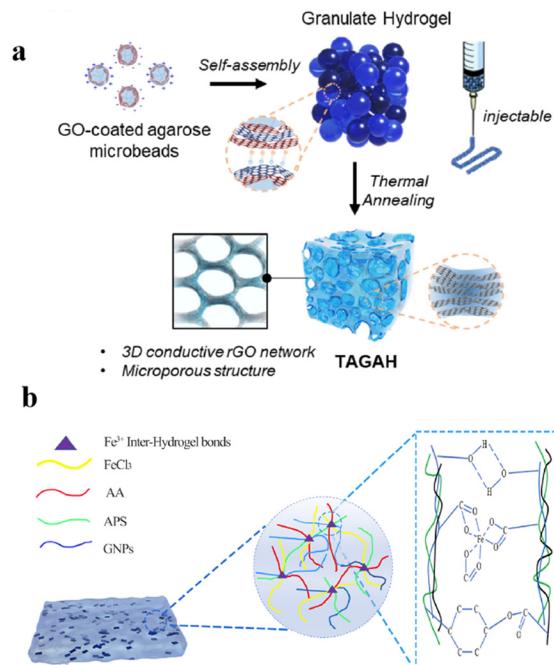


Fig. 8 (a) Schematic illustration of the fabrication of a thermally annealed graphene-connected agarose hydrogel.<sup>87</sup> GO-coated agarose microbeads were self-assembled to form GHs, and after thermal annealing, TAGAH was formed. (b) Illustration of the synthesis of self-healing hydrogel composites loaded with graphene nanoplatelets;<sup>67</sup> dynamic cross-linking exists between the PAA chains and  $\text{Fe}^{3+}$ .

material by incorporating graphene into a polyacrylate-based (PAA) double cross-linked hydrogel, as shown in Fig. 8b. The introduction of graphene enhances both the thermal conductivity and tensile properties of the hydrogel, resulting in a highly efficient thermal interface material with low interfacial thermal resistance. This effective heat dissipation mechanism makes it a key component in thermal management applications. To achieve multifunctionality, such as improved photothermal conversion performance, multiple heat conduction enhancement mechanisms can also be incorporated into the system, offering further potential for optimizing thermal management materials. For example, Luo *et al.*<sup>69</sup> improved the dispersibility of CNTs *via* metal-phenolic coordination chemistry-based interfacial engineering, and incorporated it into the PVA hydrogel. CNTs have more engineering advantages in constructing three-dimensional thermal conductivity networks, interface optimization, and anisotropic control. They are particularly suitable for composite material systems that require high directional thermal conductivity, low filler addition amount, and synergistic enhancement of mechanics and thermal conductivity. Furthermore, the synergistic effect of the metal-phenolic networks and CNTs can further enhance the light absorption characteristics of the hydrogel, making it have potential application value in fields such as solar evaporators.

In summary, incorporating high-thermal-conductivity fillers can significantly enhance the thermal transport properties of hydrogels, making them promising materials for thermal management applications. However, a key challenge lies in the poor compatibility between most thermally conductive fillers and the polymer matrix of hydrogels. To address this, surface modification of the fillers is essential to improve their dispersion within the hydrogel network. The introduction of hydrophilic functional groups can inevitably impact the thermal conductivity of the hydrogel, making it crucial to optimize the degree of modification during material design. Furthermore, for nanoscale fillers, their inherent tendency to aggregate must be carefully managed, as uncontrolled clustering can lead to uneven heat dissipation or even localized heat accumulation, ultimately compromising the hydrogel's performance.

### 3.2. Regulation of heat capacity and phase transition temperature

The enthalpy and phase change temperature range of phase change hydrogels play a crucial role in their thermal management applications. Most phase change energy storage materials exhibit a broad and tunable phase change temperature range. Typically, inorganic phase-change materials have relatively high phase change temperatures, while organic counterparts are more commonly employed in applications requiring moderate to low temperatures. Notably, for certain organic phase-change materials such as PEG and paraffin, their phase change temperatures and enthalpy values are closely linked to the length of their carbon chains. This correlation arises from factors such as intermolecular forces, the

number of chemical bonds, and the degree of molecular structural orderliness.<sup>89–91</sup> Generally, phase-change materials with longer carbon chains tend to exhibit higher phase change temperatures and enthalpy values.<sup>92</sup> However, achieving both high enthalpy values and low phase change temperatures simultaneously remains a significant challenge, often requiring careful material design and optimization.

Optimization of the phase transition temperature of phase change hydrogels can be achieved through various strategies, including chemical modification, selection of eutectic phase-change materials, nanocomposite technology, regulation of the hydrogel network structure, and thermal treatment processes.<sup>93–96</sup> For example, through cascaded latent heat storage design for a variety of PCMs with different phase change temperatures, Abhijith *et al.*<sup>97</sup> prepared a stable caprylic acid and cetyl alcohol organic binary mixture as a solid–liquid phase change material with a phase transition temperature in the range for exotic chilled refrigeration; the binary mixture can be used to maintain the temperature at 10–15 °C for food storage, transportation of pharmaceuticals, or storage in supermarket display cabinets. It should be noted that the reliability of thermal properties for melting/freezing cycles is important when it comes to application. Ma *et al.*<sup>98</sup> combined the tailorabile phase-change ability of comb/bottle-brush networks, recyclability of polysiloxane and effective photo-induced thiol–ene click reaction, and the crystalline polysiloxane networks were efficiently prepared by mixing polymethylvinylsiloxanes with different vinyl contents, 1-octadecanethiol and 1,6-hexanedithiol, followed by UV irradiation. The grafting crystalline alkyl chain content can effectively regulate the latent heat, phase-change temperature, and thermomechanical and viscoelastic properties of networks. In addition, improving the force between the phase change material and the carrier can effectively increase the load ratio of the phase change material. He *et al.*<sup>99</sup> employed a one-step substitution reaction followed by a thermal reduction technique to successfully affix Ni and Cu nanoparticles onto reduced rGO nanosheets, which were subsequently integrated into a nickel foam (NF) matrix. This process resulted in the fabrication of an NF/Ni-Cu@rGO porous carrier, facilitating the adsorption of PEG and the formation of CPCMs. Introducing Ni-Cu@rGO into the NF matrix enhanced the carrier framework, enabling improved PEG loading and leakage prevention.

Machine learning (ML) techniques play a crucial role in optimizing the performance of PCMs, particularly in thermal management applications for electronic devices. As illustrated in Fig. 9, ML-driven approaches enable the efficient design and optimization of PCM structures and material properties, enhancing thermal performance and reliability.<sup>100–102</sup> By leveraging ML models, researchers can predict the thermal response of PCMs based on their physical properties and operating conditions, guiding material selection and design. For instance, analyzing key parameters such as thermal conductivity, phase transition temperature, and specific heat capacity allows ML models to forecast PCM performance in practical applications, aiding in the selection of optimal materials.<sup>103–105</sup>

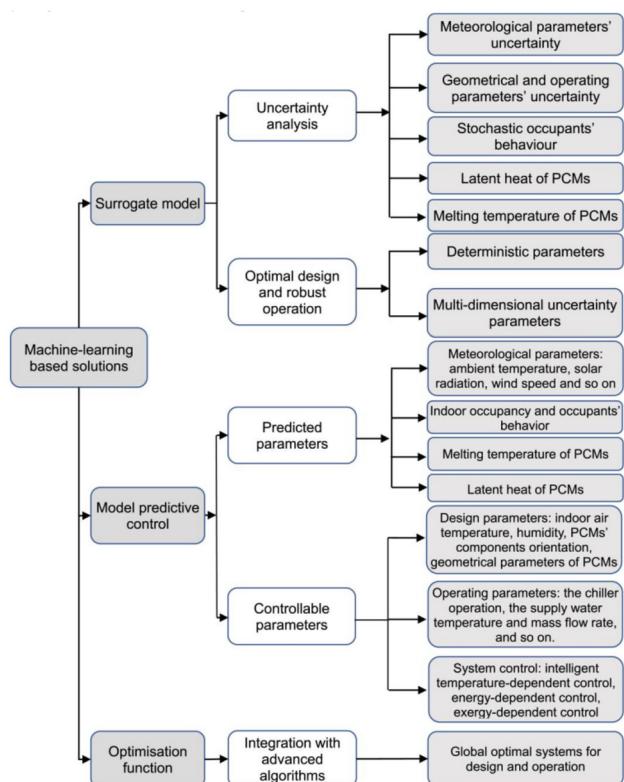


Fig. 9 Application of the machine learning method in novel PCMs cooling systems.<sup>110</sup>

Additionally, ML facilitates multi-objective optimization in thermal management systems, balancing cooling efficiency and energy consumption. Artificial Neural Networks (ANNs) and other ML models can predict PCMs' thermal behavior under various conditions, enabling adaptive thermal management strategies.<sup>106,107</sup> Furthermore, ML-driven intelligent thermal management systems can monitor and predict device temperatures in real time, optimizing cooling system performance. By accurately modeling the melting and solidification behaviors of PCMs, ML enhances control over electronic device thermal states, improving efficiency.<sup>108,109</sup> ML techniques also enable rapid prediction of PCMs' thermal performance within electronic packaging, which is critical for minimizing peak temperatures and temperature fluctuations. The ability of ML algorithms to process complex, multi-dimensional datasets reveals intricate relationships between microstructural mechanisms and macroscopic PCMs' performance. This deeper understanding facilitates the development of advanced thermal management materials and systems. Moreover, ML-assisted optimization algorithms refine the geometric structure and material composition of PCMs, further enhancing their effectiveness. In summary, ML offers a powerful tool for advancing phase change energy storage research and applications, driving scientific and technological progress while fostering industrial development.

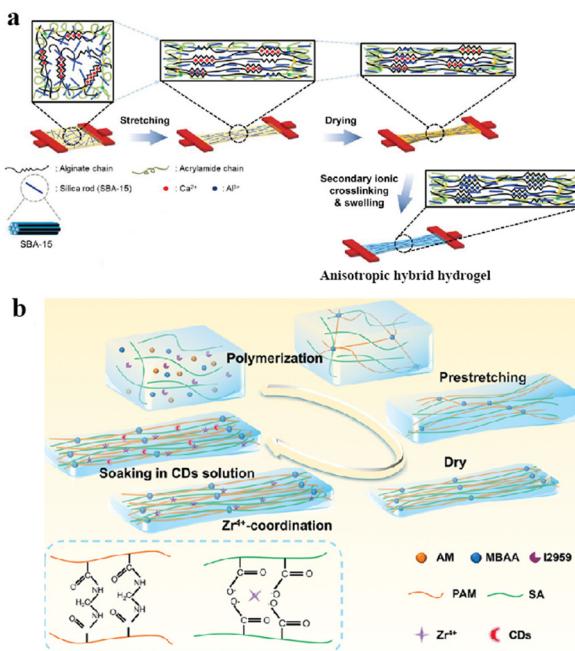
The adaptable properties of hydrogels enable them to function as energy storage materials through strategic structural

design. Therefore, tuning the enthalpy values and phase transition temperature ranges of phase change hydrogels require careful consideration of both the hydrogel matrix and the incorporated phase change energy storage materials.

### 3.3. Optimization of cycling stability and durability

For the application of phase change hydrogels in thermal management, beyond thermal conductivity and phase change behavior, recyclability and durability are also crucial factors. By fine-tuning the network structure of hydrogels, their mechanical strength and stability can be significantly enhanced to meet the demands of long-term and repeated use. Key strategies for reinforcing the mechanical integrity of hydrogels include the deliberate construction of covalent and non-covalent cross-linked networks,<sup>111–113</sup> the controlled induction of microphase separation to create tailored domain morphologies,<sup>114</sup> and the utilization of supramolecular interactions, such as hydrogen bonding, van der Waals forces, and hydrophobic interactions, to improve the overall stability and mechanical robustness.<sup>115–117</sup> Other key influences include the cross-linking density of the hydrogel matrix, the presence of thermally stable groups, the incorporation of nanofillers, surface modifications, and the interactions between phase change energy storage materials and the hydrogel.

For example, inspired by the multi-scale structural features of tendons and ligaments, Choi *et al.*<sup>118</sup> fabricated a hybrid hydrogel with a hierarchically anisotropic structure and superior mechanical properties. In their approach, alginate-polyacrylamide double-network hydrogels were reinforced with high-aspect-ratio mesoporous silica microparticles. By employing a combination of stretching, controlled drying, and ionic crosslinking, the researchers achieved a multiscale structure, where an anisotropically aligned polymer network was embedded with uniformly aligned microparticles, as illustrated in Fig. 10a. Wang *et al.*<sup>119</sup> developed a bilayer actuator with high stretchability and robust interfacial bonding, consisting of an adhesive passive layer copolymerized with amphoteric  $[(2\text{-methacryloyloxyethyl})\text{dimethyl-(3-sulfopropyl)}]$ , acrylic acid (AA), and an active hydrogel layer of poly(*N*-isopropylacrylamide) containing polydopamine-modified MXene and calcium chloride. The coordination between carboxylate and  $\text{Ca}^{2+}$  at the interface enhances bonding, enabling resistance to large strain and preventing delamination. Beyond strengthening intermolecular interactions, increasing chain entanglement also improves hydrogel mechanics. It can be seen that the adoption of a layered structure design enhances the performance of hydrogels, while anisotropic structures contribute to high tensile strength and ductility. Layered structures have been shown to improve the mechanical resilience of phase-change hydrogels by facilitating energy dissipation upon fracture. Zhong *et al.*<sup>120</sup> introduced a simple dehydration-induced entanglement strategy. In this approach, a hydrogel with low cross-linking is slightly dehydrated, forming long polymer chains that entangle more readily than shorter chains, enhancing stiffness. These entanglements dissipate energy during large deformations, improving fracture tough-

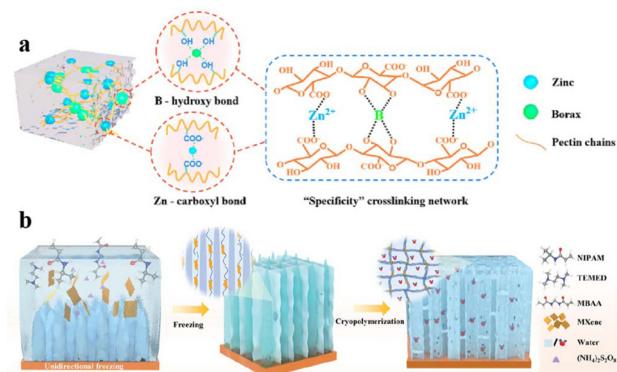


**Fig. 10** (a) Schematic preparation of a hierarchically anisotropic hydrogel inspired by natural tendons or ligaments.<sup>118</sup> Using alginate and polyacrylamide as the base materials and SBA-15 as the inorganic filler, the hydrogel achieved a unique anisotropic hierarchical structure through anisotropic reshaping, followed by drying and ionic cross-linking. (b) Preparation procedures of anisotropic PSZC hydrogels.<sup>129</sup> The cross-linked PAM/SA hydrogel was stretched to align the molecular chains, and then dried to stabilize the arranged structure.  $Zr^{4+}$  ions were introduced for metal cross-linking, and the prepared gel samples were incubated in the CDs solution. This fluorescent hydrogel, similar to human muscle fibers, has strong rigidity and anti-expansion capabilities.

ness and fatigue resistance. Unlike complex methods requiring specialized microstructures or fiber/matrix composites,<sup>121–127</sup> this technique offers a simpler alternative. In fact, by precisely controlling the dehydration process of dynamic hydrogels, the stress induced by dehydration can promote the structural reorganization of these materials at both the micro and macro levels, offering new perspectives for the design of novel biomaterials and manufacturing technologies.<sup>128</sup>

The mechanical properties of hydrogels can be tailored by adjusting the degree of stretching, the concentration and functionalization of inorganic microparticles, and the type of crosslinking cations. Inspired by the biomechanical anisotropy of muscle fibers, Zhang *et al.*<sup>129</sup> developed a mechanically robust and multifunctional anisotropic polyacrylamide/sodium alginate/zirconium ion/carbon dots (PAM/SA/ $Zr^{4+}$ /CDs, PSZC) hydrogel through the synergistic effect of mechanical-assisted stretching,  $Zr^{4+}$  metal-coordination and CD embedding, as presented in Fig. 10b. Attributed to the oriented alignment of PAM and SA molecular chains induced by mechanical-assisted stretching and metal-coordination, the resulting hydrogel exhibited an impressive tensile strength; the mechanical properties can be controlled by the pre-strain, crosslinker content. The highly ordered structure facilitated stress trans-

mission along the stretching direction, minimizing energy dissipation and imparting exceptional resilience. A common approach to enhance the mechanical properties of hydrogels involves the implementation of specific crosslinking strategies. For example, Yang *et al.*<sup>130</sup> prepared a hydrogel from a mixture of pectin and borax, followed by immersion in a zinc acetate solution; a superionic conductive hydrogel with a Zn-carboxyl physical crosslinking and B-hydroxyl chemical crosslinking network can be achieved, as shown in Fig. 11a. This approach allows for tailored molecular-level crosslinking, where pectin serves as the polymeric backbone. Specific crosslinking is achieved through dynamic Zn-carboxyl physical bonds and B-hydroxyl chemical bonds.<sup>131,132</sup> The Zn-carboxyl bonds provide stability under significant strain, while the B-hydroxyl chemical bonds are dynamic, continuously dissociating and associating during stretching, which enhances the hydrogel's durability. The crosslinking strategy significantly impacts the mechanical properties of hydrogels. By incorporating covalent or non-covalent crosslinks within the polymer network, the hydrogel's mechanical stability, strength, and toughness can be fine-tuned. Covalent crosslinks, formed through chemical reactions, create permanent bonds between polymer chains, enhancing resistance to deformation and improving structural integrity. In contrast, non-covalent crosslinks, such as hydrogen bonds, ionic interactions, and van der Waals forces, provide reversible, dynamic interactions that contribute to self-healing and adaptability. The density and distribution of these crosslinks are crucial factors influencing the hydrogel's balance between elasticity and brittleness, as well as its response to external forces.



**Fig. 11** (a) Structure of stretchable ionic hydrogels.<sup>130</sup> The mixture of pectin and borax was poured into a glass mold and dried at 60 °C to form a pectin hydrogel film, and then the pectin hydrogel film was immersed in a zinc acetate solution to achieve SICH with a Zn-carboxyl physical crosslinking and B-hydroxyl chemical crosslinking network. (b) Schematic diagram of the synthesis process of  $Ti_3C_2T_x$  MXene-based nanocomposite hydrogels.<sup>133</sup> NIPAM monomers were mixed with the MXene suspension, and then the PNIPAM/MXene (PM) hydrogel was prepared with a crosslinker and initiator under an argon atmosphere. The PMD hydrogel was presented with an orderly internal orientation structure by the directional freezing method using liquid nitrogen. Finally, the composite hydrogels with ordered orientation filled with water were acquired as the ice crystals melted.

The synergistic effects of various interaction forces to construct hydrogels with enhanced mechanical properties have also been frequently reported. Luo *et al.*<sup>134</sup> reported a synergistic confinement strategy of ionic, covalent and hydrogen bond crosslinking for manufacturing flexible and form-stable inorganic hydrated salt gels. By constructing a double-gel network to anchor the hydrogel, sodium sulfate decahydrate was homogeneously dispersed within the matrix, stabilized by hydrogen bonding with the gel network and electrostatic attraction to the calcium alginate framework. These multi-scale interactions endowed the hydrogel with excellent form stability and cycling durability. Incorporating nanofillers with high mechanical strength further enhances energy dissipation, leading to a toughening effect.<sup>132,135</sup> Wang *et al.*<sup>136</sup> developed tough hydrogels *via* stretch-induced alignment of silver nanowires, deformable liquid metal, and PVA nanocrystals. The integration of these components effectively mitigated stress concentration at crack tips, imparting crack propagation insensitivity and fatigue resistance to the hydrogel.

The phase change hydrogels prepared by the above-mentioned chemical methods vary in performance and application scenarios, as listed in Table 3. In addition to the chemical methods, the mechanical properties of hydrogels can also be improved by using physical methods such as the ice template method and magnetic field orientation.<sup>137–139</sup> Inspired by the pulvinus of *Mimosa pudica*, Yan *et al.*<sup>133</sup> developed  $Ti_3C_2T_x$  MXene-based nanocomposite hydrogels by a directional freezing method, as shown in Fig. 11b. The oriented arrangement of polymers and nanosheets inside the hydrogel gives it an ordered and delicate structure that can withstand more forces, thus exhibiting superior mechanical capabilities when forces are directed parallel to the hydrogel network's direction. Similar to this method of improving the stability of hydrogels by constructing energy dissipation paths, applying mechanical stretching during the preparation or post-treatment of hydrogels to induce better alignment and cross-linking of polymer chains can also improve their thermal stability. Other common physical methods to enhance the stability of hydrogels include appropriate thermal treatment, such as thermal curing or crosslinking, which can strengthen the stability of their network structure and consequently enhance their thermal

stability. Thermocompression treatment of hydrogels can promote close contact and partial crosslinking between polymer chains, thereby improving their thermal stability. Alternatively, stress relaxation treatment can release residual stresses within the hydrogel, enhancing its stability during thermal cycling. Physical methods can be used independently or in combination with chemical modification methods for better results. Enhancing the thermal stability of phase change hydrogels through physical methods allows for the optimization of performance without altering their chemical composition.

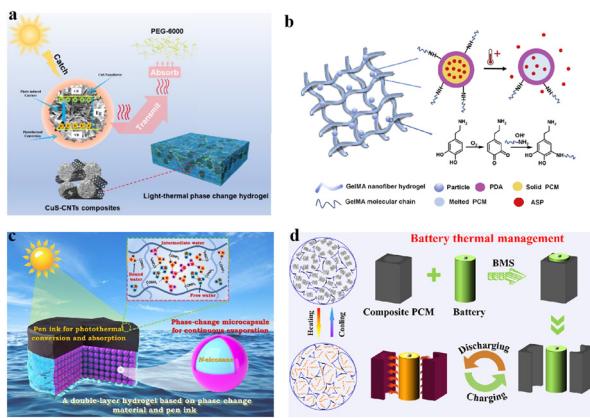
## 4. Application of phase change hydrogels

The application of phase change hydrogels in the fields of electronic equipment cooling and building energy conservation has been widely reported.<sup>142</sup> Electronic components in electronic devices can suffer performance degradation due to exposure to high heat fluxes.<sup>143</sup> Several typical applications of phase change hydrogels are presented in Fig. 12. With the capacity to absorb and store significant amounts of thermal energy, phase change hydrogels can prevent equipment from overheating, extending the lifespan of devices, and enhancing energy efficiency. In the construction area, phase change hydrogels can be integrated into windows, walls, roof insulation materials, floors, and ceilings to modulate light transmittance and thermal energy permeability in response to external temperature fluctuations,<sup>144</sup> thereby balancing indoor temperatures, improving indoor comfort, reducing the thermal load on buildings, and decreasing the energy consumption of air conditioning and heating systems.<sup>32,145,146</sup>

Additionally, phase change hydrogels can serve as thermal energy storage media in solar water heating systems, improving system efficiency and stability. For example, Tan *et al.*<sup>31</sup> designed and prepared a photothermal-thermal storage integrated system with excellent light absorption and photothermal conversion performance based on the sodium alginate (SA) hydrogel combined with photothermal materials nano-flower structure CuS and pure polyethylene glycol-6000

**Table 3** Comparative analysis of hydrogel-based PCM systems

System composition	Thermal conductivity ( $\text{W m}^{-1} \text{K}^{-1}$ )	Phase-change enthalpy ( $\text{J g}^{-1}$ )	Cycling stability (cycles)	Application	Ref.
NaAc·3H <sub>2</sub> O/PAM hydrogel	1.2	202.4	300	Solar energy storage	15
PEG/3D-HPC carbon hydrogel	—	109.1	150	Building, electronic device regulators	18
Paraffin@PVA microsphere hydrogel	0.17	126.0	200	Infrared stealth, wearable devices	25
SAT/PDA/PAM	—	180.0	100	Building, wearable devices	140
PEG/SA	0.492	150.8	300	Solar storage systems	31
Na <sub>2</sub> SO <sub>4</sub> ·10H <sub>2</sub> O/MXene/PAM	—	163.3	200	Wearable devices, healthcare devices	4
Na <sub>2</sub> SO <sub>4</sub> ·10H <sub>2</sub> O/MXene-Ag/PAM/SA	—	176.0	800	Wearable devices, healthcare devices	27
SAT/expanded graphite/PAM/chitosan	1.153	119.7	50	Personalized healthcare	29
Na <sub>2</sub> SO <sub>4</sub> ·10H <sub>2</sub> O/PAM-PAA	—	133.3	500	Building	28
Na <sub>2</sub> SO <sub>4</sub> ·10H <sub>2</sub> O/PAM/SA	—	181.4	10	Desalination	30
PEG/sodium polyacrylate/cement	—	15.74	—	Cooling pavement	141



**Fig. 12** (a) Phase change hydrogel for photothermal-thermal storage.<sup>31</sup> PEG-6000 in the hydrogel undergoes a solid–liquid phase change during heating, absorbing heat from the hot plate and thus suppressing the temperature rise. (b) Phase change hydrogel for wound dressing.<sup>24</sup> At the eutectic point, the PDA nanoparticles were retained in the solid, which can prevent the ASP from divulging through spread. When temperature of the wound bed rises, the nanoparticles will be heated, resulting in diffusion of fatty acids and releasing of the loaded drug. (c) Phase change hydrogel for seawater desalination.<sup>147</sup> A pen ink layer as a solar photothermal responsive component was covered on the upper surface of the hydrogel to convert solar photothermal energy to drive seawater evaporation. (d) Phase change hydrogel for battery thermal management.<sup>148</sup> CPCM absorbs and stores the heat generated during the battery discharge, thereby lowering the battery temperature, and releases the stored heat during the charging process.

(PEG-6000), as shown in Fig. 12a. The hierarchical pore structures of hydrogel provide a good structural basis for the encapsulation of PCMs and offer opportunities for the design of phase change energy storage hydrogels. The light-thermal phase change hydrogel has great potential in solar energy storage systems, providing insights into the functional integration design of PCMs.

Owing to the flexibility of phase change hydrogels, it has been investigated for thermal management applications. For instance, in aircraft or automobiles, phase change hydrogels can be utilized to regulate cabin temperatures or absorb heat generated by electronic devices during operation to prevent overheating. And they are capable of storing thermal energy to provide warmth when temperatures drop. For example, in the aerospace field, phase-change energy storage hydrogels can be used in the thermal control system of satellites or space stations by absorbing and releasing heat to maintain the normal operating temperature of equipment.<sup>12,149,150</sup> In the field of biomedicine, this material can also be used in drug delivery systems, controlling the release rate and amount of drugs through temperature-responsive behavior.<sup>151–155</sup> Zhang *et al.*<sup>24</sup> fabricated an extracellular matrix-like methacrylate gelatin nanofibrous hydrogel *via* electrospinning and UV-cross-linking, followed by a depot of fatty acids/aspirin (ASP) encapsulated polydopamine. With the temperature regulation performance of fatty acids, the as-prepared fibrous hydrogels showed a significantly faster ASP release at 40 °C compared

with those at 25 °C and 37 °C, and an “on-off” drug release was achieved at heating and cooling cycles between 40 °C and 25 °C, as illustrated in Fig. 12b. Such a thermosensitive drug-releasing nanofibrous hydrogel demonstrates considerable promise for wound care. Furthermore, the contribution of phase change hydrogels to sustainable development and environmental protection also cannot be ignored. Zheng *et al.*<sup>147</sup> developed a new double-layered hydrogel based on polyacrylamide, *n*-eicosane (core)@TiO<sub>2</sub> (shell) phase-change microcapsules, and pen ink to construct a hydrogel-based evaporator for efficient and continuous seawater desalination, as presented in Fig. 12c. The phase-change microcapsules dispersed in the lower layer of the hydrogel can act as a latent heat-storage unit to store a great quantity of photothermal energy through melting phase change by the phase change material core. This imparts a consecutive evaporation capability to the developed hydrogel-based evaporator.

It is evident that in recent years, research on phase change hydrogels has made remarkable progress. Whether it is cooling of electronic devices,<sup>156</sup> personal thermal management,<sup>135</sup> healthcare,<sup>157,158</sup> or solar thermal energy storage,<sup>159</sup> the features of temperature controllability, excellent flexibility, and plasticity of phase change hydrogels provide them with unique advantages in the field of thermal management.

## 5. Conclusions and outlook

This paper reviews recent advances in phase change hydrogels for thermal management, focusing on fabrication strategies, performance enhancement, and applications. With excellent thermal storage and regulation capabilities, these hydrogels show notable efficacy in building energy conservation, electronics cooling, and healthcare. However, the following challenges and unresolved issues still exist.

There are several scientific barriers that currently hinder the broader application of phase change hydrogels. In extreme environments, these materials often exhibit compromised stability. For instance, high humidity can induce hydrogel swelling, leading to structural collapse, while intense irradiation (*e.g.*, UV or  $\gamma$ -rays) accelerates polymer chain degradation. Additionally, exposure to extremely low temperatures can damage the hydrogel structure and significantly diminish its mechanical properties. The synergistic effects of multiple stressors, such as thermal cycling, humidity, and mechanical loading, have received limited attention and remain poorly understood. Furthermore, few studies have systematically assessed the long-term durability of these materials, particularly regarding the decay of phase change enthalpy and mechanical fatigue under real-world operating conditions. A further challenge lies in the interfacial mismatch between organic hydrogel matrices and inorganic phase change fillers, which frequently leads to delamination during repeated phase transitions, especially under shear stress.

Moving forward, there are four key perspectives that we would like to propose:

### (1) Fabrication strategies and structural control

The fabrication strategies of phase change hydrogels play a crucial role in determining their microstructure, phase transition behavior, and overall performance. Various synthesis methods, including solution mixing, radiation crosslinking, and template-assisted approaches, have been employed to fine-tune their structural properties. Moving forward, integrating computational modeling and simulation techniques into the design process presents a promising direction. These tools enable the prediction of the effects of different crosslinking and modification strategies on the mechanical, thermal, and phase change properties of hydrogels, thereby guiding experimental optimization.

Future advancements should focus on developing precise fabrication techniques that offer enhanced structural control at multiple scales. This includes hierarchical structuring to improve phase change efficiency, tunable porosity for optimized thermal conductivity, and responsive architectures that dynamically adapt to external stimuli. Additionally, incorporating smart self-healing mechanisms and bioinspired designs could further enhance the durability and functionality of phase change hydrogels, paving the way for their expanded applications in energy-efficient buildings, electronic cooling, and wearable thermal management.

Furthermore, there are three critical challenges that must be overcome in fabrication:

First, the development of environmentally adaptive designs requires the creation of temperature- and humidity-resistant crosslinking networks (*e.g.*, zwitterionic polymers) and radiation-tolerant matrix materials (*e.g.*, aromatic-rich backbones) without compromising the phase transition enthalpy. Second, achieving multi-scale structural control remains problematic, as current methodologies struggle to simultaneously regulate grain size (10–100 nm), mesoporous architecture (100–500 nm), and macroscopic morphology, which is an essential prerequisite for enabling hydrogel applications under extreme conditions. Third, inherent contradictions between process stability constraints must be resolved, particularly the competing demands of high filler loading for enhanced thermal capacity *versus* maintained processability, necessitating innovative manufacturing approaches.

### (2) Performance enhancement techniques

Improving the thermal conductivity, stability, and durability of phase change hydrogels is essential for their practical applications. While significant strides have been made in enhancing their thermal response speed, several challenges remain, including stability issues in real-world environments and the limited application range of inorganic hydrated salt-based phase change materials for medium- and low-temperature thermal energy storage. For instance, in humid environments, the plasticization effect of moisture on polymers reduces mechanical modulus and accelerates leakage of PCMs. This necessitates the development of novel hydrophobic modification strategies combined with moisture-resistant structures to enhance the hydrothermal stability of hydrogels. Additionally, under prolonged intense ultraviolet (UV)

irradiation, conventional hydrogels suffer from chain scission, leading to reduced enthalpy. Thus, exploring radiation-resistant systems incorporating  $\pi$ -conjugated polymers or semiconductor nanoparticles is critical. Furthermore, improving the electrochemical performance and mechanical stability of hydrogels remains a key area for future research. Concurrently, current evaluations of cycling stability lack standardized protocols, such as accelerated aging tests integrating temperature fluctuations, humidity cycling, and mechanical deformation.

Future improvements in phase change hydrogels should focus on several key aspects: first, enhancing thermal conductivity through chemical modifications and nanocomposite strategies to improve heat transfer efficiency. Second, optimizing molecular structures and crosslinking networks to boost both thermal stability and cycling durability. Third, exploring new phase change energy storage materials that incorporate smart, responsive features, enabling adaptive thermal regulation based on changes in temperature, humidity, or light exposure. Fourth, developing hydrogels derived from renewable and biodegradable materials to promote sustainability and minimize environmental impact. Lastly, conducting cost-benefit analyses for large-scale production to enable the commercialization of phase change hydrogels for applications in building energy efficiency, electronic device cooling, and personal thermal management.

### (3) Thermal management applications

Phase change hydrogels are increasingly recognized for their role in energy-efficient solutions, driving innovation in thermal management. Future research should focus on integrating multifunctional capabilities, such as photothermal and thermoelectric conversion, to enhance thermal management efficiency and broaden applications, including information encryption. Additionally, improving the environmental adaptability of these hydrogels will ensure their performance in extreme environments, meeting diverse needs. For example, intense solar radiation in desert regions or corrosive seawater salts in marine environments can pose significant challenges to the structural integrity of hydrogels. So future research must prioritize developing standardized testing protocols that replicate real-world extreme conditions rather than ideal laboratory environments. Particular emphasis should be placed on *in situ* characterization techniques to understand degradation mechanisms. In addition, developing hydrogels from renewable, biodegradable materials will reduce environmental impact and support sustainability. Incorporating smart responsive technologies (*e.g.*, temperature, humidity, and light) will enable dynamic thermal management, making hydrogels more versatile for applications in personal thermal management, building insulation, and electronics. Finally, large-scale production and commercialization, supported by cost-benefit analyses and the integration of smart thermal management systems, will help phase change hydrogels become widely adopted across industries.

### (4) Challenges and strategies of industrialization

To advance phase change hydrogels to practical applications, it is imperative to establish a standardized evaluation system for their performance, such as assessing their long-term service life

under composite environmental conditions (e.g., thermal shock, humidity cycling, and mechanical deformation). Furthermore, the development of novel materials that overcome the limitations of conventional PEG/paraffin systems is essential, such as bio-based lipid materials and eutectic materials.

However, industrial-scale production of phase change hydrogels faces multifaceted challenges. For instance, when regulating phase-change behavior by designing hydrogel microstructures (e.g., networked scaffolds and layered sandwich structures), structural inhomogeneity often arises, compromising performance stability. Similarly, incorporating functional fillers to enhance specific properties may lead to difficulties in balancing mechanical performance and functionality. Additionally, multistep processes (e.g., stepwise polymerization and post-processing) introduce complexity and reduce production efficiency. While *in situ* monomer polymerization is a common approach for large-scale manufacturing, its reliance on stringent reaction conditions (e.g., the concentrations of monomer, promoter, and initiator; temperature; atmosphere) can result in gelation failure during scale-up.

To address these challenges, optimizing material design and fabrication processes is critical. This includes simplifying structural configurations to minimize dependence on intricate procedures, adopting efficient and green synthesis methods (e.g., one-pot synthesis and photopolymerization) to reduce energy consumption and operational steps, and leveraging dynamic crosslinking mechanisms (e.g., hydrogen bonds and ionic interactions) to enhance mechanical and environmental stability. Innovations in production equipment, such as integrating 3D printing or continuous flow reactors, could enable precise structural control and high-throughput manufacturing. Concurrently, automated monitoring systems should be developed to dynamically regulate reaction parameters (e.g., temperature and pH) in real time, ensuring product uniformity.

## Author contributions

Zhuoni Jiang: conceptualization, writing – original draft, writing – review & editing, and funding acquisition. Jian Tang: data curation, and investigation. Chaochao Zhang: investigation and data curation. Yiping Xie: writing – review & editing, investigation, and validation. Jinxing Chen: conceptualization, writing – review & editing, supervision, funding acquisition, and validation.

## Conflicts of interest

There are no conflicts to declare.

## Data availability

No primary research results, software or code have been included and no new data were generated or analysed as part of this review.

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