Materials Advances

This article can be cited before page numbers have been issued, to do this please use: S. Awasthi, *Mater. Adv.*, 2026, DOI: 10.1039/D6MA00167J.

View Article Online
DOI: 10.1039/D6MA00167J

Interfacial Interaction-Driven Comparison of Phase-Transition and Phase-Change Nanocomposite Hydrogels for Multifunctional Energy-Therapeutic Platforms

Shikha Awasthi[1,2*]

[1]Jagran School of Pharmacy, Jagran Lakecity University, Bhopal - 462044, Madhya Pradesh, India

[2]Sunway Center for Electrochemical Energy Storage and Sustainable Technology, Faculty of Engineering and Technology, Sunway University, No. 5 Jalan Universiti, Bandar Sunway, 47500 Petaling Jaya, Selangor Darul Ehsan, Malaysia

Email: awas.shikha2212@gmail.com and shikhaa@sunway.edu.my

Abstract

View Article Online
DOI: 10.1039/D6MA00167J

A new class of soft materials known as phase-transition and phase-change hydrogels (PTHs and PCHs) may undergo reversible changes in volume or structure in response to external stimuli, including heat, light, pH, or electromagnetic fields. For sophisticated healthcare, energy, and smart device applications, their chemical adaptability and adjustable mechanical properties have attracted significant attention. The basic ideas that determine hydrogel responsiveness and multifunctionality are introduced at the outset of this review, which is then followed by classifications and design guidelines. After that, the review delves into synthesis techniques for customizing PTHs and PCHs, with a focus on using nanoparticles as reinforcing agents to improve functional variety, robustness, and responsiveness. To demonstrate how nanoscale interactions regulate mechanical reinforcement, energy dissipation, and phase-change kinetics, the molecular processes and structure-property correlations driving phase transitions are examined. Energy technologies (thermal management, storage, wearable electronics), smart devices (actuators, sensors, bioelectronics), and therapeutic systems (drug delivery, tissue engineering, photothermal treatment) are among the recent advancements in multifunctional applications that are reviewed. Finally, the study addresses key challenges such as biocompatibility, long-term stability, and scalability of production, and offers future perspectives for flexible, sustainable, and multipurpose soft materials. Design, synthesis, and application knowledge converge to make nanoparticulate PTHs and PCHs flexible platforms at the intersection of nanotechnology, materials science, and biomedical engineering.

Keywords: Hydrogels; Phase change; Sustainable livelihoods; Energy; Biomedical

View Article Online
DOI: 10.1039/D6MA00167J

1. Introduction

A special type of soft material called hydrogels is made up of networks of three-dimensional crosslinked polymers that can hold a lot of water while yet being structurally sound [1–4]. They are essential in a variety of fields, from biomedicine to energy technologies, due to their high water content, adjustable mechanical characteristics, and likeness to biological tissues [5,6]. Recently, there has been growing interest in specialized types of hydrogels, such as phase-transition hydrogels (PTHs) and phase-change hydrogels (PCHs), which can reversibly change their volume, structure, or conformation in response to external stimuli [7–9]. Phase-changeable hydrogels are a captivating class of soft materials that hold great scientific and practical interest. PTHs can change their volume, structure, or physical properties very rapidly in response to changes in temperature, pH, ionic strength, light, electric and magnetic fields. These transitions are typically due to interactions between the polymer and the solvent and the construction of the network, and can be swelling and deswelling events that can occur dynamically and reversibly [7]. Poly(N-isopropylacrylamide)-based hydrogels are a famous example, which have a lower critical solution temperature (LCST) reaction and quickly release water over a certain temperature [10]. Similarly, pH-sensitive hydrogels are based on the presence of ionizable groups that change the charge density and osmotic pressure[11]. Photo-responsive systems, on the other hand, use chromophores to cause conformational changes when exposed to light [12]. These changes, caused by stimuli, provide PTHs with special abilities, such as regulated release, the capacity to change form, and the ability to respond to complex biological or environmental signals. Due to their versatility, PTHs are interesting materials for biomedical applications, drug delivery, tissue scaffolding, biosensing, and soft robotics. Their use is usually limited by their fragility, their slow response and their durability when repeated use, which demonstrates the need for more robust structures and more functional integration.

Phase-change hydrogels (PCHs) are based on the concept of PTHs and incorporate additional phase transitions between other material states, such as gel-sol or solid-liquid[13,14]. While most of the time, the volume or shape of the material changes reversibly, PCHs are intended for larger phase changes that allow you to alter the properties of the material. These systems are beneficial for injectable hydrogels, thermal energy storage, and shape-programmable devices, where solidification or liquefaction may be reversed to allow for controlled deployment and recovery [14–16]. PCHs become more mechanically strong and can

View Article Online
DOI: 10.1039/D6MA00167J

do more things, including photothermal heating, magneto-responsiveness, or electrical conductivity, by adding nanoparticle reinforcements. This makes them more useful in more circumstances. So, PCHs are a natural step up from PTHs. They integrate reversible transitions and multi-functional reinforcement to create flexible platforms for the latest health, energy and smart device technologies. When taken as a whole, these nanoparticle-integrated systems represent an expanding family of smart or intelligent materials that are intended to react to their surroundings flexibly.

Because of their adaptability, PTHs and PCHs are especially appealing for therapeutic uses such as photothermal cancer treatment, less invasive scaffolds for tissue engineering, and on-demand drug delivery [15,17]. Their innate capacity to store and release energy during phase transitions has also sparked applications in wearable electronics, energy storage, and temperature control. Moreover, multipurpose platforms for intelligent devices are also gaining interest, as they are being integrated into flexible sensors, actuators, and soft robotics. Nevertheless, despite this potential, the actual translation of these resources is constrained by a number of inherent restrictions. The traditional hydrogels are generally not multifunctional, have long reaction times, low mechanical strength and poor fatigue resistance [18]. These constraints limit their effectiveness and long-term stability, especially in challenging biomedical or energy applications.

Researchers are increasingly using nanoparticle reinforcement techniques to get over these obstacles [19,20]. When inorganic, organic or hybrid nanostructures are embedded in hydrogel matrices, significant progress has been achieved in the structural and functional aspects. Nanoparticles act as secondary crosslinking sites mechanically to dissipate stress and enhance elasticity, toughness and durability [21,22]. In terms of functionality, they provide additional levels of responsiveness: metallic nanoparticles allow photothermal conversion, carbon-based nanomaterials add electrical conductivity, catalytic nanostructures speed up reaction kinetics, and magnetic nanoparticles give magneto-sensitivity [23]. The dynamic hydrogel networks and nanoscale reinforcements work together to create hybrid systems with both phase transition properties and enhanced and multifunctional properties. The scientific significance of nanoparticle-reinforced PTHs and PCHs is due to the basic molecular-level interactions between polymer networks and nanostructures. Phase-change kinetics, energy dissipation routes, and network rearrangement are all determined by these interactions. For instance, hydrogen bonding, ionic contacts or supramolecular host–guest complexes between nanoparticles and polymer chains can provide reversible but strong bonds that enable repeated phase changes. In addition, the large surface area and tunable surface chemistry of nanoparticles enable precise control of the hydrogel microstructure, swelling properties, and stimuli-responsiveness. Compared to their unaltered counterparts, the resultant nanocomposites show phase transitions with more fidelity, enhanced reversibility, and wider stimulus sensitivity.

A paradigm change in the design of multifunctional soft matter is represented by nanoparticle-reinforced PTHs and PCHs in the larger context of materials science and nanotechnology. They are excellent examples of hierarchical structuring at the molecular, nanoscale and macroscale levels that can be applied to the design of flexible systems that are technically useful and function in a biologically inspired manner. Engineering-wise, these materials offer opportunities for application-specific, scalable platforms that can address challenges in environmental sustainability, soft robotics, precision medicine and renewable energy. A comprehensive assessment of nanoparticle-reinforced PTHs and PCHs is warranted, given the speed at which research is progressing. The purpose of this article is to provide such an overview by (i) describing the categories and design tenets that support hydrogel systems that undergo phase transitions and changes. (ii) Providing an overview of synthetic methods for creating pure and nanoparticle-reinforced versions. (iii) Talking about the structure-property correlations and processes that control their responsive behaviour. (iv) Demonstrating multi-purpose applications of energy technologies, smart gadgets and therapeutic platforms. (v) Identifying barriers, limitations and future prospects for bringing these technologies to fruition. Although a few studies have encapsulated the progress of hydrogel research, most of the existing literature is dedicated to the generic stimuli-responsive hydrogels or specific application fields such as biomedical systems, thermal management, or soft robotics [24–26]. No special and systematic research is available that concentrates on the unique and rapidly emerging class of nanoparticle-reinforced phase-transition hydrogels (PTHs) and phase-change hydrogels (PCHs). This article fills that gap by giving a critical and organized discussion that brings together molecular design methodologies, synthesis pathways, structure-property connections, and multifunctional applications.

Based on this perspective, a systematic comparison of PTHs and PCHs is required to differentiate their operating principles and performance characteristics. Although both classes are considered as phase-responsive soft materials, they exhibit very different underlying processes, response dynamics, thermodynamic behaviour, mechanical properties and energy storage capacities. To give a clear and organized understanding, a comparison summary (**Table 1**) is provided, highlighting important performance indicators like response rate, mechanical characteristics, thermodynamic parameters, and cycle stability. This framework not only

View Article Online
DOI: 10.1039/D6MA00167J

clarifies the basic differences between PTHs and PCHs but also facilitates rational design and selection of these systems for various applications in energy storage, biomedical engineering, and flexible soft material platforms.

Table 1: Comparative analysis of PTHs vs PCHs.

Parameter	**Phase-Transition Hydrogels (PTHs)**	**Phase-Change Hydrogels (PCHs)**	**Key Implication**	**Reference**
Driving Mechanism	Polymer–solvent interactions (LCST/UCST)	Solid–liquid phase transition of PCM	Entropy-driven vs enthalpy-driven	[27]
Response Rate	Fast (seconds to minutes)	Moderate to slow (minutes to hours)	PTHs suitable for rapid actuation	[28]
Reversibility	Highly reversible swelling-deswelling	Reversible melting–crystallisation	Both cyclic but different mechanisms	[27]
Mechanical Properties	Soft, tunable, sometimes weak	Reinforced, often more stable due to PCM confinement	PCHs have better structural stability	[29]
Thermodynamic Basis	Conformational entropy change	Latent heat storage/release	Distinct energy storage pathways	[27]
Energy Storage Capacity	Low to moderate	High (latent heat)	PCHs are ideal for thermal storage	[9]

View Article Online
DOI: 10.1039/D6MA00167J

Thermal Conductivity	Generally low	Often enhanced using fillers	PCHs require conductivity tuning	[30]
Cycling Stability	Good, but may degrade due to fatigue	High with proper encapsulation	PCHs suitable for long-term use	[30]
Volume Change	Significant (swelling/deswelling)	Minimal to moderate	PTHs are useful for actuation	[31]
Diffusion/Transport	High (dependent on mesh size)	Restricted by PCM domains	Impacts, release, and heat transfer	[31]
Stimuli Sensitivity	High (temperature, pH, light, etc.)	Primarily temperature-driven	PTHs more multifunctional	[27]
Typical Applications	Drug delivery, actuators, sensors	Thermal energy storage, thermal regulation	Clear application distinction	[9]

The unique feature of this review is its narrow emphasis, which separates nanoparticle-engineered phase-transition hydrogels (PTHs) and phase-change hydrogels (PCHs) from the more general category of responsive hydrogels. This study emphasizes the differences and similarities between PTHs and PCHs, and their common adaptive nature, compared to previous studies that tend to lump stimuli-responsive hydrogels together. The mechanistic understanding it offers is a significant innovation, with special focus on the molecular interactions between nanoparticles and hydrogel networks and how these interactions affect reinforcement, phase-change dynamics, and multifunctionality. While there are many papers that refer to nanoparticles as reinforcements, few explain in detail how the microscopic interactions relate

View Article Online
DOI: 10.1039/D6MA00167J

to the macroscopic behavior, as this study does. Moreover, the majority of the surveys available focus on one of the three areas: drug delivery, thermal storage, or tissue engineering. This article, on the other hand, advocates for a cross-domain integration, linking biomedical, energy, and smart device applications together and emphasizing the ability of these applications to work together, not separately. Importantly, the paper provides a critical perspective on the limitations that remain to be addressed for translation into practical applications, such as slow reaction rates, fatigue resistance, long-term stability, and the challenges of scaling up production. In the future, it describes methods to get around these problems and shows the path to multipurpose, sustainable, and adaptable hydrogel systems that are prepared for real-world use. The categorization and range of applications for PTHs and PCHs are shown in **Figure 1**. PTHs may be used in wearable electronics, biomedical devices, soft robotics, optical systems, and other stimulus-sensitive fields. They are classified as thermoresponsive, pH-responsive, light-responsive, and other varieties. PCHs, on the other hand, offer larger applications in energy and thermal management, biological systems, and smart devices by including sol–gel, solid–liquid, and other reversible phase transitions. The complementary adaptive roles of PTHs and PCHs for next-generation multifunctional platforms are highlighted by these schematic routes taken together.

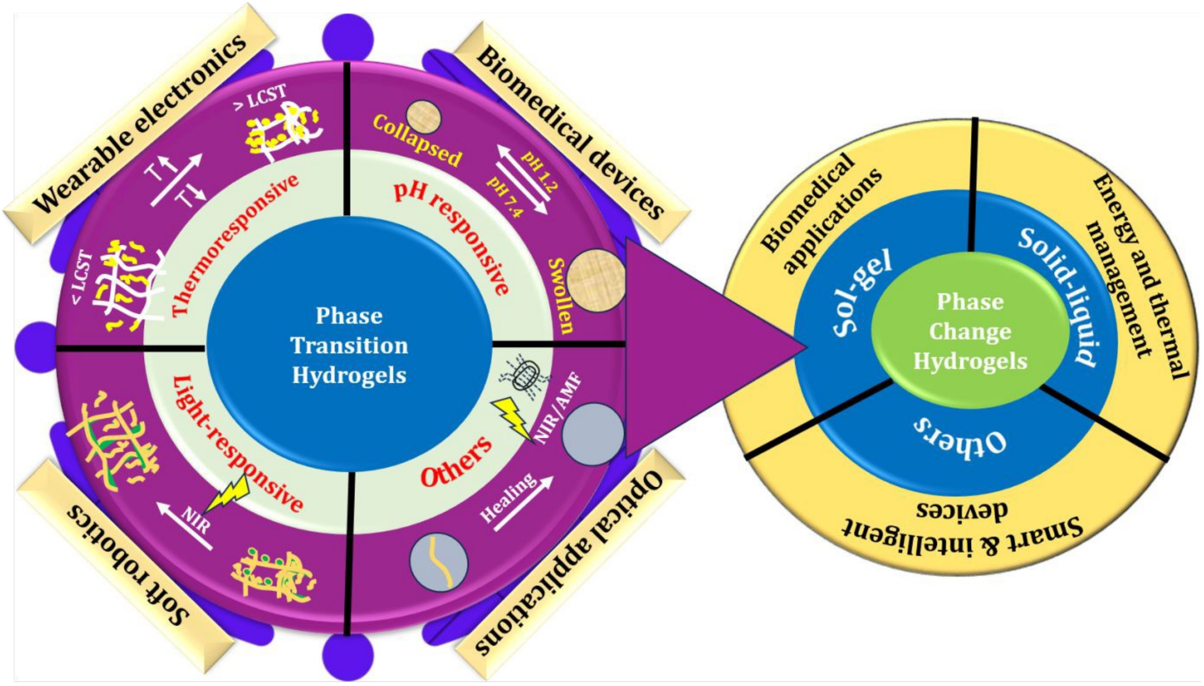


Figure 1: Phase-transition hydrogels and phase-change hydrogels are shown schematically, with an emphasis on their kinds and typical uses in the energy, biomedical, and smart device

View Article Online
DOI: 10.1039/D6MA00167J

sectors. (T: temperature, NIR: Near Infrared, AMF: Alternating Magnetic Field and LCST: Lower Critical Solution Temperature). Original illustration.

2. Classifications of PTHs and PCHs

Phase-transition hydrogels are among the most dynamic types of soft materials; they respond to environmental stimuli by undergoing reversible and often sudden physical changes. Volume collapse, swelling, solvation-desolvation, or shape transformation are common manifestations of these changes, which are regulated by the balance of polymer–polymer, polymer-solvent, and polymer-stimulus interactions. Phase change hydrogels, on the other hand, exhibit reversible enthalpy-driven phase transitions, such as sol–gel, solid–liquid, or amorphous–crystalline transitions, and can store energy, modulate volume, or modulate mechanical properties. PCHs are grouped according to the characteristics of the phase transition itself, while PTHs are grouped according to the kind of stimulus that causes the reaction. To attain desired kinetics, stability, and multifunctionality, all crucial for applications in biomedical devices, soft robotics, thermal management, and environmental systems, design strategies in both situations center on adjusting polymer chemistry, crosslinking architecture, network interactions, and reinforcement.

2.1 Types of phase transition hydrogels

2.1.1 Thermoresponsive hydrogels

Because of their distinct lower or upper critical solution temperature (LCST/UCST) behaviour, thermoresponsive PTHs are the most extensively researched subclass[32,33]. Polymers like poly(N-isopropylacrylamide) (PNIPAm) have an LCST of around 32 °C, which is close to body temperature, they are especially appealing for use in biomedical applications [34,35]. Hydrophobic interactions between polymer backbones drive chain breakdown and water ejection above the LCST, but polymer–water hydrogen bonding predominates below it, maintaining the hydrogel's swelling. Although less frequent, UCST-type structures behave oppositely and collapse at lower temperatures. The hydrophobic comonomer ratio, crosslinking density, polymer composition, and the presence of reinforcing nanoparticles all have a significant impact on the transition's hysteresis and sharpness [36,37]. Thermoresponsive hydrogels have been used in tissue engineering, biosensors, controlled drug administration, and actuators, where temperature acts as a simple, non-invasive trigger.

2.1.2 pH-responsive hydrogels

Another basic subclass is pH-responsive PTHs, which rely on the protonation or deprotonation of ionizable functional groups in the polymer network. Because of electrostatic repulsion between deprotonated groups, hydrogels with acidic groups like carboxylic acid (-COOH) swell at higher pH values, while hydrogels with basic groups, like amines ($-NH_2$), swell in acidic environments [38,39]. The degree of swelling, transition threshold and reversibility are influenced by the kind and density of ionizable moieties, buffer ionic strength and ambient factors. These hydrogels are highly relevant in biological applications where local pH changes, such as in inflammatory tissues, the gastrointestinal system, and tumor microenvironments, could serve as natural triggers [40,41]. Surface functions of nanoparticles can be used to adjust pH sensitivity further, and mechanical stability and responsiveness can be improved by introducing them into the material.

2.1.3 Light-responsive

Light-responsive PTHs take advantage of the reversible structural changes that photoactive moieties that are connected as pendant groups or incorporated in the polymer backbone may undergo when exposed to radiation. Common tactics include the trans-cis isomerization of azobenzene-based chromophores, the hydrophobic/hydrophilic state flipping of spiropyran units, or the formation and breaking of crosslinks by coumarin groups when exposed to UV/visible light [42]. The hydrogels are attractive for spatiotemporal control of volume transitions, for precise drug administration, and for cell culture platforms and optomechanical actuators. By turning light into heat or starting photochemical reactions, light-responsive nanoparticles like graphene oxide, gold nanorods, or upconversion nanoparticles can increase efficiency even more and expand the operating wavelength window to include biologically significant near-infrared regions[43–45].

2.1.4 Hydrogels that are electro-and magnetoresponsive

By adding conductive polymers, ionic groups, or magnetic nanoparticles, electro- and magnetoresponsive PTHs are made to react to electric or magnetic fields [46,47]. Swelling or deswelling can occur in an electroresponsive system due to electrostatic interactions, pH changes resulting from electrolysis, or electrophoretic ion migration. To improve charge transfer and actuation efficiency, conducting polymers like polypyrrole and polyaniline are often used [48,49]. Superparamagnetic nanoparticles are often included in magnetoresponsive hydrogels, allowing for quick, reversible, and distant changes under alternating magnetic fields. The high accuracy and non-invasive response tuning capabilities of these technologies are

View Article Online
DOI: 10.1039/D6MA00167J

particularly interesting for targeted therapeutics, wireless soft actuators, and microfluidic devices.

2.1.5 Multi-stimuli responsive hydrogels

Many of the complex PTHs have been developed to respond to multiple stimuli, and these are called multi-stimuli responsive hydrogels [50]. The magnetic nanoparticles can be used to achieve triple responsiveness (thermal, pH, and magnetic), while PNIPAm-based hydrogels with acrylic acid can be made both thermally and pH sensitive [51,52]. These designs open up possibilities for complicated biomedical devices, logic-gated medication release, and adaptive robotics by enabling synergistic control of swelling kinetics and functionality. Since numerous signals coexist and affect material behaviour in biological contexts, multi-stimuli systems are at the forefront of PTH design.

The main categories of nanocomposite PTHs are shown in **Table 2**, which also summarizes the operation of thermo, pH, light, electro-/magneto, and multi-responsive systems, as well as their main applications and the main obstacles preventing their widespread adoption.

Table 2: Types of nanocomposites PTHs grouped according to their primary limitations, application fields, and reaction mechanisms.

Response type	Response mechanism	Application domains	Key limitations	Reference
Thermo-responsive nanocomposite hydrogels	Polymers with lower or upper critical solution temperature (LCST/UCST) undergo hydrophilic–hydrophobic switching near transition points, driving changes in volume, porosity,	Targeted drug release, biosensing, 3D bioprinting	Transition temperature shifts with ionic strength, cosolvents, and additives, complicating precise control. Loss of performance at elevated temperatures due to dehydration or	[53]

View Article Online
DOI: 10.1039/D6MA00167J

	and molecular diffusion.		shrinkage, leading to uncontrolled release in biomedical use.	
pH nanocomposite hydrogels	Ionizable groups such as –COOH or –NH_2 in polymer matrices alter charge state with pH, regulating swelling, solubility, and diffusion properties.	Controlled delivery systems, adaptive smart materials	Limited pH window tied to pKa restricts function in variable biological environments. Irreversible structural damage possible under prolonged extreme pH. Difficulties in reproducible large-scale synthesis.	[40,54]
Light-responsive nanocomposite hydrogels	Incorporation of photoactive moieties or nanofillers enables conformational changes, bond cleavage, or photocatalytic activity under specific light wavelengths.	Biomedical phototherapies, environmental cleanup, energy harvesting and actuation	Poor penetration of UV and visible light into deep tissues. Responsiveness depends on light intensity, wavelength, and duration. Reduced stability after repeated irradiation cycles.	[55]
Electro- and magneto-responsive	Conductive or magnetic fillers facilitate charge migration or	Wearable and flexible sensors, bioelectronic interfaces,	High ionic/electronic resistance reduces sensitivity and	[46,56]

View Article Online
DOI: 10.1039/D6MA00167J

nanocomposite hydrogels	dipole alignment under external electrical or magnetic fields, resulting in deformation, swelling, or conductivity changes.	remote-controlled actuators	speed. Maintaining lubrication and performance in liquid environments is challenging. Magnetic fillers may cause aggregation or biocompatibility concerns.	
Multi-responsive nanocomposite hydrogels	Integration of diverse functional groups or nanofillers enables response to multiple stimuli (e.g., pH, temperature, light, magnetic fields) within a single network.	Smart drug delivery with spatiotemporal control, adaptive soft robotics, multifunctional biosensors	Complex design makes reproducible synthesis and scale-up difficult. Potential for conflicting responses (e.g., thermal vs. pH). Balancing responsiveness with mechanical robustness remains a challenge.	[57]

2.2 Classifications of phase change hydrogels

2.2.1 Sol-gel phase change hydrogels

Phase shift of sol-gel is usually caused by temperature, pH, or ionic strength; hydrogels may reversibly change from a low-viscosity sol to a high-viscosity gel state. Covalent crosslinks provide structural stability, whereas reversible physical crosslinks, including hydrogen bonds, hydrophobic interactions, and electrostatic forces, facilitate the change [58]. Commonly utilized polymers include gelatin, agarose, chitosan, PNIPAm, and polyvinyl alcohol (PVA); their concentration, molecular weight, and crosslinking density determine the mechanical characteristics, gelation temperature, and kinetics [58–60]. In biomedical applications, sol-gel PCHs are exceedingly adaptable, allowing for regulated drug administration, in situ tissue creation, and injectable scaffolds [61–63]. While the gel state offers mechanical support and regulates diffusion for prolonged release, the sol state makes it simple to load or distribute medicines. Nanofibers or nanoparticles may be included to improve responsiveness, conductivity, or mechanical robustness [20]. Sol-gel PCHs are being investigated for industrial and environmental uses outside of biomedicine, including absorbents, smart filters, and switchable encapsulating systems. Controlling gelation kinetics, preserving mechanical stability, and guaranteeing reversible cycling free of hysteresis are important design issues.

2.2.2 Solid-liquid PCHs

Latent heat storage and temperature control are made possible by the reversible melting and solidification of crystalline domains within the polymer network that characterizes solid–liquid PCHs [9]. In order to preserve structural integrity during multiple phase transitions, crosslinking is often used in polymers, including polyethene glycol (PEG), PVA, poloxamers, and polyethene oxide (PEO) derivatives [64]. Mechanical strength, homogeneous crystallization, and heat conductivity may all be enhanced by the addition of nanoparticles such as graphene, silica, or metal oxides. When precise temperature control or energy storage is needed, solid-liquid PCHs are used in thermal management, wearable technology, smart fabrics, and biomedical scaffolds [65]. Thermal expansion and contraction may alter release kinetics in medication delivery. Because the hydrogel must tolerate volumetric variations during melting and solidification without breaking or leaking, mechanical resilience is essential. To combine utility and durability, design concerns include maximizing the transition temperature, latent heat, cycle stability, and the coupling of permanent covalent networks with reversible physical crosslinks [66].

2.2.3 Amorphous-crystalline or amorphous-amorphous PCHs

Amorphous-crystalline and amorphous–amorphous PCHs affect mechanical properties, mobility of water, and shape memory effect by ordering or disordering polymer chains without bulk melting [67,68]. Such transitions can be relatively small but can be very functional, affecting diffusion, swelling and mechanical stiffness, and can be induced by temperature, solvent interactions, or external stress. These hydrogels are extremely useful in shape-memory scaffolds, bioelectronics and controlled drug delivery due to the reversible microstructural rearrangements that control their performance [69]. The use of reinforcement with nanoparticles can enhance the mechanical stability, promote uniform ordering, and incorporate additional properties such as photothermal responsiveness or electrical conductivity. The degree of crystallinity, reversibility over multiple cycles, and mechanical strength and responsiveness are important design considerations.

2.2.4 Other types of PCHs

In addition to the sol-gel, solid-liquid and amorphous-crystalline or amorphous–amorphous phase transition hydrogel types, other types of PCHs include multi- or hybrid systems, and organic and inorganic variations [70,71]. Enhanced responsiveness and adjustable thermal, mechanical, and functional qualities are made possible by multi- or hybrid PCHs, which integrate two or more functional components or phase change mechanisms, such as sol-gel with solid–liquid transitions, into a single network. In order to maximize transition kinetics, mechanical resilience, and multifunctional performance, these hydrogels often combine chemical and physical crosslinks, hierarchical mesh topologies, and nanoparticle reinforcement. Classified according to their composition, organic and inorganic PCHs have unique structural and functional properties that provide adaptability for certain applications. PCHs may be grouped according to their composition as well as how receptive they are to outside stimuli. Thermoresponsive hydrogels undergo reversible gel–sol transitions upon changes in temperature; photoresponsive systems undergo changes when exposed to light, using photoreactive components; electroresponsive hydrogels undergo changes when exposed to an electric field, using electroactive polymers or shape memory materials; and pH-responsive hydrogels change their swelling, gelation, or structural configuration when the pH of the solution changes. PCHs are further divided into physical and chemical networks based on the type of crosslinking. While physically crosslinked hydrogels provide dynamic and reversible interactions, chemically crosslinked hydrogels have better mechanical stability, durability, and consistent gel states across a wider temperature range. The source material is also important. Synthetic PCHs can be prepared by polymerization, copolymerization and crosslinking of chemical monomers, and can be precisely controlled to achieve the desired network structure, mechanical properties and phase change behavior, whereas natural PCHs prepared from plant polysaccharides, algal polymers or animal tissues have excellent biocompatibility, biodegradability and minimal toxicity, suitable for biomedical,

View Article Online
DOI: 10.1039/D6MA00167J

pharmaceutical and food-related applications. Furthermore, depending on the material composition, PCHs may be categorized as composite hydrogels or polymeric hydrogels. While composite PCHs combine several materials, such as organic phase change materials, inorganic nanoparticles, or polymer nanocomposites, to improve functionality and performance, polymeric hydrogels are made entirely of polymer networks and water and provide good tunability and biocompatibility. These many PCH variants are used in multifunctional energy systems, soft robotics, actuators, and flexible biomedical devices. Nevertheless, there are still issues with synchronizing several phase transitions, reducing hysteresis, guaranteeing cyclic stability, and striking a balance between mechanical performance and energy storage efficiency or biocompatibility, especially in hybrid and multi-phase systems intended for intricate responsive applications.

Phase Change Hydrogels' (PCHs') varied behaviour and uses, from sol-gel injectability to solid-liquid heat storage and amorphous-crystalline shape-memory effects, emphasize how important it is to optimize their performance via logical design. To ensure that each type of PCH can undergo reversible and reliable phase transitions, exhibit optimal mechanical properties and be capable of performing a specific function for a particular application, careful control of polymer chemistry, crosslinking, network design, and reinforcement is required. The design approach should also incorporate features such as latent heat capacity, crystallization kinetics and cycle stability to ensure long term operation and responsiveness. These factors form the foundation for developing PCHs that are structurally designed to meet their functional requirements in the real world and are applicable to biological, environmental and energy systems. Like the design principles developed for PTHs, the following section outlines a methodical review of PCH design principles, but with an emphasis on phase-change-specific design principles.

3. Design principles of PTHs and PCHs

The network architecture, crosslinking, reinforcing techniques, and polymer chemistry are all crucial for the design of phase-change hydrogels or Phase-transition hydrogels. Unlike PCHs, which undergo enthalpy-driven phase changes such as sol-gel, solid-liquid and amorphous-crystalline transitions, PTHs respond to external stimuli such as temperature, pH, light, and electric/magnetic fields. Both classes aim to achieve reversible, rapid and secure transitions, while maintaining suitable mechanical and functional properties, despite these differences. Here is a summary of important design elements, highlighting parallels and differences as appropriate.

View Article Online
DOI: 10.1039/D6MA00167J

3.1 Functional groups and the polymer backbone

The responsiveness of the PTHs depends on the selection of the polymer backbone: covalently linked light-sensitive groups can be isomerized by light; hydrophilic-hydrophobic balance can be controlled by heat; acidic or basic pendant groups can be used to control the pH sensitivity[31,72]. Enthalpy changes, crystallinity and phase transition temperature are the properties governed by the backbone in PCHs [9]. The sol-gel transitions can be achieved by the use of PNIPAm, gelatin or agarose, whereas polymers like PEG or PVA create crystalline domains for solid–liquid transitions. Besides the adjustment of mechanical strength, swelling and interaction with the nanoparticles, copolymerization can also be used to introduce different functional groups, so that hybrid or multi-stimuli responsiveness can be achieved in both PTHs and PCHs [13].

3.2 The approach of crosslinking

Both PTHs and PCHs make a compromise between dynamic reversibility and mechanical stability through crosslinking. Reversible physical crosslinks (hydrogen bonding, host-guest interaction, and ionic association) offer flexibility, whereas covalent crosslinks in PTHs help to retain the structure [73,74]. PCHs require covalent scaffolds to support crystalline or molten domains, as well as physical crosslinks for reversible phase shifts [75,76]. The double-network and semi-IPN designs are used in both classes, providing responsive behaviour with toughness, which gives good durability and allows for multiple cycles of melting/solidification (PCHs) or swelling/deswelling (PTHs).

3.3 Mesh size and network density

The number of meshes and the density of the network.Number of meshes and network density. The mesh size and network density are important structural properties for both PTHs and PCHs that determine their physicochemical behaviour and phase responsiveness. The distance between crosslinks in a polymer network is called the mesh size (ξ), and it directly affects transport properties, swelling rates and diffusion of solvents [77,78]. On the other hand, network density reflects the degree of crosslinking and determines the stiffness and structure of the hydrogel matrix. Thermoresponsive swelling-deswelling behaviour of PTHs is controlled by mesh size. A higher mesh size allows for more water uptake and faster diffusion, leading to noticeable volume phase shifts, while a more strongly crosslinked network will have less chain mobility and will be less responsive. Modifications in the network density, in turn,

View Article Online
DOI: 10.1039/D6MA00167J

affect the LCST and the sharpness of the transition, as well as the equilibrium between the interactions between the polymer-polymer and polymer-solvent [79,80]. Hence, it is necessary to fine-tune the mesh size to control actuation speed, permeability and stimulus sensitivity.

The mesh size and network density of PCHs have a significant effect on the encapsulation, confinement and crystallzation behaviour of phase change materials (PCMs). A close crosslinked network with a small mesh size can be used to effectively prevent PCM leakage and improve the form stability, in addition to the effect on nucleation and crystal growth in phase changes. Higher mesh size, however, can provide greater latent heat storage capacity, but can compromise structural integrity under repeated thermal cycling. Therefore, the interaction between the mesh size and the interfacial confinement is a key factor in the control of the thermal conductivity, the efficiency of the phase transition and the durability of PCH systems [81]. Overall, the ability to control mesh size and network density provides a fundamental design strategy to tailor mass transfer, mechanical properties, and phase behaviour in hydrogel systems for optimal performance in energy storage and responsive applications.

3.4 Reinforcement by nanoparticles

Nanoparticle inclusion has changed PTHs, while it has historically been thought of in relation to PCHs. Multipurpose crosslinkers, stress relievers, and stimulus transducers are all functions of nanoparticles. For example, graphene improves conductivity and durability, iron oxide nanoparticles enable magneto-responsiveness, and gold nanorods facilitate photothermal heating. Strong polymer–nanoparticle interactions are made possible by their vast surface area and adjustable surface chemistry, which enhances transition fidelity and functional diversity [82,83]. By functioning as crosslinking agents, stress relievers, stimulus transducers, or nucleation sites, nanoparticles improve the performance of PCH and PTH [84,85]. For PTHs, photothermal effects are made possible by gold nanorods, magneto-responsiveness is made possible by iron oxide, and conductivity is enhanced by graphene [86,87]. Nanoparticles in PCHs promote mechanical stability, enhance thermal conductivity, and enable uniform crystallization during repeated solid-liquid or sol-gel transitions. In all hydrogel forms, effective energy transfer and functional integration are guaranteed by strong polymer–nanoparticle interactions.

3.5 Transition kinetics and reversibility

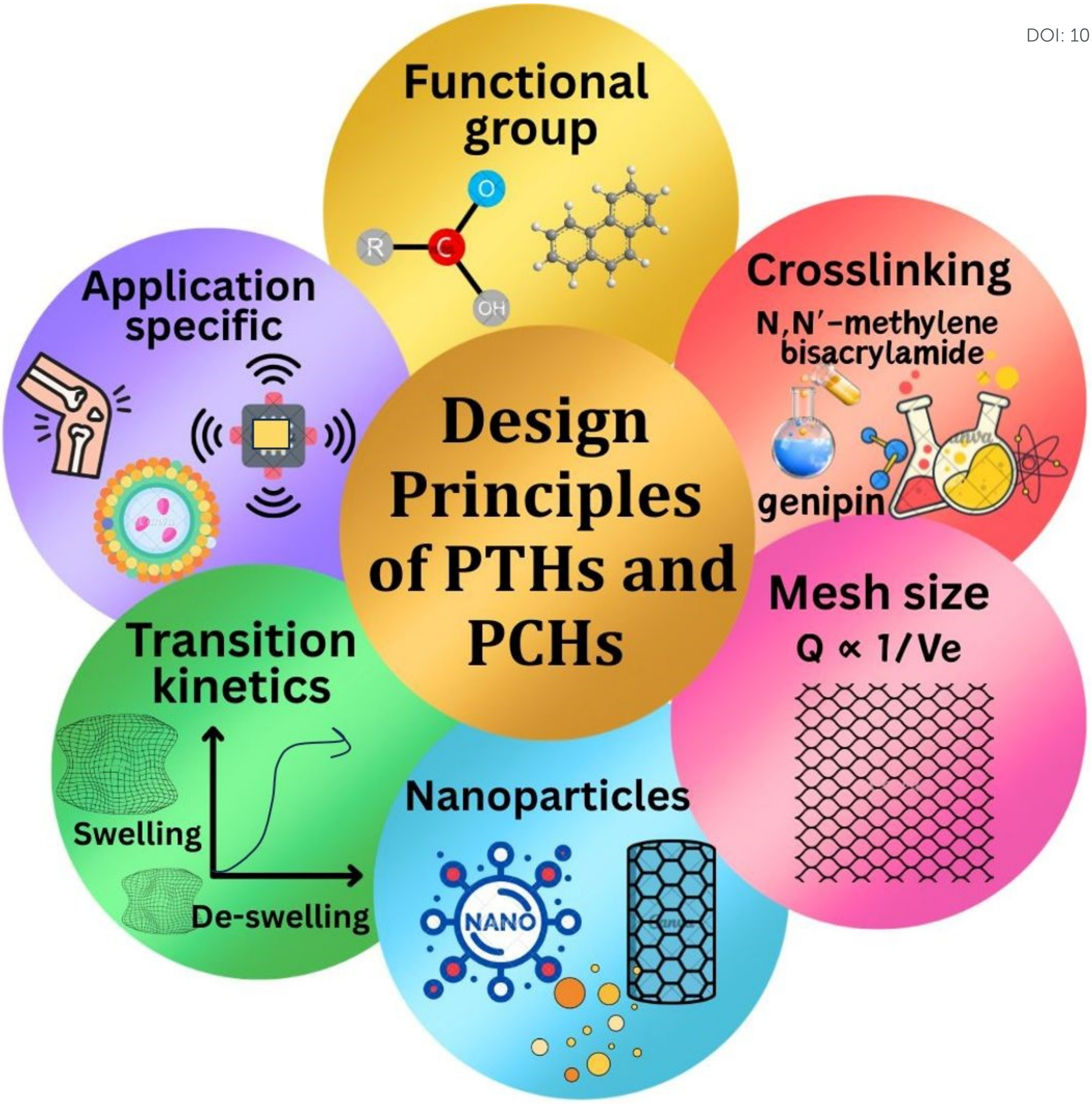
View Article Online
DOI: 10.1039/D6MA00167J

Both hydrogel classes need quick and reversible changes. Melting/solidification, sol–gel, or amorphous ordering must be constant throughout many cycles in PCHs, while swelling/deswelling cycles in PTHs must be reproducible without hysteresis. Predictable kinetics, dependability, and long-term functioning may be preserved by establishing nucleation sites or hierarchical domains, decreasing network heterogeneity, and optimizing polymer mobility [88].

3.6 Application-specific limitations and biocompatibility

PCHs and PTHs both need customization that is focused on the application. While PCHs for biomedical usage must have phase transitions close to physiological temperatures and preserve latent heat storage without sacrificing biocompatibility, biomedical PTHs must be cytocompatible, non-toxic, and often biodegradable. PCHs may provide latent heat capacity, conductivity, or cycle stability precedence over biodegradability in energy or electrical applications. To create multifunctional hydrogels that are sensitive, long-lasting, and customized for their particular uses, designers combine polymer chemistry, crosslinking, network architecture, nanoparticle reinforcement, and phase-transition kinetics [89–91]. **Figure 2** summarizes the general design concepts of PTHs and PCHs, including the importance of polymer backbone selection, functional groups, crosslinking strategy, mesh size and network density, nanoparticle reinforcement, transition kinetics, and application-specific tailoring. These factors synergistically improve the responsiveness, mechanical strength, reversibility and multifunctionality of the hydrogel. For example, crosslinking provides a compromise between rigidity and flexibility, the chemistry of the polymer backbone determines its sensitivity to stimuli or phase-change properties, and nanoparticles can act as stress relievers, stimulus transducers, or nucleation sites for crystallization. Optimized transition kinetics provide fast and repeatable response, and mesh architecture controls swelling, diffusion and latent heat transfer. Application-specific tailoring incorporates requirements like biocompatibility, biodegradability, thermal regulation or conductivity, depending on the intended use. These design principles collectively offer a blueprint for the design of next-generation hydrogels that can reliably function in biomedical, environmental, energy, and soft

View Article Online
DOI: 10.1039/D6MA00167J



robotic applications, both in response to external stimuli (PTHs) and in response to enthalpy-driven phase transitions (PCHs).

Figure 2: Design concepts of phase-transition (PTH) and phase-change (PCH) hydrogels. Responsiveness and phase behaviour are controlled by functional groups and the polymer backbone, such as photoisomerization, hydrophilic–hydrophobic balance, ionization, or crystallization. Crosslinking (N,N′-methylenebisacrylamide, genipin or other agents) is a combination of reversible interactions for flexibility and covalent bonds for stability, enhanced by double-network and semi-IPN architectures. Nanoparticles (such as carbon nanotubes, graphene, carbon dots, metal oxides) serve as crosslinkers, stress relievers, stimulus transducers and nucleation sites, providing environmental responsiveness and phase-change fidelity. Mechanical strength, swelling kinetics, diffusion and latent heat transfer (Q is the swelling ratio and Ve is the effective crosslink density) are controlled by mesh size and network porosity.

View Article Online
DOI: 10.1039/D6MA00167J

Optimized transition kinetics and reversibility enable reliable, repeatable cycles for swelling/deswelling (PTHs) or melting/solidification (PCHs). Application-specific tailoring ensures biocompatibility, biodegradability, thermal regulation, or conductivity, depending on the target use, including bone tissue engineering, drug delivery, soft robotics, sensors, and energy systems.

4. Synthesis strategies and mechanisms of PTHs and PCHs

4.1 Fabrication routes and functional mechanisms of phase-transition hydrogels

4.1.1 Preparation approaches for thermoresponsive PTHs

Phase transition hydrogels use a variety of gels and methods and react to a range of external stimuli, including heat, pH, light, magnetic, ultrasonic, and electric fields. Their behaviors vary according to the stimulus. Designing the molecular backbone and response groups, controlling the necessary conditions for phase transition, and controlling the phase transition rate are all part of the process of creating phase transition hydrogels. Gel molecular chains undergo reversible breakdown and reconfiguration in response to external stimuli via the use of reversible and dynamic cross-linking processes. The kind of response, the key circumstances for phase change, and the pace of phase transition may all be controlled by varying the system's hydrophilic/hydrophobic monomer ratio, the kind of responsive groups, and the density and structure of the crosslinked network. The several operating principles that control the behaviour of phase transition hydrogels are thoroughly discussed in this section.

Although thermally responsive polymers have shown potential use in a wide range of applications, it is challenging to precisely regulate their thermal response behaviour due to the fact that their critical solution temperature (CST) is influenced by several variables. The way that polymer molecules and solvents interact may be changed by varying the proportion of salt. Changes in CST and difficulties precisely controlling the temperature nodes of thermal response in real-world applications result from the strengthening of the interaction between ions and polymer molecular chains caused by an increase in salt concentration. This interaction may also break the hydrogen bonds between solvent and polymer molecules [92].

CST is significantly impacted by additives as well. Certain additives have the ability to change the solvation environment around polymers or create novel chemical or physical interactions with them [93]. For instance, certain macromolecular additives may raise CST through complexation or steric hindrance, whereas specific small-molecule additives may be incorporated between polymer segments to improve segment activity and lower CST. Certain thermal-responsive hydrogels function poorly and are unstable in high-temperature settings. Thermal-responsive hydrogels will dry and shrink if the surrounding temperature rises above their particular temperature. At this point, the hydrogel's swelling, mechanical strength, and loading, as well as the release of medications and other substances, are all affected because many water molecules are separated, and its three-dimensional network structure contracts. In the biomedical area, for instance, contraction brought on by dehydration at high temperatures may result in uncontrolled medication release, which might compromise the therapeutic impact [94].

The phase transition behaviour seen at LCST or UCST (i.e., cloud point) serves as the basis for the design idea of the great majority of temperature-responsive components. Interestingly, certain interactions between solvent and polymer molecules are necessary for the LCST behaviour. Cosolvents, salt, and surfactants are examples of additives that may change the transition temperature and modify polymer-solvent interactions. Therefore, it is necessary to carefully determine and design the transition temperature in line with the intended use [95].

Chen et al. [96] synthesized a nanofibrous membrane/thermoresponsive hydrogel composite (PBNCs@PLA/P(NIPAM-AM), that has the capacity to control heat production for the treatment of infected wounds. The PB NCs@PLA/P(NIPAM-AM) hydrogels were made by loading prussian blue nanocubes (PB NCs) onto the polylactic acid (PLA) nanofibrous membrane and decorating it. NIPAM and acrylamide (AM) were free radical polymerized to create P(NIPAM-AM) hydrogels. Strong NIR light absorption (λ = 700–900 nm) and high photothermal conversion efficiency were characteristics of PB NCs that gave the nanofiber membrane exceptional photothermal qualities. Furthermore, by altering the ratio of NIPAM to AM, the phase transition temperature of P(NIPAM-AM) hydrogels was brought into line with the optimal treatment temperature (45–50 °C). Under 808 nm laser irradiation, PB NCs@PLA nanofibrous membranes containing PB NCs conducted a significant amount of photothermal energy to P(NIPAM-AM) hydrogels, raising the hydrogel's temperature to the phase transition temperature and causing a phase transition to form quickly to block light from passing through. The PB NCs@PLA nanofiber membrane's photothermal temperature is maintained at or near cloud point temperature (T_{cp}) via the aforementioned dynamic and reversible P(NIPAM-AM) hydrogel processes. Results from both in vitro and in vivo experiments showed that PB NCs@PLA/P(NIPAM-AM) hydrogels had a strong antibacterial impact and had little negative effect on the healthy tissues around the wound, which successfully sped up the healing of infected wounds (**Figure 3A**).

Zhou and co-workers [97] developed and used a thermoresponsive hydrogel stabilizer

View Article Online
DOI: 10.1039/D6MA00167J

(HSZ) in ecological soil. The thermoresponsive hydrogel curing agent, known as HSZ, was made up of a responsive carrier and a precursor solution with a mass ratio of 6.4:1. In contrast, NSZ, a traditional hydrogel curing agent made of a crosslinker and a precursor solution, was also made. 0.44 g of poly(vinyl alcohol) (PVA) was dissolved in 100 g of deionized water at 95 °C for 30 minutes while being stirred magnetically to produce precursor solution A. Following cooling, 0.21 g tetramethylethylenediamine (TMEDA), 0.066 g N,N′-methylenebisacrylamide (BIS), and 5.89 g acrylamide (AAm) were added and physically mixed. Two stages were taken in the fabrication of the thermally sensitive carrier:

i. Porous ceramic particle activation: porous ceramsite was immersed in 30 wt.% ammonium persulfate (APS) solution under vacuum (<20 kPa) for 12 h, then vacuum-dried for 4 h, yielding APS-loaded porous ceramic particles.
ii. Encapsulation: paraffin was melted at 75 °C, and 3 wt.% activated carbon powder was mixed in. The APS-loaded porous ceramics were added, coated evenly with molten paraffin, and then cooled in water to solidify.

Figure 3B provides a graphic representation of this procedure. According to the experimental findings, the ecological soil containing HSZ has a high flowability before to response, but after reaction, its flowability quickly drops and its consistency abruptly rises. The ecological soil's 7-day unconfined compressive strength is 1.55 MPa with the addition of HSZ. The ecological soil typically has a pH between 6.5 and 8.0, and plants grow well in a simulated vegetation box. When the core-shell microcarriers respond thermally, they release crosslinking components from the carrier, which quickly react with the components of the precursor solution to create a curing system, as shown by conductivity and viscosity tests. This work significantly increases the ecological soil's ability to respond to a variety of difficult situations by offering a unique way to control it using a responsive stabilizer.

To address the growing problems of coal spontaneous combustion (CSC) and solid waste management, a thermosensitive composite hydrogel was made by Hao et al. [98] from solid waste, is presented together with its application engineering design. In a mortar, a predetermined quantity of fly ash (FA) powder and hydroxypropyl guar gum (HGG) powder were combined and crushed. To get a consistent suspension, the mixed powder was dissolved in deionized water and swirled for five minutes. To create a stable solution, a determined quantity of crosslinking agent solution was then added and stirred. The mixture was let to remain at room temperature for three minutes to create the HGG@FA hydrogel. The same process was used to make the HGG hydrogel for comparison, with fly ash omitted. **Figure 3C**

View Article Online
DOI: 10.1039/D6MA00167J

depicts the HGG@FA hydrogel preparation procedure. The final HGG@FA hydrogel produced in the lab showed excellent structural integrity and strong elasticity. Rapid gelation, robust water retention, transport-friendly viscosity, and clever thermal responsiveness for coal cooling and sealing were all shown by the hydrogel. Strong fire suppression and anti-reignition capabilities were shown, together with good inhibition of oxidation, delayed ignition, and decreased gas emissions. HGG@FA hydrogel has potential for large-scale mine fire control applications because of its affordability, environmental friendliness, and ease of preparation.

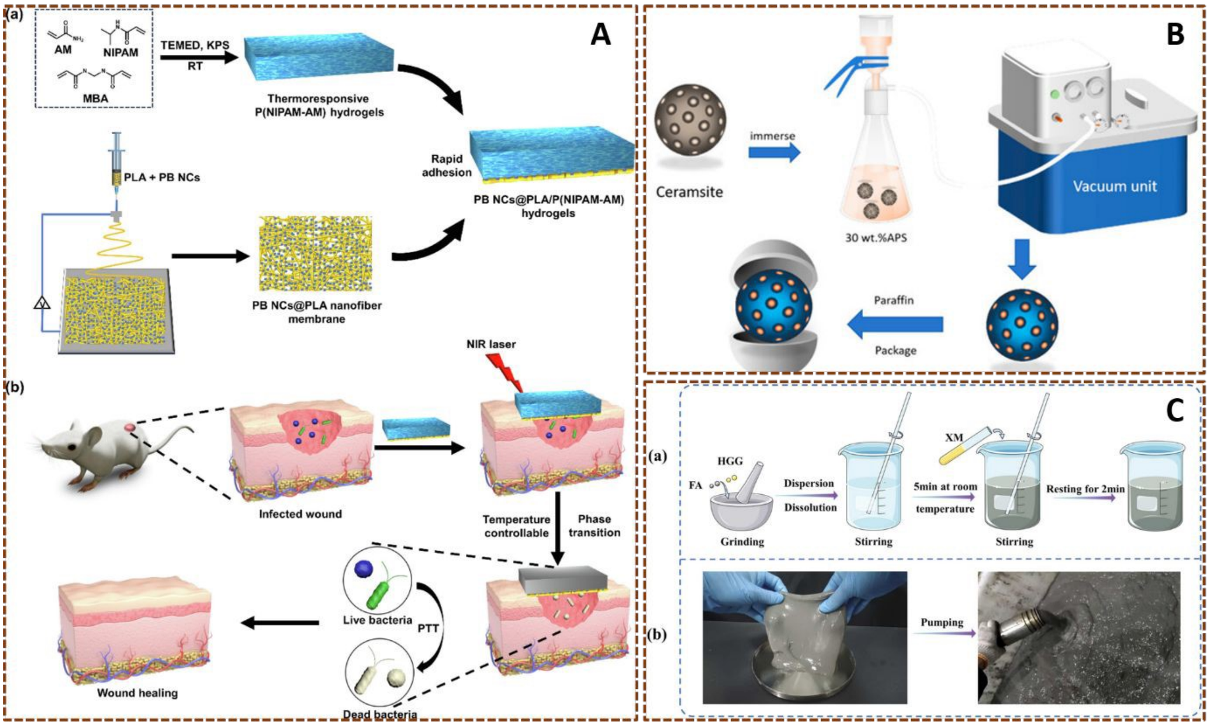


Figure 3: (A) Diagrammatic representations of (a) PB NCs@PLA/P(NIPAM-AM) hydrogel synthesis and (b) PB NCs@PLA/P(NIPAM-AM) hydrogels for wound healing enhancement. Reproduced with permission from ref. [96]. Copyright 2025 Elsevier. **(B)** The procedure for making thermally sensitive carriers [97] (Open Access). **(C)** Preparation process of HGG@FA hydrogel: (a) Laboratory synthesis steps, (b) Fabricated HGG@FA hydrogel ready for pumping. Reproduced with permission from ref. [98]. Copyright 2025 Elsevier.

Functional Mechanism of Thermoresponsive Hydrogels

Depending on whether they are lower critical solution temperature (LCST) or upper critical solution temperature (UCST) kinds, thermoresponsive hydrogels display different modes of operation. Both hydrophilic and hydrophobic segments are present in LCST hydrogels; a reversible sol–gel transition is induced when the temperature increases over the critical solution temperature (CST), and hydrophobic interactions take over [99,100]. Changes in optical transmittance, volume, and adhesion often accompany this process, which qualifies these systems for multipurpose uses [101–103]. On the other hand, UCST hydrogels depend on various driving factors depending on their composition. While cooling below the UCST causes polymer dehydration, chain collapse, and gel formation, higher temperatures cause nonionic UCST hydrogels, rich in hydrophilic groups, to experience weaker hydrogen bonding, improving polymer solubility. Instead, zwitterionic UCST hydrogels experience phase transitions controlled by electrostatic interactions; the sol–gel transition is triggered when polymer chains assemble due to changes in hydration and ion shielding caused by temperature [104–106].

4.1.2 Fabrication strategies and action mechanisms of pH-responsive hydrogels

Hydrogels that react to pH variations are ideal for regulated and site-specific medication delivery [107]. Because functional groups like amines, carboxyl (-COOH), and imines may donate or take protons based on the pH of the surrounding environment, they provide the hydrogel structure pH sensitivity and change its chemical and physical characteristics [108]. Hydrogel design has made use of a variety of synthetic polymers because of their biodegradability, non-toxicity, and biocompatibility [109]. Using a free-radical polymerization technique, hydrogels were created by Liaqat et al. [110] with the primary goal of releasing mesalamine (5-aminosalicylic acid) in the colon in a modified way. Methylenebis (acrylamide) was used to cross-link varying amounts of hyaluronic acid with methacrylic and acrylic acids. After dissolving hyaluronic acid in water while being magnetically stirred, methacrylic and acrylic acids were added gradually. Methylene bisacrylamide (MBA) was dissolved as a crosslinker, while ammonium persulfate (APS) solution was made independently and used as an initiator. After combining and thoroughly mixing the ingredients, they were put into glass tubes, sealed, and polymerized in a water bath at 55 °C for one hour and 60 °C for two hours. Fourier transform infrared spectroscopy confirmed the formation of a new polymeric network and successful drug incorporation. Thermogravimetric analysis revealed improved thermal stability and SEM revealed a rough surface which is desirable for drug loading and medium penetration. The swelling and release studies showed that the drug release was maximum at pH 7.4 and the higher the concentration of the polymer, the higher the loading, release and gel fraction. Toxicity studies demonstrated biocompatibility, making these pH-sensitive hydrogels potential carriers for colon-targeted therapy, especially for inflammatory bowel disease (IBD)

View Article Online
DOI: 10.1039/D6MA00167J

(Figure 4A).

Wang and the team [111] designed a study that used cationic guar gum (CG) and hyaluronic acid (HA) to create a multifunctional pH-responsive polysaccharide hydrogel. Hydrogen bonding and electrostatic interactions helped to produce the basic CG-HA hydrogel. The FA/β-CD combination was created by encapsulating ferulic acid (FA) using β-cyclodextrin (β-CD) via host–guest inclusion complexation in order to improve therapeutic activity. The FA/β-CD@CG-HA-Zn hydrogel drug delivery platform was created by integrating this combination with zinc ions (Zn^{2+}) into the CG-HA hydrogel. Because of its pH responsiveness and self-healing properties, the resultant FA/β-CD@CG-HA-Zn hydrogel may be injected into wounds with uneven shapes for efficient wound covering and protection. Surprisingly, this technology served as an intelligent drug delivery mechanism that allowed therapeutic compounds to be released on demand and in response to pH. Furthermore, the hydrogel showed antibacterial activity and the ability to suppress the production of pro-inflammatory cytokines, which sped up the healing of wounds (**Figure 4B**). All things considered, the pH-sensitive, self-healing FA/β-CD@CG-HA-Zn hydrogel with several uses provides regulated drug release and has great promise for the treatment of infected wounds.

A recent research used free radical polymerization to create a pH-responsive hydrogel system based on Pluronic F127 for the regulated administration of Famciclovir [112]. With little expansion in gastric fluid (pH 1.2) and noticeable swelling in intestinal fluid (pH 7.4), the formulations displayed pH-dependent swelling. Drug incorporation, amorphous conversion, and porous structure were validated by FTIR, PXRD, and SEM investigations, which showed that drug loading varied from 60.89% to 73.74%. With limited drug release in stomach fluid (25.55–34.89%) and almost full release in intestinal fluid (90.12–99.13%) over 36 hours, the hydrogels showed improved heat stability and sustained release. For hydrogels, release kinetics were zero-order, as opposed to first-order for commercially available tablets. Biosafety was established by toxicity studies conducted on rabbits. All things considered, the Pluronic-based approach exhibits potential as a site-specific intestinal carrier of Famciclovir, allowing for longer release, fewer stomach adverse effects, and a lower frequency of dosage in the treatment of herpes and varicella zoster.

Underlying mechanism of pH-responsive PTHs

Typically, networks of polymers with ionizable groups like carboxyl, sulfonic, or amine functionalities are used to create pH-sensitive hydrogels. These groups undergo protonation or deprotonation in settings with different pH values, which causes electrostatic repulsion within the network. Hydrogel swelling is caused by this repulsion, which also increases the distance between polymer chains and improves water absorption. On the other hand, decreased charge repulsion causes the network to contract when these groups revert to their neutral, non-ionized state. Controlling the density and intensity of hydrogen bonding connections between functional groups is another way to alter the degree of pH responsiveness. In order to precisely control swelling and deswelling behaviour, Yang and colleagues, for example, created a phase-transition hydrogel called poly(acrylic acid-co-acrylamide)-hydroxypropyl cellulose (PACA-HPC), in which the hydrogen bonding between the carboxyl groups of poly(acrylic acid-co-acrylamide) and the hydroxyl groups of hydroxypropyl cellulose was pH-dependent [113].

4.1.3 Design and mechanistic pathways of light-responsive PTHs

Viscoelasticity can be precisely tuned thanks to light-based techniques, which provide quick, reversible, and targeted control of hydrogel mechanics [114,115]. Although most methods depend on UV light and external photoinitiators, which may be harmful and modify the chemistry of hydrogels, techniques like allyl sulfide cross-linkers, thiolene additions, and disulfide exchanges provide dynamic control [116–118]. For the safe, long-term control of viscoelasticity, innovative techniques that use biocompatible light sources and eschew extra chemical components are required since stress relaxation in tissues takes place across a variety of durations. Lu et al. [55] provided two mechanisms for controlling stress relaxation in dynamic hydrogels: intrinsic and light-responsive. Through reversible imine metathesis, the intrinsic relaxation period may be adjusted from 3400 s to 500 s using dialdehyde crosslinkers that create Schiff base bonds with carboxymethyl chitosan (**Figure 4C**). Thiuram disulfide (TDS) was added to increase visible light responsiveness. Under light, S–S bonds interchange more quickly, cutting down on relaxation time. This design functions under non-cytotoxic light, allows for prolonged exposure, and provides accurate, on-demand control without the need for external substances. Using this platform, photopatterned surfaces were used to study cell migration and spreading under spatiotemporal changes in viscoelasticity.

The majority of hydrogel models provide static mechanics, although mechanotransduction governs cell function via dynamic changes in tissue elasticity throughout development, remodelling, and ageing. Although azobenzene-based photoresponsive hydrogels provide adjustable stiffness, their dependence on UV light poses a danger to DNA and cells, making it more difficult to interpret the findings. This was addressed by developing a polyacrylamide–azobenzene (PAMA) hydrogel system that reacts to biocompatible blue and green light, allowing for a clear investigation of the dynamics of cellular mechanosensing

View Article Online
DOI: 10.1039/D6MA00167J

[119]. After heating a polymer solution containing 40 mM tetrachloroazobenzene dimethacrylate and 45% acrylamide in DMSO to 100 °C until it became transparent, 1% AIBN was added as a thermal initiator. A ≈200 μm gel layer was created by casting around 30 μL of the mixture between a silanized coverslip and a CTMS-silanized glass slide. The gel layer was then cured for three minutes at 100 °C. To guarantee solvent removal, the produced gels were carefully removed and submerged in PBS at 4 °C for two days, with regular solution exchanges. The gels were activated with sulfo-SANPAH under UV light, completely cleaned with PBS, and incubated with 50 μg/mL collagen-I for a whole night at 4 °C to ensure cell compatibility. The collagen-coated gels were prepared for cell seeding and further research after a final washing. Rapid and reversible stiffness modulation between 19 and 4 kPa is made possible by the hydrogel platform, which causes mesenchymal stromal cells (MSCs) to exhibit quick mechanosensitive reactions, including changes in shape and YAP (yes-associated protein) localization. Cell spreading allows early-passage MSCs to swiftly adjust to changes in dynamic stiffness, but late-passage MSCs react more slowly and have noticeable nuclear lamina wrinkling, which indicates decreased mechanosensitivity. Key elements of mechanosensing during cellular ageing are highlighted by these findings. This system has wide-ranging promise in regenerative medicine and cancer research since it can replicate tissue formation, homeostasis, ageing, and pathological states, including fibrosis and tumour growth.

The complex and unfavourable microenvironment of diabetic wounds makes them a great therapeutic challenge, as it slows down the natural healing process. Unlike conventional wound dressings, hydrogels have several functions such as antibacterial, anti-inflammatory and tissue repair. Ma and the coworkers [120] designed a light-responsive, injectable hydrogel based on chitosan methacryloyl (CSMA) that contained soy isoflavones (SIs) and gold nanoparticles (AuNPs). After making a solution of soy isoflavones in ultrapure water to the necessary concentration, lyophilized CSMA was added to get the final concentration down to 20 mg/mL. After adding 10 mg/mL of the photoinitiator lithium phenyl-2,4,6-trimethylbenzoylphosphinate (LAP), the liquid was agitated until the CSMA was completely dissolved. The AuNP solution was then added at a 1:10 ratio and well mixed. The CSMA/SI/AuNP hydrogel was formed by casting this final combination into moulds and exposing it to UV radiation (365 nm, 4 mW/cm^2) for 30 seconds. According to the results, the hydrogel has exceptional histocompatibility and significantly boosts tissue regeneration via collagen deposition, angiogenesis stimulation, local inflammation reduction, wound closure speed, and skin appendage restoration. It is a light-responsive, injectable substance that easily adapts to wound sites and gains advantageous mechanical strength with very little stimulation.

View Article Online
DOI: 10.1039/D6MA00167J

It is also non-toxic, which makes it ideal for treating chronic wounds and complicated diabetes conditions. All things considered, this hydrogel is a multipurpose and effective biomaterial that can quickly restore the damaged skin's structural integrity and functional qualities, making it the perfect therapeutic wound dressing.

For enhanced utility, soft robots need spatially controlled actuation and sophisticated shape-morphing capabilities. Self-healing, near-infrared (NIR) light-responsive shape memory granular hydrogels (SLSH) were created in this context and printed in three dimensions [120]. The system was designed using four different types of microgels: epoxy-containing microgels (MGs), which react with Jeffamine to form a chemically cross-linked shape memory network; polydopamine-modified microgels (PMGs), which impart photothermal responsiveness and antibacterial properties; and β-cyclodextrin-modified microgels (CDMGs) and 1-adamantylamine-modified microgels (AMGs), which create host-guest interactions that provide self-healing and shear-thinning behaviour (**Figure 4D**). The work increased the flexibility and performance of hydrogels in complicated settings by combining many functional microgels, each of which played a unique role. It offers novel strategies for application in dynamic robotics and fills important gaps in current knowledge of granular hydrogels. Special properties, such as self-healing ability, reliable shape memory effect and shear-thinning behaviour, are introduced by the incorporation of microgels functionalized with β-cyclodextrin, polydopamine, 1-adamantylamine and epoxy. Strong thermal reactivity to NIR stimulation is another feature of these hydrogels that enables precise control over shape recovery and versatility for a range of applications. Additionally, polydopamine's antibacterial and photothermal qualities make it more appropriate for use in biomedicine, especially in infection control and less invasive procedures. These efforts collectively lay the foundation for the creation of smart materials that can adapt to varying conditions without compromising their functionality and biocompatibility.

View Article Online
DOI: 10.1039/D5MA00167J

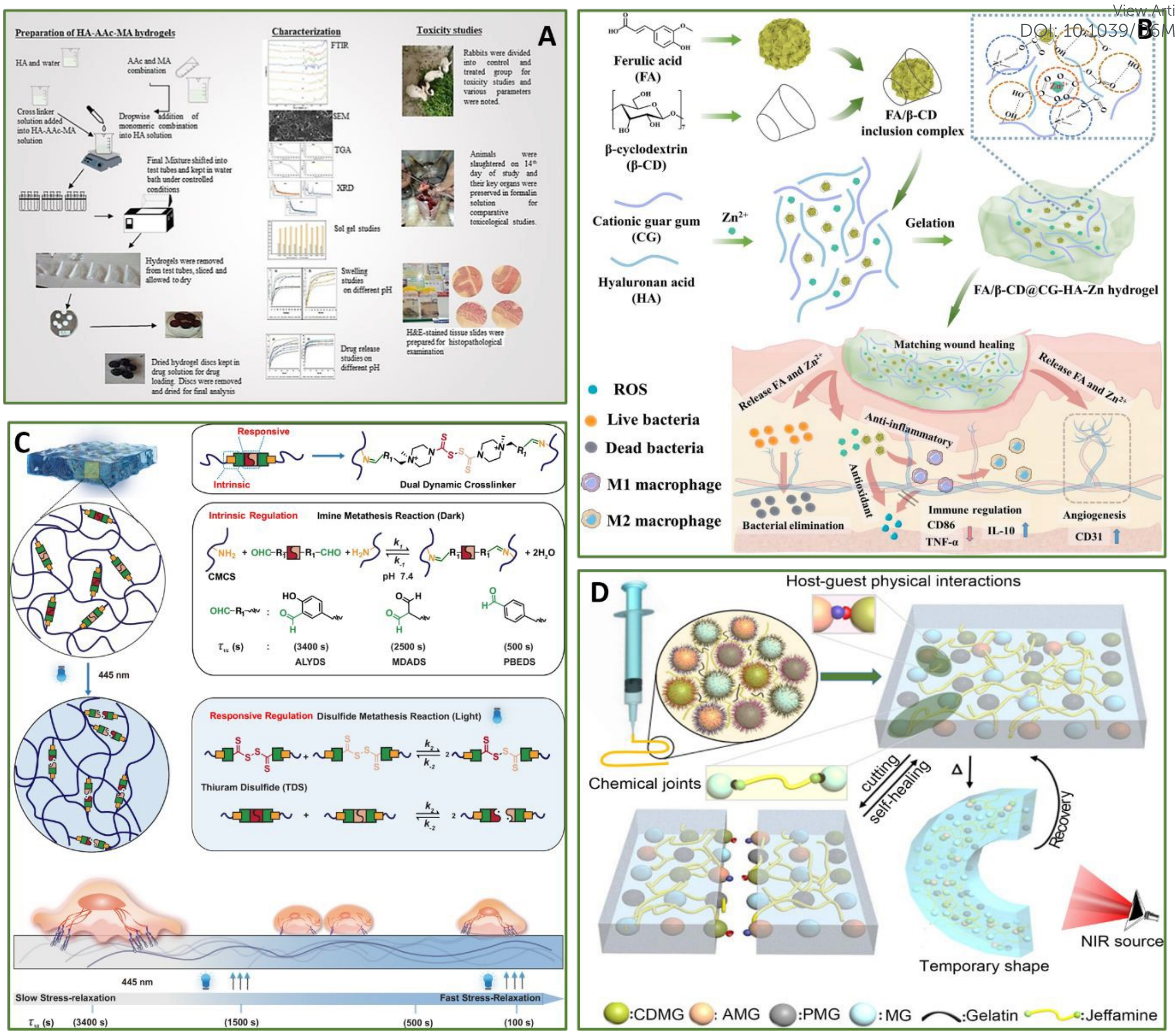


Figure 4: (A) Preparation, characterization, and toxicity studies of pH-responsive hydrogel [110] (Open Access). **(B)** Schematic illustration of the fabrication and application of the self-healing, pH-responsive FA/β-CD@CG-HA-Zn hydrogel as a multifunctional dressing for treating infected wounds. Reproduced with permission from ref. [111]. Copyright 2025 Elsevier. **(C)** The diagram illustrates the hydrogel's dual regulation of stress relaxation: an intrinsic mechanism and a light-triggered responsive mechanism. The material is synthesized via a Schiff base reaction between dual dynamic crosslinkers and carboxymethyl-chitosan (CMCS, 30 kDa). Intrinsic stress relaxation, adjustable from 3400 s to 500 s, is achieved by modifying the aldehyde components through imine metathesis. Meanwhile, the incorporated TDS unit undergoes radical-mediated disulfide metathesis under light exposure, causing dose-dependent polymer network reorganization and enabling precise control of stress relaxation [55] (Open Access). **(D)** Diagrammatic illustration of the extrusion of SLSH hydrogels and the corresponding chemical and physical interactions throughout the printing process. Particle

View Article Online
DOI: 10.1039/D6MA00167J

jamming might occur during this procedure if contact forces between nearby microgels are created by particle interaction forces. After the extrusion process, Jeffamine reacts with the epoxy groups on MG particles to form chemical cross-links. Reproduced with permission from ref. [120]. Copyright 2025 American Chemical Society.

Light-responsive hydrogels' mode of action

Adding photoactive dyes to temperature-sensitive polymers is necessary for the creation of light-responsive phase transition hydrogels. These dyes raise the temperature of the system by using the photothermal effect to transform certain wavelengths of light into heat. A phase transition in the hydrogel is started when the ensuing heat shift modifies the strength and quantity of hydrogen bonds between polymer chains. For instance, Zhang et al. combined gold nanorods (AuNRs) with N-acrylamide (AAm), acrylonitrile (AN), and N,N′-dimethylacrylamide (DMA) to create P(AAm-co-AN)-b-PDMA-b-P(AAm-co-AN) triblock copolymer [121–123]. The AuNRs demonstrated longitudinal surface plasmon resonance (SPR) in response to 785 nm near-infrared (NIR) light. This produced localized heat that raised the hydrogel's temperature over its critical threshold, causing a transition from gel to solution. By adding different photoactive dyes, such as spiropyrans, azobenzenes, or upconversion nanoparticles, which absorb different wavelengths and cause structural or conformational changes within the polymer network, the responsive wavelength of such systems may be precisely tuned. The responsiveness of the hydrogel can be precisely controlled by changing the molecular structure or dye concentration, which can be applied to photothermal treatment (PTT), controlled drug delivery, and optically controlled soft actuators.

4.1.4 Fabrication methods for electro-magneto-responsive and multi-responsive PTHs

Electro- and magneto-responsive polyelectrolyte hydrogel soft actuators have emerged as viable options in artificial muscles and bionic soft robotics due to their quick reaction, accurate control, and biocompatibility. However, the actuators based on electro-responsive polyelectrolyte hydrogels currently available have poor actuation efficiency when stimulated by low voltage, which restricts their use in practice. To overcome this drawback, Wang et al. [125] developed an electrohydrodynamic (EHD) printed hydrogel actuator using a synergistic approach that combines electric field-guided alignment with in-situ photopolymerization, which is composed of two anionic polyelectrolytes, sodium alginate (SA), sodium p-styrenesulfonate (NaSS), and acrylamide (AM). Polyelectrolyte hydrogels were synthesized using a hybrid electrohydrodynamic printing and in-situ photopolymerization approach

(**Figure 5i a**). The EHD configuration included a three-axis motion platform, a printing system, a control unit, and a high-voltage power supply. Printing was conducted at a flow rate of 200 μL/s, a voltage of 3 kV, a height of 1 mm, and a speed of 0.5 mm/s, with the precursor solution contained inside the nozzle. A stable jet was deposited into a PTFE mold (5 × 20 × 1 mm^3) under an electric field, followed by 12 minutes of UV irradiation to promote photopolymerization and produce the final hydrogel (**Figure 5i b**). The optimized P(SA/NaSS/AM) hydrogel demonstrated a dual-anion synergistic effect when compared to single-anion polyelectrolyte hydrogels like P(SA/AM) and P(NaSS/AM). It achieved an equilibrium water content of 98.3%, a compressive strength of 2.7 MPa, and an enhanced ionic conductivity of 4.547×10^{-3} S/cm. Under a 10 V stimulus, the actuator impressively demonstrated enhanced electro-responsive actuation performance, reaching a maximum bending angle of 169°. Additionally, to confirm its practical application, a bionic fish-tail actuator and an underwater cantilever-type bionic actuator were built. All things considered, this study presents a unique method for creating electro-responsive soft actuators with high actuation efficiency and low energy consumption, underscoring its promising use in underwater robotic systems and artificial muscles.

Hyaluronic acid (clickHA) crosslinked with a modified polyethylene glycol (PEG) precursor was used to create electro-responsive click-hydrogels to treat skin wounds that are difficult to heal [124]. Oxidative polymerization was used to incorporate a conducting polymer, poly(hydroxymethyl-3,4-ethylenedioxythiophene) (PEDOT-MeOH), into a semi-interpenetrating network. In order to create optimized HA-S-ene-PEG hydrogels (clickHA), 31.3 mg of HA-SH was dissolved in 300 μL PBS, pH 7.4 was adjusted with 12–15 μL KOH, and 3.5 mg of alkyne PEG crosslinker (31A) was added in 100 μL PBS. After 20 to 30 seconds of stirring, the liquid was left to gel overnight at 4 °C and then given two water washes. The initial dry weight (w_0), swelling in water for three days to eliminate unreacted precursors, lyophilizing, and reweighing (w_L) were used to calculate the gel fraction (GF, n = 3). These values were used to compute GF.

$$GF\ (\%) = \frac{w_L}{w_0} \times 100 \qquad (1)$$

The hydrogel's biocompatibility and potential for skin restoration were validated by scratch wound-healing and cell survival tests using epithelial cells. Notably, clickHA/PEDOT-MeOH was exposed to a moderate voltage of 0.5 V for 15 minutes, which greatly increased cell migration and allowed for wound closure in about one hour. The conducting

View Article Online
DOI: 10.1039/D6MA00167J

clickHA/PEDOT-MeOH hydrogel in conjunction with electrical stimulation is a viable method for full skin regeneration, according to these results, which validate the original idea. Future research will concentrate on sophisticated tests that use 3D-printed skin models to evaluate angiogenesis, migration, and cell proliferation in more detail.

Zhang et al. [125] created a "fried egg" structure using chitosan-coated ferroferric oxide magnetic nanoparticles (Fe_3O_4@CS MNPs) embedded in a hydrogel. To overcome the issues of graft-associated inflammation, insufficient mechanical strength, and the lack of external control in standard 3D-printed scaffolds *in vivo*, magneto-responsive hydrogel scaffolds were created using 4D printing. The multipurpose composite scaffold for ear cartilage tissue engineering that targets external control of cartilage regeneration *in vitro* and *in vivo*, chondrocyte proliferation, and inflammation suppression. Fe_3O_4@CS MNPs were created, added to GelMA hydrogel, and paired with CT-based 3D ear modelling, as shown in **Figure 5ii A**. Rabbit cartilage auricular chondrocytes were cultivated, combined with the magnetic hydrogel composite to create bio-ink, and then 3D printed with UV crosslinking into ear-shaped scaffolds. The printed scaffolds were placed in naked mice and exposed to external magnetic fields (MF) for 4D regulation, as shown in **Figure 5ii B,C**. This improved macrophage M2 polarization, ECM deposition, and chondrocyte proliferation. In vivo culture time was greatly reduced by the FOGS (GelMA + Fe_3O_4@CS) scaffold, which demonstrated improved chondrocyte development and ECM secretion under MF. Three months later, the scaffold's morphology, histology, and mechanics were very similar to those of natural cartilage. Additionally, by encouraging M2 polarization via the activation of the JAK2/STAT3 pathway, lowering inflammation, and boosting cartilage formation, FOGS enhanced the immune microenvironment. All things considered, this magnetic hydrogel scaffold exhibits excellent mechanical integrity, anti-inflammatory properties, and biocompatibility, providing a viable approach to ear cartilage regeneration.

A new pyrene-appended dipeptide, Py-E-A, with L-glutamic acid (E) and L-alanine (A) forming the dipeptide core, was used to create a strong, self-healing hydrogel (**Figure 5iii and Figure 5iv a**) [126]. In order to (i) impart amphiphilicity, (ii) promote π–π stacking–driven self-assembly, (iii) act as an electron donor for charge-transfer (CT) gelation, and (iv) take advantage of the luminescent property of pyrene for imaging, Py-E-A combines the hydrophilic E-A unit, which contains two carboxyl (–COOH) groups, with a hydrophobic pyrenyl group. While Py-E-A dissolves in ethanol (EtOH), methanol (MeOH), and dimethylformamide (DMF), it produces opaque hydrogels with a critical gelation concentration (CGC) of about 8.2 mM when water is added (450 µL to 50 µL of solution) (**Figure 5iv c**). Its ambidextrous gelator

View Article Online
DOI: 10.1039/D6MA00167J

character is confirmed by the fact that gelation also takes place in acetonitrile (ACN) as stable organogels and in EtOH/H_2O, MeOH/H_2O, and DMF/H_2O mixtures at greater CGC.

Additionally, naphthalenetetracarboxylic dianhydride (NDI) and β-alanine (β-Ala) were combined to create NDI-β-A (**Figure 5iv b**), which dissolves in DMF. A deep violet CT complex was produced by combining NDI-β-A (acceptor) and Py-E-A (donor) in a 1:1 ratio. With a CGC of around 1.4 mM, far lower than that of Py-E-A alone, this CT complex instantly produced a stable hydrogel (**Figure 5iv d**) with the addition of water, identifying it as a supergelator with significantly improved hierarchical co-assembly efficiency.

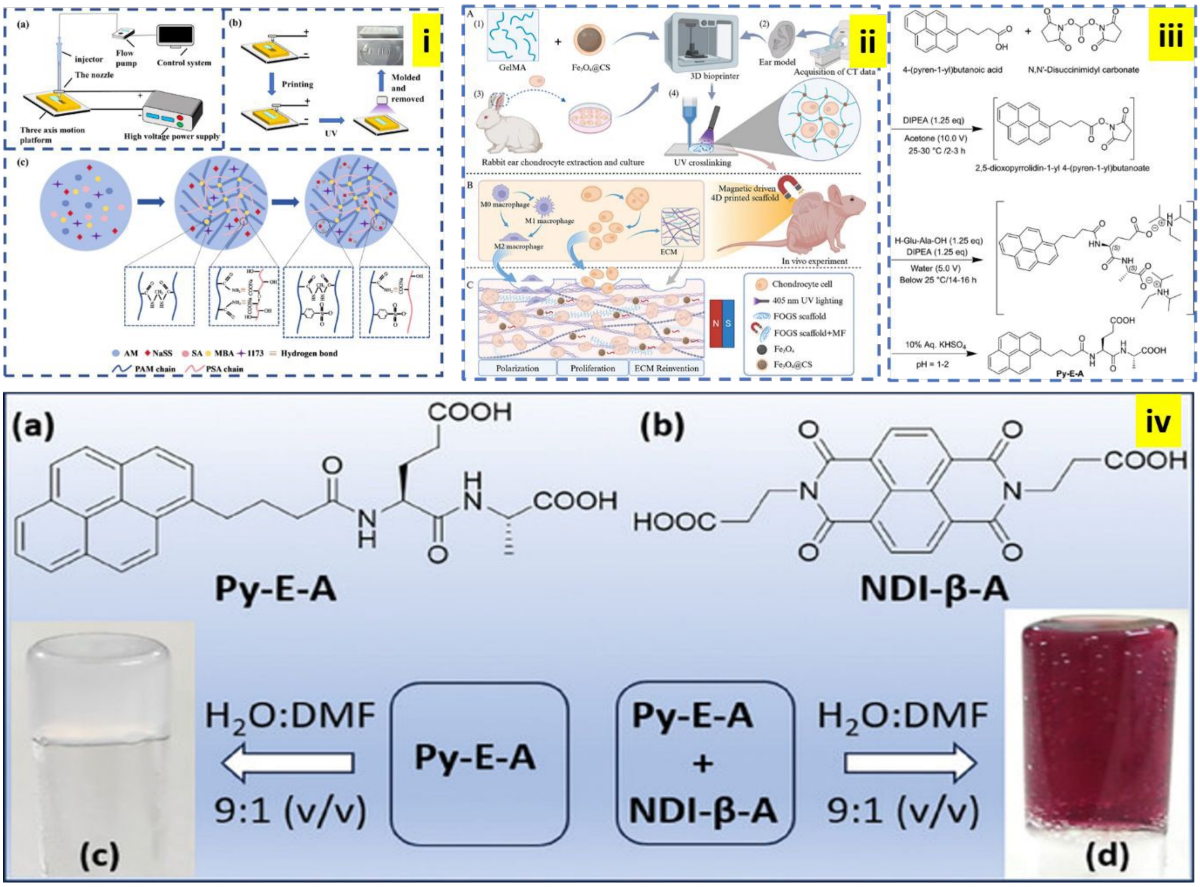


Figure 5: **(i)** (a) Diagram of the EHD printing apparatus, (b) production method of the polyelectrolyte hydrogel, and (c) mechanism of P(SA/NaSS/AM) hydrogel synthesis. Reproduced with permission from ref. [127]. Copyright 2025 Elsevier. **(ii)** The experimental process is shown in the figure and is broken down into four primary stages: A) Magnetic Hydrogel Scaffolds 3D Printed: 1) GelMA hydrogel is used to prepare and functionalize magnetic nanoparticles; 2) CT scanning and computer modeling are used to create a 3D ear model; 3) auricular chondrocytes are taken from rabbit ear cartilage, cultured in vitro, and then

combined with the magnetic nanoparticles hydrogel composite made in 1) to create the magnetic nanoparticles hydrogel composite bio-ink; 4) Ear-shaped magnetic nanoparticles hydrogel composite scaffolds are printed using 3D printing technology in conjunction with UV crosslinking. B) Implantation and Magnetic Driven 4D Printed Scaffold: Mice were given the 3D printed scaffold subcutaneously, and to accomplish 4D magnetic driven scaffold regulation, an external MF was applied. C) *In vivo* Scaffold Development: The scaffold experiences physiological alterations such as chondrocyte proliferation, extracellular matrix secretion, and macrophage M2 polarization, eventually resulting in cartilage regeneration, under the influence of the external MF and the subcutaneous environment. Reproduced with permission from ref. [125]. Copyright 2025 Wiley. **(iii)** Py-E-A synthetic process. **(iv)** a, b) Py-E-A and NDI-β-A's chemical structures, respectively. c) Py-E-A hydrogel photo taken at [Py-E-A] = 8.2 mM. d) Image of the CT hydrogel in H2O:DMF (9:1, v/v) generated at [Py-E-A] = [NDI-β-A] = 8.2 mM. Reproduced with permission from ref. [126]. Copyright 2025 Wiley.

Devices built from optimized multifunctional materials are required in many application fields, particularly biomedicine, to fulfil specific requirements, such as guaranteeing biocompatibility, focusing on specific sites, producing heat, or allowing the controlled release or permeation of substances in response to external stimuli [128–131]. It gets more challenging to include so many features as device dimensions decrease. The integration of different nanomaterials or components into hybrid structures, which combine their unique properties, is a common method to achieve such multifunctionality. Given their capacity to undergo phase transitions in response to temperature changes or other external stimuli, "smart materials" like responsive polymers hold special promise for biological applications, such as medication delivery, gene transfer, and tissue engineering [132–134]. The capacity to release the medications in a particular location is lost when thermo-responsive hydrogels are employed solely because they either need tissue heating or require the pharmaceuticals to be released at body temperature. Combining magnetic nanoparticles with responsive polymers to create composite hydrogels known as "ferrogels" is one method to provide a controlled release [86,135]. Magnetic fields may be used to transport these ferrogels to a specific place inside the body, and magnetic hyperthermia can then cause the hydrogels to undergo a phase change [136,137].

In the work carried out by Schutz and the team [57], a twofold thermo-responsive graft copolymer was used to synthesize and characterize a magneto-plasmonic $CoFe_2O_4$@Au@Polymer hybrid. The trithiocarbonate (TTC) end group of the polymers, which were made via reversible addition–fragmentation chain transfer (RAFT) polymerization, was used to attach to the surfaces of the particles. Crosslinking the colloidal hybrid created multi-responsive hydrogels with temperature-dependent swelling and the magnetic/optical characteristics of magneto-plasmonic nanoparticles (MP-NPs). Effective water ejection was demonstrated by photothermal studies conducted under near-infrared (NIR) irradiation (**Figure 6A**), exhibiting component synergy. This approach, which was based on MP-NPs and TTC-terminated polymers, may be modified to create materials with surface functionalities and transition temperatures that can be adjusted. The promise of these hybrid systems for biological applications, including targeted release and controlled distribution, was highlighted by their combined responsiveness, heat production, and release capability.

Because of their deep tissue penetration and powerful remote actuation, hydrogels sensitive to second near-infrared (NIR-II) light have great promise for biomedical applications; nevertheless, single-response systems often fall short of clinical complexity. In order to get around this, Li et al. [138] combined phenylboronic acid-modified PEI functionalized multi-walled carbon nanotubes (PP-CNT) with alginate dialdehyde (ADA), polyethyleneimine (PEI), and poly(N-isopropylacrylamide) (PNIPAM) to create a multi-responsive nanocomposite hydrogel (APN/PP-CNT). Stabilized by dynamic imine and boronate ester linkages, the network exhibits self-healing and shear-thinning characteristics and reacts to a variety of stimuli, including temperature, pH, hydrogen peroxide ($H2O_2$), and NIR-II light. Uniform CNT dispersion improved hydrogel stability, prolonged release profiles, and decreased nanotube leakage, whereas PNIPAM allowed thermally induced release by contraction under NIR-II irradiation. Effective NIR-II-triggered drug release was shown in both in vitro and in vivo tests, proving APN/PP-CNT as a flexible option for targeted delivery applications (**Figure 6B**).

View Article Online
DOI: 10.1039/D6MA00167J

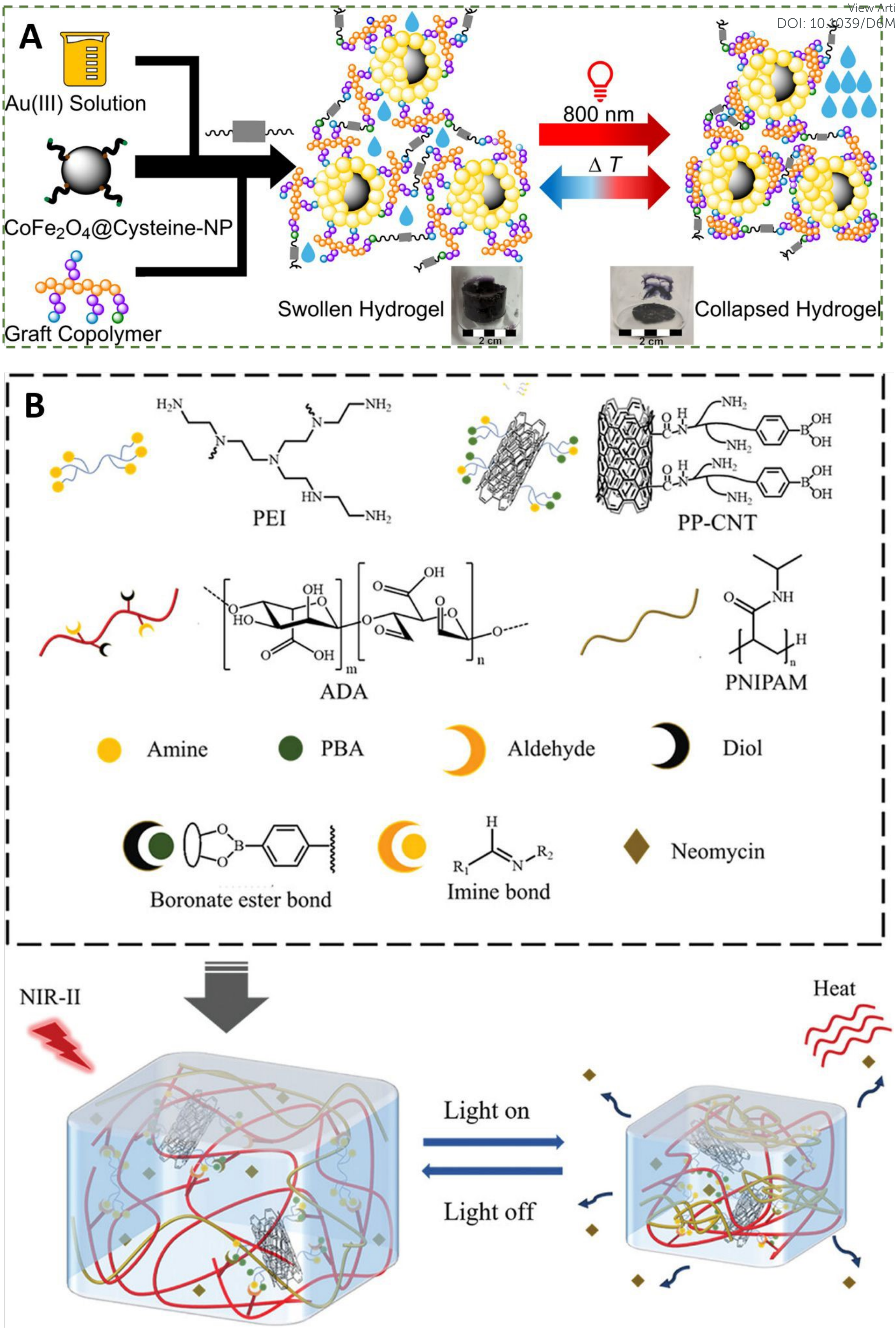


Figure 6: **(A)** Creation and evaluation of a twofold thermoresponsive graft copolymer-based magneto-plasmonic $CoFe_2O_4$@Au@Polymer hybrid material. Reproduced with permission from ref. [57]. Copyright 2025 Elsevier. **(B)** The materials and chemical crosslinks of the APN/PP-CNT nanocomposite hydrogel are shown schematically, along with a description of the drug release from the hydrogel network that is NIR-II sensitive. Reproduced with permission from ref. [138]. Copyright 2025 Wiley.

Mechanistic insights

Phase transition hydrogels have a lot of promise for use in medicine, especially in the treatment of cancer. By concentrating ultrasound beams on certain locations, the well-known imaging and therapeutic method known as high-intensity focused ultrasound (HIFU) may heat thermoresponsive hydrogels selectively. Reversible phase changes are triggered by the ensuing localized temperature increase, enabling regulated therapeutic action [139]. Similarly, when magnetic particles are added to the network, magnetic fields improve the performance of phase transition hydrogels [140,141]. These particles produce heat when exposed to an alternating magnetic field (AMF), which modifies the hydrogel's hydrogen bonding and causes a reversible sol–gel transition [7]. For example, when exposed to AMF, Chen et al.'s magnetic hydrogel cluster, which was made up of tissue adhesion monomers, thermoresponsive hydrogel matrices, and magnetic particles, heated quickly from 25.6 to 102.9 °C in only three minutes [142].

Furthermore, by altering hydration states and polymer chain conformations, electric fields may control PTHs [143]. Hydrogel expansion or contraction is driven by ion migration and the electric double-layer effect under an applied field, which permits phase transitions and offers exact control over their characteristics, increasing their potential uses.

4.2 Synthesis techniques and mechanism for PCHs

4.2.1 Preparation methods of PCHs

Hydrogels have a lot of benefits, but in real-world applications, their performance suffers because the gel state varies with temperature. To overcome these temperature-related constraints, scientists have concentrated on creating a range of temperature-regulating materials with distinct qualities, such as wettability, controlled release capabilities, energy storage and release, temperature adjustment and control, and environmental sustainability. In this regard, PCMs are essential as they are often included in hydrogel network matrices to

View Article Online
DOI: 10.1039/D6MA00167J

improve their performance for energy storage uses. The creation of hydrogels that are shape-stable and capable of efficiently encasing PCMs is thus crucial. The methods utilized to produce the hydrogel network may be used to characterize PCH preparation processes. Interpenetrating polymer networks, chemical crosslinking, and physical crosslinking are some of these techniques. Each method has unique benefits and drawbacks that affect how well the final hydrogels work and how useful they are for TES applications. Three different approaches to hydrogel preparation are compared in **Table 3**. It discusses their inherent limitations and their salient features, their strengths and underlying processes. The chart also lists the situations when each approach is most suitable. When combined, these comparisons provide a thorough viewpoint for choosing the best strategy based on application requirements and performance requirements.

Table 3: Comparative overview of preparation methods: mechanisms, features, benefits, limitations and applications of PCHs.

Preparation Method	**Mechanism**	**Key Features**	**Strengths**	**Limitations**	**Applications**	**Ref.**
Physical Crosslinking	Network formation occurs through non-covalent interactions such as hydrogen bonding, hydrophobic association, or ionic forces.	Carried out under mild conditions without chemical reagents.	Easy to implement, preserves the intrinsic properties of materials, avoids harsh chemicals, and maintains biocompatibility.	Weaker network strength, lower durability, and susceptibility to structural disruption under stress or extreme environments.	Suitable for systems where high strength is unnecessary, e.g., biomedical scaffolds requiring biocompatibility but not heavy load-bearing capacity.	[30,144]
Chemical Crosslinking	Involves covalent bond formation between monomers or polymer chains	Provides permanent and strong bonding	Produces stable and robust hydrogels with tunable	Requires strict reaction conditions, catalysts, or	Best for applications demanding strong, durable, and stable hydrogels, such as	[145,146]

	via chemical reactions to yield a 3D crosslinked matrix.	between molecules.	properties; allows fine control of crosslink density; high mechanical and chemical stability.	elevated temperatures; may involve complex purification or post-treatment steps.	controlled drug release, tissue engineering scaffolds, and wound dressings.	
Interpenetrating Polymer Networks (IPNs)	Fabricated by combining two or more polymer networks that interlace without a covalent linkage between them.	Integrates complementary properties of different polymers into one system.	Enables design of multifunctional hydrogels with superior elasticity, toughness, and performance; allows tailoring of advanced material properties.	Preparation is technically challenging, time-intensive, and costly, with multiple synthesis and processing steps.	High-performance automotive components, biomedical hydrogels (drug delivery, tissue engineering), sound/vibration damping, separation membranes, and intelligent adhesives	[147,148]

View Article Online
DOI: 10.1039/D6MA00167J

4.2.1.1 Physical crosslinking

Physically cross-linked hydrogels' primary feature is their structural stability, which is achieved by dynamic and reversible non-covalent bonding interactions. Reversibility, the capability to alter the network structure under various physical circumstances, a relatively low degree of cross-linking, a high swelling capacity, and the ability to swiftly release energy or reattach in response to external stimuli are characteristics of these bonds. Hydrogen bonds, ionic cross-linking, electrostatic contacts, and hydrophobic interactions are examples of common non-covalent bonding interactions. The properties make natural polymers with abundant functional groups, such as carboxyl and hydroxyl groups, more appropriate for physical cross-linking. In addition, the external environment such as pH and temperature can affect the interactions between the molecular chains of these hydrogels, providing the possibility of controlled synthesis and performance tuning of physically crosslinked natural polymer hydrogels. These hydrogels are primarily prepared using freeze-thaw and solution crosslinking procedures. The experiment's most straightforward approach involves physically altering the material by directly adding inorganic salts, tiny organic molecules, surfactants, etc., and then cross-linking it into a mesh structure via host-guest interactions, hydrogen bonds, electrostatic forces, and molecular interactions. The gel's phase transition temperature may be easily lowered with this technique, but modulation is often restricted. For instance, by physically cross-linking at 37 °C via a sol-gel transition, methyl cellulose (MC) and polyvinyl alcohol (PVA) may be mixed in phosphate buffer solution (PBS) to create an MC-PVA gel.

Using a physical non-covalent cross-linking technique, Li and the team [149] created a novel smart window with high frost resistance. Their technique uses glycerol-water (GGW) to create a binary solvent system with significant synergistic hydrogen bonding between the two solvents, in contrast to the covalent cross-linking method (**Figure 7A and B**). Moreover, N-isopropylacrylamide (ONIPAM) undergoes a very quick transition from nematic to spherical non-covalent cross-linking. In contrast to (A), where window breaking occurs as the temperature drops because of covalent cross-linking, these characteristics provide the new smart windows with superior frost resistance, preventing the windows from shrinking and cracking when the temperature drops. An essential contact force in the physical cross-linking process is hydrogen bonding. In order to create a three-dimensional network, Yin et al. [150] employed polyacrylamide as a hydrogel matrix. Aside from the hydrophobic interaction with octadecyl acrylate, where POA was utilized as a PCM to create micro-regions for phase-change energy storage, stable hydrogen bonding was possible between the PAM and the other

View Article Online
DOI: 10.1039/D6MA00167J

experimental PCMs. By combining several solid-liquid organic PCMs that melt at low temperatures due to hydrogen bonding forces, the polyacrylamide PAM structure creates an organogel system with a stable shape, high energy density, and dual phase transition capabilities, as shown in **Figure 7C.** The method of hydrogen bond creation under various environmental circumstances is better clarified by this example.

View Article Online
DOI: 10.1039/D6MA00167J

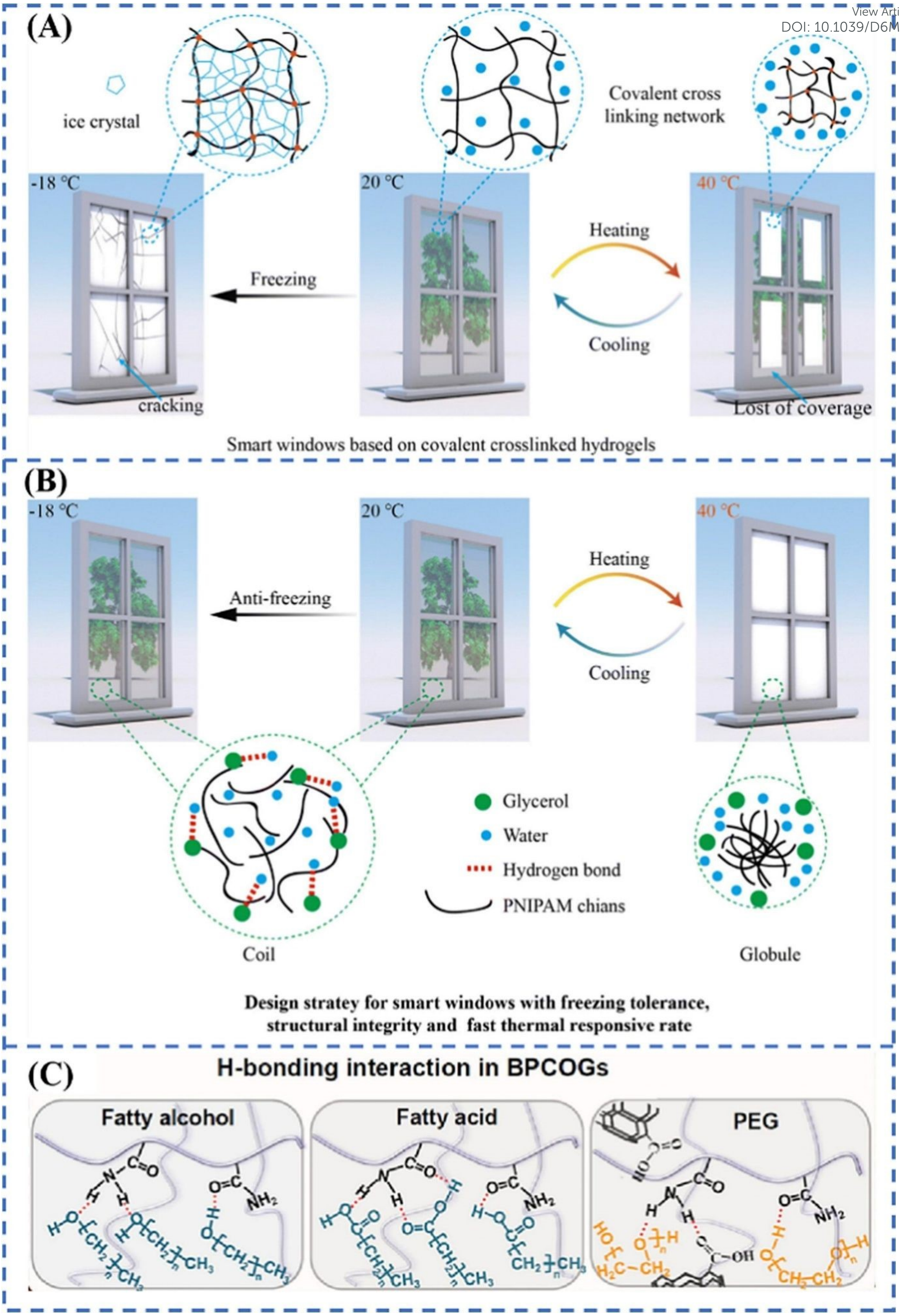


Figure 7: (A) General smart window design strategies using covalently crosslinked hydrogels. **(B)** Smart window concept using a non-covalently crosslinked PNIPAM GW solution, showing structural stability, frost resistance, and fast thermal responsiveness; **(C)** Illustration of hydrogen bonding interactions, as demonstrated by Xgel/fatty acid, Xgel/fatty alcohol, and CNT-Xgel/PEG BPCOG systems. Reproduced with permission from ref. [30]. Copyright 2025 Elsevier.

4.2.1.2 Crosslinking of PCHs using chemical process

Strong three-dimensional networks are produced by chemically crosslinked hydrogels, which are created by processes like click chemistry and free-radical polymerization that create irreversible covalent connections. Chemically crosslinked hydrogels have better structural stability and durability than physically crosslinked ones. Stimuli that encourage the creation of chemical bonds within the macromolecular chains, such as light, heat, high-energy radiation, mechanical forces, ultrasound, or cross-linking agents, often start the cross-linking process. These chemically crosslinked hydrogels are appropriate for applications requiring exceptional mechanical performance and endurance because of their high mechanical strength, limited swelling capacity, and notable chemical stability.

Hydrated salt has been used for thermal management and solar energy applications due to its phase transition properties. However, problems that need immediate care, such as leakage, phase separation, and supercooling, restrict its practical implementation. To overcome these obstacles, a new method was put forward by creating a photothermal phase change hydrogel using chemical crosslinking through acrylamide and konjac glucomannan as the support structure, sodium acetate trihydrate as the phase change material, and graphene oxide as the photothermal agent [66]. The production route of sodium acetate trihydrate–konjac glucomannan/polyacrylamide (SAT-KPAM) is shown in Figure 8A. The photothermal material is graphene oxide (GO), and the crosslinking agent is N, N′-methylenebisacrylamide (MBAA). The liquid SAT is used to dissolve acrylamide (AM) and konjac glucomannan (KGM) to form a homogenous precursor solution, as shown in **Figure 8A.** Thermal initiation triggers polymerization, resulting in a three-dimensional network of polyacrylamide-based composite hydrogel (PCH) that guarantees flexibility and morphological stability. A porous gel structure is encouraged by the interpenetration of konjac glucomannan chains into the polyacrylamide framework via hydrogen-bond interactions in **Figure 8A b**. Finally, acrylamide forms the main backbone in **Figure 8A c** and is crosslinked by N, N′-methylenebisacrylamide to create a strong gel network. The hydrogel's three-dimensional

View Article Online
DOI: 10.1039/D6MA00167J

network limits supercooling to 1.2–3.1 °C and reduces salt leakage. The phase change hydrogel achieves an energy storage density of 179.2 J/g and a photothermal conversion efficiency of 89.7% due to graphene oxide's excellent light absorption. The network topology guarantees effective heat transmission with a thermal conductivity of 0.868 W/(m·K), while the solid–liquid transition of hydrated salts causes a noticeable change in stiffness. These characteristics demonstrate phase change hydrogels' potential for solar energy and personal temperature control.

A family of adhesion-switchable hydrogels is created by combining the adhesion techniques of the snail's epiphragm, which uses dehydration-induced solidification to lock surfaces, and the gecko's toe, which uses surface adhesion of spatulas to enhance contact area. The hydrogels were created by creating supersaturated and stimuli-responsive phase change materials (PAM-SA) by adding $CH_3COONa\cdot 3H_2O$ salt (SA) to polyacrylamide (PAM) aqueous networks [151]. In a mixed solution with sodium acetate, the monomer (acrylamide, AAm) and crosslinker (N,N′-methylenebisacrylamide, MBAA) were polymerized to create PAM-SA. The low crosslinking density (H-PAM-SA) makes the as-prepared samples soft and transparent. A range of H-PAM-SA hydrogels with varying SA contents was made. Rigid and white crystalline composites (C-PAM-SA) were also formed as a result of the $CH_3COONa\cdot 3H_2O$ seed initiating the process. The mechanical characteristics are significantly strengthened by crystallization, and the crystalline C-PAM-SA-120% and soft PAM hydrogel have tensile Young's moduli of 340.7 and 0.1 MPa, respectively. In contrast to PAM hydrogel adhesive, which has an adhesion strength of 7 kPa, PAM-SA-120% exhibits exceptional adhesive performance with an adhesion strength of 348 kPa. H-PAM-SA-120% phase change hydrogels' stimuli-induced crystallization releases heat in a controlled manner, making them suitable for thermochromic materials and thermotherapy.

High-performance photothermal conversion phase change materials (PCMs) have significant potential for improving solar energy use. Using a straightforward one-step heat-initiated chemical crosslinking polymerization technique, Hu et al. [152] created shape-stable phase change hydrogels by adding hydrated salt particles to hydrogel polymer networks. The researchers initially created a significant amount of hydrated salt particles (SEA) using iron-ethyl acetate (Fe–EA) nanoparticles and sodium acetate trihydrate (SAT) as the phase transition core. **Figure 8B a** illustrates the process of creating polyacrylamide/sodium acetate trihydrate–iron–ethyl acetate (PAM/SEA) phase change hydrogels. SEA particles were first weighed and melted at 75 °C in a water bath. N,N′-methylenebisacrylamide (MBA, 2 wt.% relative to AM) and a predetermined quantity of acrylamide (AM) monomer were then added, and the mixture was agitated for 30 minutes in the water bath until a homogeneous dispersion was achieved. After adding ammonium persulfate (APS, 1 wt.% in relation to AM) as the initiator, the liquid was quickly poured into molds to create PAM/SEA hydrogels after being agitated for 20 to 30 seconds. The synthesis procedure is shown in **Figure 8B b**, where acrylamide is thermally initiated to form a crosslinked polymer network in the presence of MBA, and the solvent is a molten SEA solution. SEA loadings of 75, 80, 85, and 90 wt.% were used to create PAM/SEA hydrogels, which are referred to as 75%-PAM/SEA, 80%-PAM/SEA, 85%-PAM/SEA, and 90%-PAM/SEA, respectively. Pure polyacrylamide (PAM) hydrogels devoid of SEA particles were also produced as a point of comparison.

4.2.1.3 The process of interpenetrating polymer networks

When two polymers interweave in a networked structure, with at least one of them chemically crosslinked and the other piercing the network without creating covalent connections, the result is known as an interpenetrating polymer network (IPN). This arrangement preserves the unique qualities of each polymer by producing hydrogels with a dual-component network structure, as seen in **Figure 8C**. A structure like this improves the hydrogel's overall performance by increasing its mechanical strength, improving its thermal stability, improving its water retention, and lowering its swelling ratio. A particular kind of IPN called a double network (DN) is made up of two polymer networks with quite different structures and characteristics. This process is often used to produce hydrogels. This technique is often used to create hydrogels, which may then be crosslinked via techniques like freeze-drying.

If there is no systematic procedure, such a network system can be formed in one pot by reacting all the components of the matrix. By using a freeze-drying technique, Zhu et al. [153] created composite scaffolds of MXene/microcrystalline cellulose (MCC) and silver-multi-walled carbon nanotubes (Ag-MWCNT). This included the addition of a suspension of MXene, MCC, and silver-loaded Ag-MWCNT in an ice bath with stirring and the addition of epichlorohydrin as a cross-linking agent. The MXene, MCC and silver-loaded Ag-MWCNT suspension was directly transferred into an ice bath with stirring, and epichlorohydrin was used as a cross-linking agent to form a three-dimensional networked composite hydrogel in the freezing environment. Then, using the vacuum impregnation process, palmitic acid (PA) PCM was added to the composite scaffold. As a result, the layered porous structure of the Ag-

MWCNT composite scaffold was suitable for the proper containment of the PA PCM. The researchers used Fourier transform infrared spectroscopy (FTIR) to characterize the final PCH to confirm that the structure was as hypothesized. They found that no new characteristic peaks appeared, which meant that only physical interaction between the Ag-MWCNT composite scaffolds and the PCM took place, and no new substances were formed. The interpenetrating network topology also facilitates effective heat transmission, according to thermal conductivity tests.

Recently, some researchers discovered that the gel network may dynamically interact with the PCM, which can result in the gel's self-healing. This is an even more alluring characteristic than the potential for each individual to impose their unique capabilities. Composite hydrogels, which are dynamic dual-network conducting hydrogels based on natural polymers, were created by Peng and co-workers [154] using SA-borax and gelatin. In addition to hydrogen bonding between gelatin molecules, this hydrogel incorporates dynamic borate bonds created by borax and sodium alginate as a matrix. While the secondary network, created by dynamic borate ester bonds, gives outstanding conductivity and quick self-healing qualities, the main network, created by hydrogen bonding, guarantees exceptional mechanical characteristics and shape recovery capabilities. Sodium sulfate (Na2SO$_4$) distributed throughout the hydrogel experienced a phase shift upon cooling using a "gelation-first, phase-change-later" technique; the microcrystalline structure of its hydrate improved the hydrogel's mechanical characteristics and ionic conductivity.

4.2.2 Mechanistic pathways of PCHs

The solid-liquid transitions of the embedded materials, which allow for the storage and release of latent heat, are the main factors influencing the phase change mechanism of hydrogels containing phase change materials (PCHs). Phase change materials are contained and stabilized inside the hydrogel's three-dimensional polymer network via chemical crosslinking, hydrogen bonding, or physical adsorption, which improves cycle stability and stops leakage. When PCMs are heated, their ordered crystalline structure changes into a disordered liquid state and absorbs heat; when they cool, the opposite process occurs, releasing energy. Different behaviour in organic and inorganic PCHs is caused by different mechanisms, which differ depending on the kind of PCM utilized.

Paraffin-based and non-paraffin-based materials are the two main categories into which organic PCMs for energy storage fall. The primary constituents of paraffin-based PCMs are straight-chain alkanes with the formula C_nH_{2n+2}. The phase transition temperature and enthalpy both grow in proportion to the length of the carbon chain. As efficient PCMs, paraffin compounds with carbon chain lengths between 12 and 25 atoms are often used. On the other hand, non-paraffin-based materials include PVA, polyesters, polyethers, and fatty acids. Within a certain temperature range, these materials experience phase transitions, changing from solid to liquid or vice versa, while absorbing or releasing energy [155].

Hydrogels, which are composed of polymer chains and cross-linking agents that create a stable three-dimensional network structure, are often soluble, soft, and slightly elastic before the phase change. The key mechanisms that keep PCMs structurally stable include physical adsorption, chemical bond formation, and other intermolecular interactions. PCMs are embedded in the spaces of the network created by the matrix polymer chains [150,156]. The movement and reorganization of molecular chains is the primary mechanism for the phase transition process of organic PCMs. For example, during the heating process, paraffin waxes change their molecular chains from a regular crystalline arrangement (solid state) to a disordered liquid state; polyol-based PCMs store energy by changing from a layered body-centered structure to an isotropic face-centered one, which eliminates the need for a solid-liquid transition and lowers the possibility of PCM leakage. As seen from a microscopic perspective, this shift entails the dissolution of interlayer hydrogen bonds, which results in a phase transition from crystalline to amorphous and the release of bond energy (**Figure 8D**). The quantity of hydroxyl groups in each molecule determines the significant enthalpy of phase change of long-chain polymer compounds; the more hydroxyl groups there are in a molecule, the higher the enthalpy of phase transition. Because of this, these materials have extended service lives, stable characteristics, and a high enthalpy of phase transition.

However, during the phase shift process, organic PCMs often undergo volume expansion or contraction. Such variations in volume may cause the PCM to separate from the hydrogel matrix or even leak. In addition, organic PCMs typically exhibit high fluidity and low viscosity, which can lead to leakage during phase changes. Therefore, careful selection of polymer network architectures and tuning of the crosslinking density can lead to a slight decrease in PCM leakage [150].

Crystalline hydrated salts, molten salts, metals, or their alloys are examples of inorganic PCHs, a class of energy storage technologies that use inorganic materials as phase change materials (PCMs). These materials have a high thermal conductivity for efficient heat management and energy storage, as well as outstanding chemical stability. Hydrophilic groups present naturally in hydrogels enable efficient binding with hydrated salts. They also have high efficiency as carriers for these PCMs and promote the development of flexible inorganic PCHs due to their crosslinked three-dimensional network structure, which facilitates the encapsulation of molten hydrated salts [66]. During phase transitions, the lattice properties of inorganic phase transition materials can change. For example, sodium sulfate decahydrate dehydrates to an anhydrous salt during the phase transformation process, which is accompanied by a considerable heat absorption process [157]. When the phase transition takes place, the ions in the molten salt are rearranged, and the heat produced increases the thermal vibrations in the material. This leads to the atoms or ions breaking free from the lattice energy and a change in the crystal structure. It's interesting to note that when hydrated salts crystallize, the crystals align in certain directions, creating a preferred orientation that results in optical anisotropy and a distinctive optical texture [66,158]. The optical texture and birefringence of the hydrogel and crystals vanish when the temperature approaches the PCM's melting temperature (T_{mp}), signifying a change from an anisotropic solid phase to an isotropic liquid phase. Conversely, the phase transition process of hydrated salt PCMs may be clearly recognized as the crystal structure gradually reorganizes, as shown by the return of the optical texture and birefringence upon cooling to the freezing point. There are three types of water found in hydrogels: free, intermediate, and bound water [159]. Water molecules mostly reside as bound water when the ambient temperature is lower than the PCM T_{mp}, and the hydrogel's solid crystalline structure predominates. This process occurs concurrently with the hydrated salt's transformation into various crystalline forms when the temperature approaches the T_{mp}: the hydrated salt's crystalline water content gradually drops, and a significant portion of it is transformed into intermediate water in the hydrogel, ultimately leading to the formation of anhydrous salts (as illustrated in **Figure 8E**). [153]. The PCM in the inorganic PCH directly influences the ambient temperature because it changes structurally during the phase transition and absorbs or releases a significant quantity of latent heat. In particular, the PCM absorbs heat from the surroundings during its transition from a solid to a liquid, causing the ambient temperature to drop to almost the PCM's T_{mp}. The temperature does not increase further until the phase change process is complete. Likewise, the PCM exotherms, raising the surrounding temperature, as it transforms from a liquid to a solid. The kind of PCM determines the temperature range that the PCH can control; hence, the right phase change material must be chosen for a given situation.

View Article Online
DOI: 10.1039/D6MA00167J

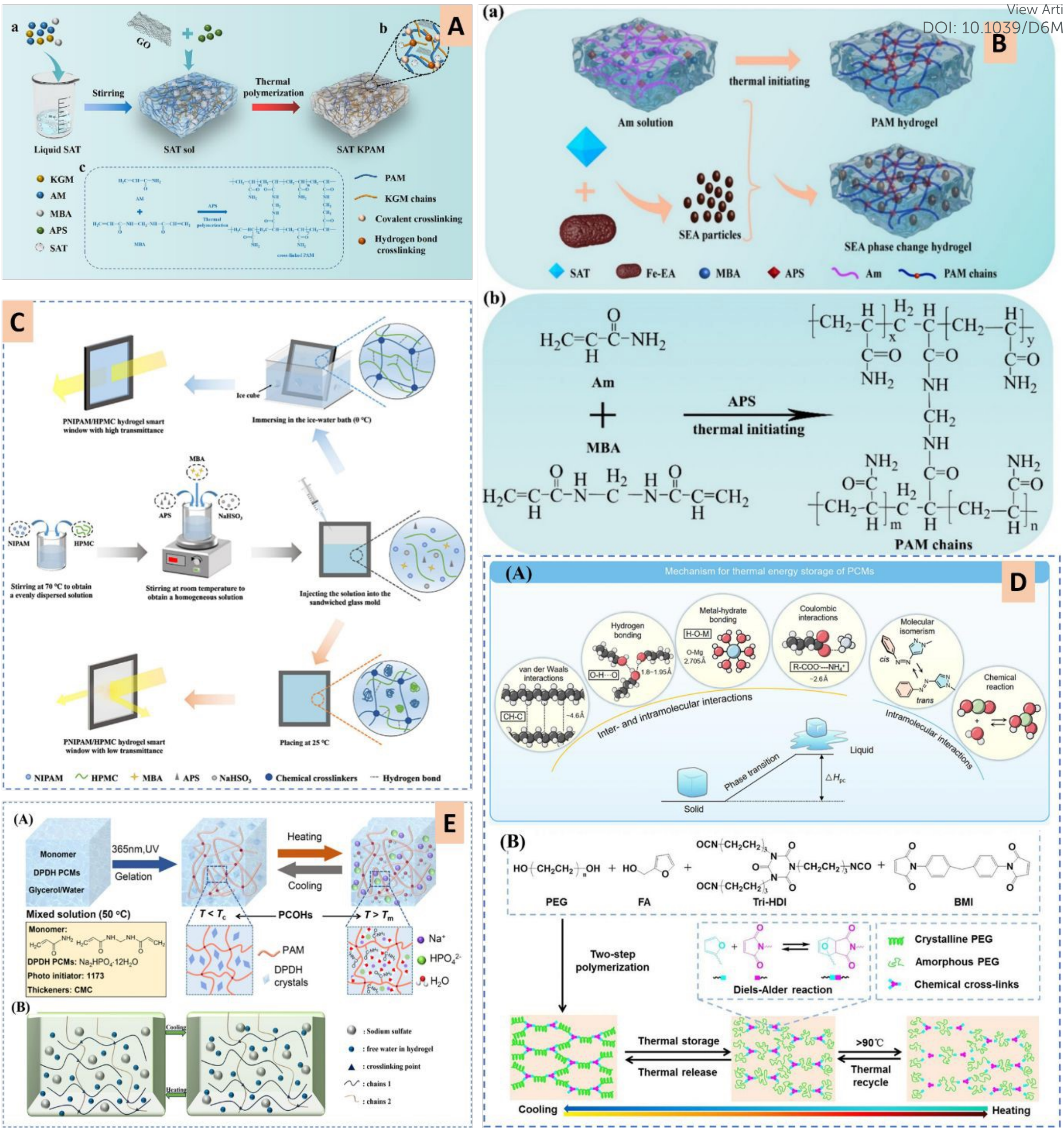


Figure 8: (A) (a) The SAT KPAM preparation schematic diagram. (a) Schematic depiction of a semi-interlaced network with a linear KGM chain interspersed with PAM forming a chemical crosslinking network. (c) The chemical processes of heat polymerization and APS started. Reproduced with permission from ref. [160]. Copyright 2023 Elsevier. **(B)** (a) The PAM/SEA phase change hydrogel fabrication procedure; (b) The chemical processes behind the production of PAM hydrogels. Reproduced with permission from ref. [152]. Copyright 2024 Elsevier. (C) Procedure for making PNIPAM/HPMC hydrogel and its mechanism. Reproduced with permission from Ref. [161]. Copyright 2024 American Chemical Society. **(D)** Mechanistic pathway of organic PCHs. **(E)** Phase change pathway for inorganic PCHs.

View Article Online
DOI: 10.1039/D6MA00167J

Reproduced with permission from ref. [30]. Copyright 2025 Elsevier.

PTHs and PCHs show great promise for real-world application, building on the understanding of their production and phase change processes. PTHs work effectively in applications that need dynamic flexibility because of their reversible structural reactions to environmental stimuli. However, PCHs can be employed in energy management systems due to their high latent heat storage and effective temperature control. The adjustable properties, stability and flexibility extend the usefulness of both materials. The main applications of PTHs and PCHs are summarized below, with a focus on their role in energy-related, environmental and biological technologies.

4.3 Sustainability considerations: biodegradability and green synthesis routes of PTHs and PCHs

Biodegradability and green synthesis routes of PTHs and PCHs: sustainability considerations. The biodegradability and ecologically safe production of phase-transition hydrogels (PTHs) and phase-change hydrogels (PCHs) supplemented with nanoparticles have received a lot of interest due to the increasing focus on sustainable materials design. The life-cycle effect, degradability, and synthesis methods of these materials must be in line with green chemistry and circular economy concepts as they move toward large-scale and practical applications, especially in the biomedical and environmental sectors. For hydrogels intended for transitory and biomedical applications, biodegradability is a crucial need [162,163]. Despite having adjustable mechanical and responsive qualities, conventional synthetic polymer networks often have poor degradability, which results in long-term persistence in biological or environmental systems. More and more efforts are being made to incorporate biodegradable polymers like proteins, synthetic biodegradable polyesters, and polysaccharides (like cellulose, chitosan, and alginate) [164,165]. These can be broken down in a controlled way, either in the body or in the environment, through hydrolytic or enzymatic reactions. In tissue engineering and drug delivery applications, biodegradable networks in PTHs can be engineered to be responsive and to gradually degrade in physiological environments. The challenge of PCHs is to achieve a balance between biodegradability, structural integrity and phase-change performance, as early degradation could compromise the PCM encapsulation and thermal stability.

The selection of nanoparticles is an important factor in influencing total degradability, in addition to the polymer matrix. Long-term toxicity and environmental persistence may be a concern due to the potential for the buildup of non-biodegradable inorganic nanoparticles over time. The use of biodegradable or bioresorbable nanofillers, such as silica derivatives, polymeric nanoparticles, and bio-based nanomaterials, that may break down into non-toxic byproducts, is thereby gaining popularity. Surface functionalization techniques can further enhance compatibility without compromising functional performance and can also allow for controlled deterioration [56]. Another crucial component of sustainable hydrogel design is green synthesis pathways. Scalability and environmental compatibility are limited by the use of hazardous solvents, high energy consumption, and multi-step processing in traditional manufacturing processes. The utilization of renewable feedstocks, solvent-free or aqueous-based synthesis, and energy-efficient processing methods have been the main areas of recent development. To lessen the effect on the environment, techniques such as enzymatic crosslinking, photo-initiated reactions under moderate temperatures, and in situ polymerization in water are being used more often. In addition, innovative production methods such as freeze-casting and 3D printing enable precise material structure and reduce waste generation, which is conducive to large-scale and sustainable production.

The construction of recyclable and reprocessable hydrogel systems also incorporates the concepts of green chemistry [166]. Material recovery and reuse through reversible crosslinking networks, dynamic covalent bonds and supramolecular interactions reduce waste and enhance resource efficiency. For energy storage applications, which require significant cycling of the materials, long-term material stability is extremely important, and recyclability is particularly critical for PCHs. The sustainable development of nanoparticle-reinforced PTHs and PCHs relies on the use of biodegradable components and green synthesis techniques in general [167]. In order to enable these cutting-edge materials to satisfy both technical and sustainability criteria, future research should concentrate on striking the ideal balance between performance, durability, and environmental friendliness.

4.4 Interfacial interactions governing phase behaviour in PTHs vs PCHs

The phase behaviour, structural development, and functional performance of nanocomposite hydrogels are largely determined by interfacial interactions. A range of non-covalent and dynamic interactions control the interaction between polymer chains, solvent molecules, and embedded nanostructures in phase-transition hydrogels (PTHs) and phase-change hydrogels (PCHs). Different phase development paths and energy storage mechanisms result from the substantial differences in the nature, dominance, and functional implications of these interactions between PTHs and PCHs. Therefore, to explain the variations in their phase behaviour and multifunctional capabilities, a comparative study of hydrogen bonding, hydrophobic interactions, π–π stacking, polymer–nanoparticle interfaces, and coordination-driven interactions is crucial.

4.4.1 Interactions with hydrogen bonds

One of the most basic interactions controlling PTHs' thermoresponsive behaviour is hydrogen bonding. Hydrogen bonding between polymer chains and surrounding water molecules is crucial in determining the lower critical solution temperature (LCST) behaviour in polymer networks like poly(N-isopropylacrylamide) (PNIPAM)-based hydrogels [168,169]. The inflated hydrogel network is stabilized below the LCST by strong hydrogen bonding between hydrophilic groups and water molecules. Disruption of these hydrogen bonds causes dehydration and polymer chain collapse when heated over the LCST, which results in a reversible volume phase transition [170].

In PCHs, on the other hand, hydrogen bonding has a comparatively minor yet structurally stabilizing function. Although some PCM-containing hydrogels use hydrogen bonding to anchor phase-change domains (such as paraffin derivatives or polyethylene glycol (PEG)) within the polymer matrix, latent heat related to solid–liquid transitions rather than hydrogen-bond-mediated swelling-deswelling controls the main energy storage mechanism [171,172]. Hydrogen bonding is not directly responsible for the phase change in these systems, but it does help to maintain the structural integrity and prevent leakage of PCM domains. While hydrogen bonding is a secondary stabilizing interaction that maintains network integrity during repeated melting–solidification cycles in PCHs, it is dynamic, reversible and directly responsible for the thermoresponsive phase changes in PTHs. This contrast shows how the polymer-solvent interactions have essentially different effects on the phase behaviour of both systems.

4.4.2 Phase segregation and hydrophobic interactions

Although they have quite distinct functional effects, hydrophobic interactions are a crucial driving factor in both PTHs and PCHs. Polymer chain aggregation above the LCST in PTHs is mostly caused by hydrophobic interactions. It is known that the balance between the hydrophilic and hydrophobic interactions is temperature dependent, favoring hydrophobic association of polymer backbones. This causes the hydrogel network to collapse and water

View Article Online
DOI: 10.1039/D6MA00167J

molecules to be expelled [173,174]. This reversible hydrophobic aggregation is mainly responsible for the thermoresponsive switching behaviour of PTHs.

However, latent heat storage events are tightly coupled to the hydrophobic interactions in PCHs. In the hydrogel matrix, the PCM elements undergo solid-to-liquid and liquid-to-solid phase changes, which are usually hydrophobic materials such as paraffins and fatty acids. Immiscibility of the hydrophobic PCM domains and hydrophilic polymer networks results in the formation of phase-separated structures at the micro- or nanoscale [175,176]. These domains act as thermal energy storage and release areas during melting and crystallization. Moreover, the degree of confinement and the compatibility of the interfaces between the PCM domains and the polymer matrix significantly affect the crystallization kinetics, supercooling behaviour and thermal cyclability [177]. Strong interactions at the interface can enhance thermal reliability and prevent leakage by promoting nanoscale confinement and preventing macroscopic phase separation.

Hydrophobic interactions consequently favor phase separation and latent heat energy storage in PCHs and reversible polymer collapse in PTHs. This contrast highlights the difference between enthalpy-driven phase changes in PCHs and entropy-driven conformational changes in PTHs.

4.4.3 Supramolecular interactions and π–π stacking

π–π stacking interactions between aromatic groups in polymer networks or nanofillers provide structural organization and functional improvement of nanocomposite hydrogels. In order to enhance the mechanical strength, electrical conductivity and stimuli responsiveness of PTHs, π–π interactions are frequently engineered with the nanofillers of graphene or aromatic comonomers[178,179]. In PCHs, these interactions can be reversible physical crosslinks that help to stabilize the network and dissipate energy during deformation, particularly when carbon-based nanomaterials such as graphene, carbon nanotubes, or biochar are employed. These nanostructures allow for efficient heat transfer during phase transitions of the PCM, as well as provide structural support by enhancing the thermal conductivity of the PCM [180]. In addition, π–π interactions between the PCM molecules and aromatic surfaces can affect the nucleation behaviour and crystallization dynamics. The π–π stacking interaction is more closely associated with the efficiency of phase transition and the enhancement of the heat transport properties in PCHs than it is with mechanical reinforcement and multifunctionality

View Article Online
DOI: 10.1039/D6MA00167J

in PTHs. Therefore, π-conjugated systems are present in both types of hydrogels to connect the mechanical and thermal properties.

4.4.4 Interfacial interactions between polymers and nanoparticles

Phase behaviour is greatly impacted by the intricate interfacial phenomena that are introduced when nanoparticles are added to hydrogel networks. The interactions between nanoparticles such as silica, clay or metal oxides, and the polymer chains in PTHs can occur through hydrogen bonding, electrostatic interactions or van der Waals forces [181,182]. These interactions can change the stiffness, swelling behavior and responsiveness of the network, as they are multipurpose crosslinking points.

To enhance heat conductivity and to stabilize the domains of PCM in PCHs, polymer–nanoparticle interactions are crucial. During phase transitions, nanoparticles like graphene, boron nitride, and metallic nanostructures produce thermally conductive channels that speed up heat transfer [183]. Furthermore, the surface functionalization of nanoparticles may enhance their compatibility with PCM and polymer phases, thereby lowering interfacial resistance and improving cycle stability. Nanoscale confinement caused by nanoparticles also influence the behaviour of crystallization in PCHs. Improved phase transition reversibility, decreased supercooling, and changed melting temperatures are often seen in confined PCM domains [9,183]. In PTHs, where phase transitions are primarily controlled by polymer-solvent interactions rather than restricted crystallization, these effects are less noticeable. Therefore, although polymer-nanoparticle interactions are essential for temperature management and phase transition efficiency in PCHs, they mainly affect mechanical and swelling characteristics in PTHs.

4.4.5 Dynamic crosslinking interactions and coordination

A new class of interfacial interactions that provide hydrogel systems flexibility and multifunctionality include coordination interactions and dynamic crosslinking processes. Metal–ligand coordination bonds may act as reversible crosslinks in PTHs, allowing for self-healing, stimuli-responsive behaviour, and mechanical characteristics that can be adjusted [184]. Hydrogels may react to external stimuli like temperature, pH, or ionic strength thanks to these interactions, which bring dynamic network rearrangement possibilities.

Coordination interactions are widely used in PCHs to improve structural resilience over repeated heat cycling and stabilize PCM domains. PCM molecules may be anchored inside the

View Article Online
DOI: 10.1039/D6MA00167J

polymer network by metal–ligand interactions, which improves shape stability and stops leakage [185]. Coordination networks may also increase mechanical robustness under cyclic thermal loads and enhance energy dissipation. However, while coordination interactions in PCHs can help maintain structural stability and long-term durability under phase-change conditions, they are primarily associated with dynamic responsiveness and network flexibility in PTHs.

4.4.6 A comparative view of interfacial interactions

Different mechanistic concepts controlling the phase behaviour of PTHs and PCHs are revealed by a direct comparison of their interfacial interactions. PCHs, on the other hand, depend on phase segregation and latent heat storage linked to PCM domain solid-liquid transitions. In these systems, interfacial interactions are intended to improve thermal conductivity, stabilize phase-separated structures, and guarantee effective energy storage and release. The regulating mechanisms rely on the thermodynamics of phase transitions and are mostly enthalpy-driven. Furthermore, PCHs are intended for long-term energy storage and temperature control, while PTHs show quick and reversible transitions appropriate for sensing, actuation, and drug administration applications. Thus, in PCHs, the function of interfacial contacts changes from regulating phase interfaces and heat transfer channels to controlling polymer conformation in PTHs. Similar interfacial interactions serve essentially distinct purposes in PTHs and PCHs, as shown schematically in **Figure 9**. In PTHs, reversible conformational switching, swelling-deswelling behaviour, and stimulus response are primarily controlled by interactions such as hydrogen bonding, hydrophobic association, π–π stacking, and dynamic coordination. Adaptive network rearrangements in response to external stimuli like temperature, pH, or light are governed by these interactions. The same interactions, on the other hand, mainly support PCM confinement, leakage prevention, thermal conductivity increase, and latent heat storage domain stability in PCHs. Therefore, interfacial interactions primarily assist structural stability and thermal energy management in PCHs, while they are primarily linked to dynamic responsiveness in PTHs.

View Article Online
DOI: 10.1039/D6MA00167J

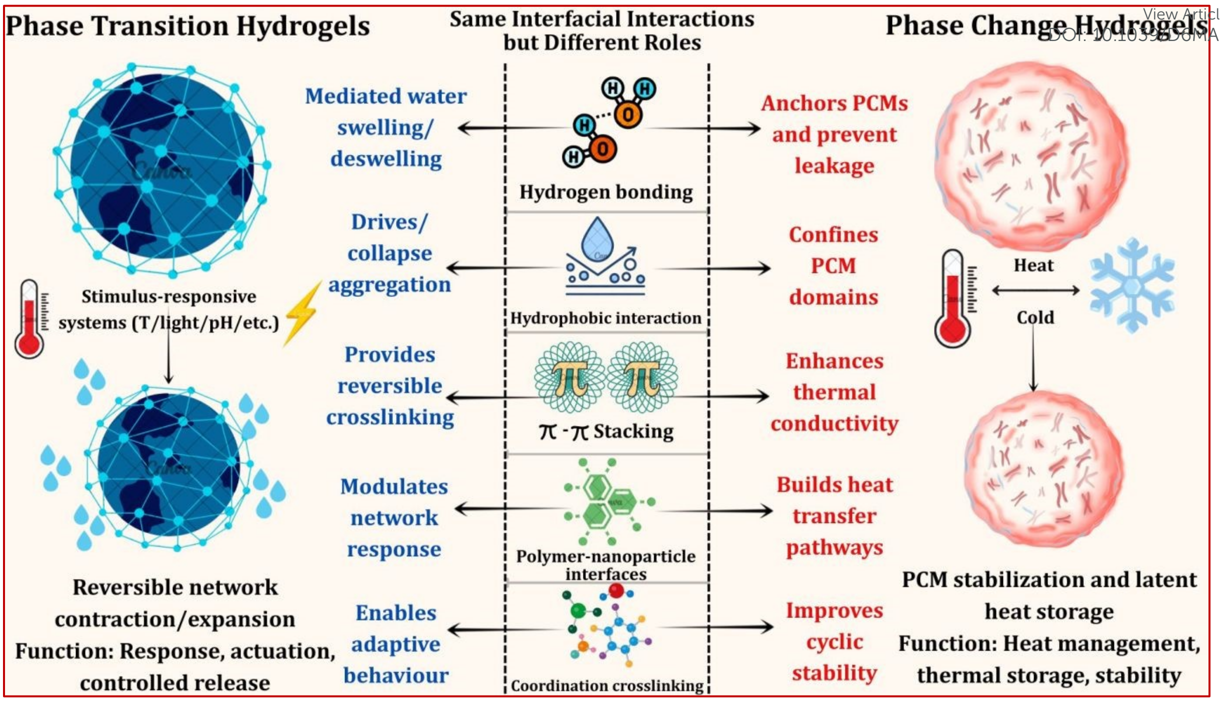


Figure 9: Diagrammatic representation contrasting the different functions of comparable interfacial interactions in phase-change hydrogels (PCHs) and phase-transition hydrogels (PTHs). Interfacial interactions in PTHs mainly control stimuli-responsive behaviour and reversible network expansion/contraction, whereas in PCHs they stabilize PCM domains, increase thermal conductivity, and improve latent heat storage and thermal cycle stability.

4.4.7 Implications for design

The rational design of next-generation multifunctional hydrogels benefits greatly from an understanding of the divergent functions of interfacial contacts in PTHs and PCHs. For PTHs, precise control over transition temperatures and responsiveness is made possible by adjusting hydrophobic balance and hydrogen bonding. For PCHs to maximize heat storage capacity, conductivity, and cycle stability, polymer-PCM and nanoparticle interface engineering is essential. These interfacial engineering techniques have great potential to advance hydrogel-based systems in soft electrical, biological, and energy applications.

5. Multifunctional applications

The ability of phase transition and phase change hydrogels to regulate temperature first led to their use in energy storage and thermal management. These hydrogels' uses have grown as a result of developments in materials science, flexible electronics, biomedicine, optics, and nanotechnology. They are extremely adaptable for use in soft robotics, healthcare, flexible bioelectronics, and optics due to their superior biocompatibility, chemical and mechanical stability, and reversible phase transition properties in response to stimuli like temperature, light, pH, magnetic fields, and electric fields.

5.1 Applications in medicine

PTHs and PCHs are becoming more and more popular in the medical profession for uses including controlled photothermal therapy, bacteriostatic skin dressings, and medication administration for conditions like cancer because of their reversible phase transition and change characteristics and high biocompatibility.

5.1.1 Systems for drug delivery

Drug delivery systems based on PTHs and PCHs may precisely regulate drug release by changing their physical state in response to external stimuli. When the drug molecules reach the desired area, external stimuli like temperature, pH, NIR light, magnetic fields, or ultrasound may cause phase transitions that control the location and rate of drug release. The drug molecules are contained inside the three-dimensional network of the hydrogel. The hydrogel's chemical stability and biocompatibility reduce tissue irritation and improve the medication's therapeutic effect [186,187].

Phase transition hydrogel drug delivery systems are optimized primarily in the areas of drug loading capacity enhancement, drug release rate tunability, gel network structure stability, and targeted delivery realization. The conjugated polymer skeleton securely attaches the photoactive drug via π–π stacking interactions, preventing premature leakage. Liu et al. [188] incorporated the NIR contrast agent, ICG, into a thermo-sensitive hydrogel based on a photo–thermal network. Phase transition hydrogel drug delivery devices may be adaptively assembled in the human body via pH responsiveness. An injectable, in situ self-assembled pH-responsive drug peptide hydrogel was created by Zhang et al. [189]. In a slightly acidic environment (pH 6.5), the small-molecule medication methotrexate was able to undergo a spontaneous gel-to-solution transition after binding to the KKFKFEFEF peptide chain through an amidation reaction. Chen et al. [142] created a magneto–thermal hydrogel swarm based on magnetic hydrogel particles for the targeted administration of medications for gastric ulcers in pigs to accomplish exact targeted delivery (**Figure 10a**) [190]. Agarose, neodymium–iron–boron (NdFeB) and Fe_3O_4 magnetic cores, and sodium caseinate made up the magnetic hydrogel. When a high-frequency AMF was applied, the magneto–thermal effect caused the hydrogel to quickly heat up and go through a solid–liquid phase transition (**Figure 10b**). At the same time,

View Article Online
DOI: 10.1039/D6MA00167J

the magnetic hot water gel cluster movement's direction and speed could be efficiently controlled by the application of an external magnetic field (**Figure 10c**). By balancing controlled release performance, targeting, and gel network stability, the aforementioned optimization may encourage a broader use of phase transition hydrogels in the drug delivery industry.

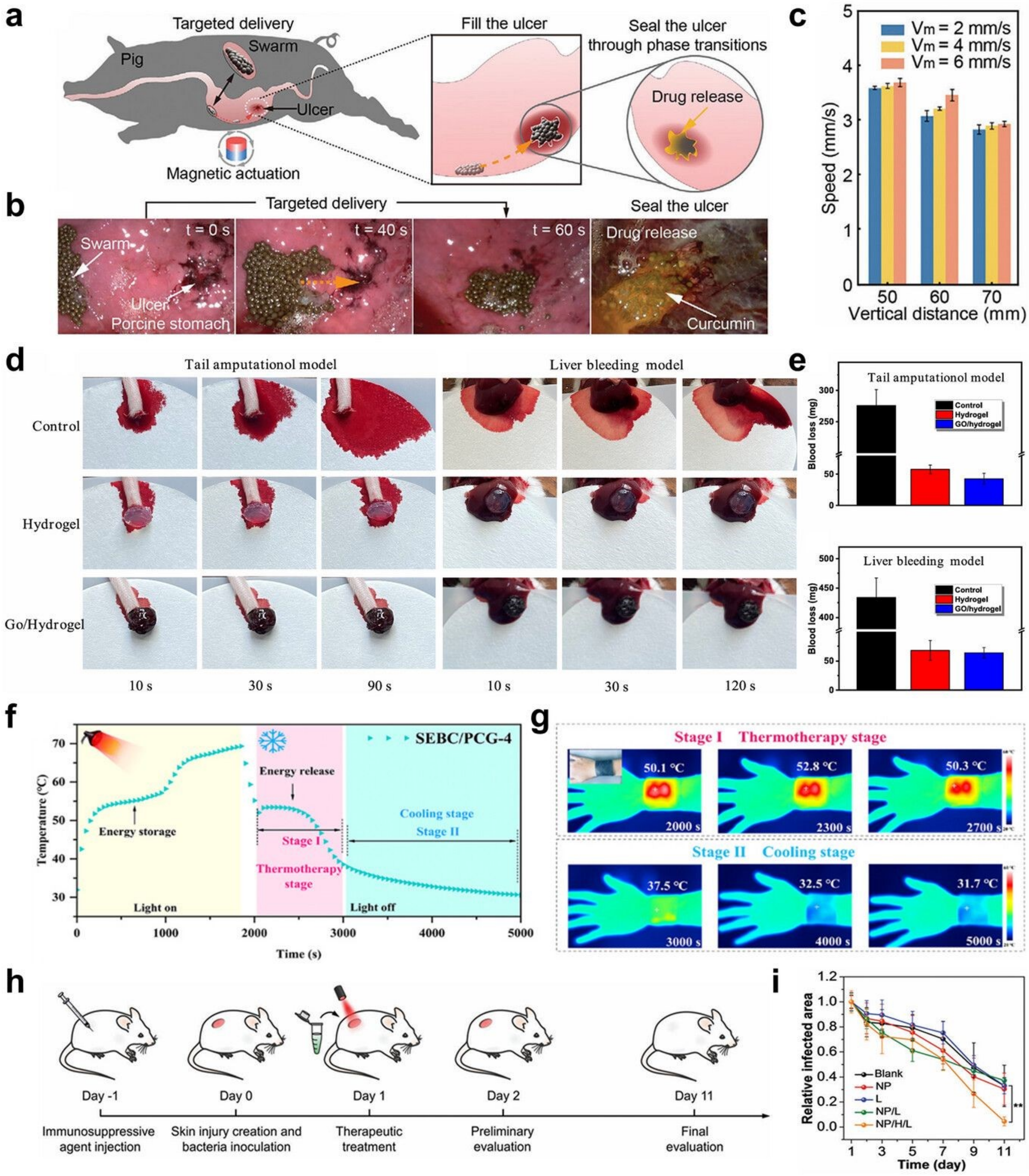


Figure 10: a) Diagrammatic representation of the in vivo porcine stomach gastric ulcer sealing experiment. b) The outcomes of an in vivo pig stomach experiment using hydrogel swarms for targeted distribution and ulcer filling. c) As the magnet travels at different speeds, the hydrogel

View Article Online
DOI: 10.1039/D6MA00167J

swarms on the stomach tissue at various vertical distances. d) The hydrogel's hemostatic property. Pictures of the liver hemorrhage and tail amputation models. e) The loss of blood in the liver hemorrhage and tail amputation models. d,e) Permitted reproduction. f) Time-temperature graphs and g) Infrared thermography pictures of SEBC/PCG-4 hydrogel composites used for wrist joint thermotherapy, powered by a physiotherapy light. h) MeO-TSI@F127 NPs' in vivo antibacterial properties under NIR irradiation with P(NIPAM-AM) hydrogel-enabled temperature control. Diagrammatic representation of the evaluation period's timeframe. i) Mice's relative infected regions with various treatments. Reproduced with permission from ref. [7]. Copyright 2025 Wiley. (Open Access).

The main quantitative performance metrics of typical phase-transition hydrogel drug delivery systems are compiled in **Table 4**. The comparison shows how thermo, pH, and magneto-responsive platforms vary in terms of stimulation conditions, release mechanisms, and biological effects. Notably, specific metrics like encapsulation efficiency and exact release percentages are still not regularly accessible, even though factors like irradiation power, pH-triggered transitions, and structural properties are frequently published. A more precise framework for assessing the effectiveness and suitability of various stimuli-responsive hydrogel systems is provided by this comparison study.

Table 4: Comparing the biological performance, release behaviour, and stimulus conditions of typical stimuli-responsive phase-transition hydrogel drug delivery systems quantitatively.

Study	**System**	**Drug/ Payload**	**Quantitative Loading/ Structure**	**Stimulus Conditions**	**Quantitative Release/ Transition**	**In Vitro Data**	**In Vivo Data**
Liu et al. [188]	Thermosensitive PNT hydrogel	ICG	ICG concentration: **5 µg/mL** (photothermal), **45 µg/mL** (cell uptake)	NIR: **808 nm**, **0.28 W/cm²** (release), **0.14 W/cm²** (therapy)	Gel–sol conversion quantified: Conversion % = (1 − remaining mass / initial mass) × 100; ON–OFF	Strong 4T1 cell killing; ROS generation; enhanced uptake after irradiation	**Near-complete tumor eradication** after single intratumoral injection

View Article Online
DOI: 10.1039/D6MA00167J

					release under pulsed NIR		
Zhang et al. [189]	pH-responsive peptide hydrogel	Methotrexate (MTX)	pH-sensitive conjugation; charge shift measured (zeta potential change)	pH trigger: **7.4 → 6.5** (tumor environment)	Sol–gel transition at pH 6.5; charge conversion improves uptake	Cell viability: **~100% for carrier (no toxicity)**; MTX systems show concentration-dependent cytotoxicity	Improved tumor inhibition; long retention; negligible side effects
Chen et al. [142]	Magneto-photothermal hydrogel (PNIPAm–MoS_2/Fe_3O_4)	Dye/ model payload	MoS_2 size: **0.4–0.8 µm**; thickness: **1.8 ± 0.2 nm → 5.0 ± 0.4 nm**	NIR + magnetic field; orientation-dependent photothermal response	Localized phase transition via photothermal effect; controlled release behavior (no % data)	Controlled dye release; orientation-dependent efficiency	Demonstrated programmable response; potential therapeutic application

5.1.2 Skin care products

Phase transition hydrogel skin dressings, which can control the moist environment, prevent the development of infections, and speed wound regeneration, are innovative trauma therapeutic materials that are often utilized in medical specialties, including burn therapy and skin restoration [191]. Phase transition hydrogels may absorb wound exudates to maintain the right humidity levels and avoid ulcers while creating a protective, moist environment close to the site when hydrated. The phase transition hydrogels have the potential to self-heal thanks to dynamic networks of covalent and hydrogen bonds, which help to ensure the long-term durability of skin dressings. Based on the dual-network structure of carboxymethyl chitosan-oxidized SA/Agar/MXene (CMCS-OSA/AG/MXene), Ma et al. [192] developed an injectable hydrogel that, when exposed to external electrical stimulation, not only quickly eliminated wound exudates but also encouraged cell proliferation, repair, and accelerated wound healing because of its superior electrical conductivity.

Phase transition hydrogels' reversible phase transition characteristics allow them to adapt well to wounds, improving comfort, and the addition of photothermal dyes and medications for on-demand drug administration and photothermally improved antibacterial treatment. Chen et al. [193] added graphene oxide nanoparticles to the thermoresponsive copolymer PNIPAM and dopamine-functionalized pectin hydrazide to create a photothermal-enhanced skin dressing that included vancomycin. Through the photothermal action, the hydrogel successfully prevented the development of wound bacteria, and the medication that was released encouraged wound healing. Its superior hemostatic and skin adhesion qualities further improved its efficacy as a skin dressing (**Figure 10d**). Mice's bleeding was considerably decreased by around 85% in the tail amputation and hepatic haemorrhage paradigm (**Figure 10e**). The usefulness of phase transition hydrogel skin dressings may be further increased by optimizing biocompatible natural polymers and biofunctional changes, such as antibacterial and anti-inflammatory ones.

5.1.3 Devices for photothermal therapy

To cure damaged tissues, photothermal therapy (PTT) uses laser irradiation, localized heat to increase blood flow, or membrane rupture and protein denaturation of diseased cells. PTT based on phase transition hydrogel has the benefits of great controllability, robust targeting, and adaptability. The secret to PTT is photothermal conversion efficiency. Li et al. [194] created a phase transition hydrogel with a high melting point enthalpy (119.7 J g^{-1}) and an outstanding photothermal conversion efficiency (92.2%) by loading SEBC composites into PAM/chitosan/GNP bi-networked hydrogels (**Figure 10f**). To prevent overstimulation of the joints by direct exposure to the physiotherapy lamp, the phase transition hydrogel's high photothermal conversion and melting point enthalpy allow it to warm up quickly during the preheating stage and release latent heat continuously and steadily during the thermotherapy stage (**Figure 10g**). Fu et al. [195] created a thermostatic PTT system based on P(NIPAM-AM) thermo-responsive hydrogel to address the issue of uncontrolled heat damage to healthy tissues and organs in conventional PTT. The majority of the heat produced by NIR light irradiation was absorbed by the phase transition hydrogel, which causes a rapid phase transition to stop

View Article Online
DOI: 10.1039/D6MA00167J

heat penetration and greatly lessens the high-temperature stimulation of tissues and organs. Mice have been used to verify the PTT system, and by setting the equilibrium temperature T_{cp} to 49 °C to guarantee sterilizing effectiveness and prevent thermal damage (**Figure 10h**), the comparatively infected area's pace of healing was markedly accelerated (**Figure 10i**). The usefulness of phase transition hydrogel-based PTT in photothermal synergistic treatment is shown by these investigations.

5.2 Wearable sensors and electrodes

The ability to track body temperature, humidity, joint movements, breathing, and sleep patterns in real-time, as well as help aphasia patients with voice rehabilitation, has made phase transition hydrogel-based flexible wearable sensors a hot topic in medical diagnostics, environmental monitoring, and health monitoring in recent years [196,197]. Adjustability, high sensitivity, low power consumption, wearability, and biocompatibility are some benefits of phase transition hydrogels, which are induced to vary their conductivity and swelling rate by external stimuli, including temperature, humidity, strain, and bioelectric signals.

Using the effects of temperature on carrier mobility, the swelling state of the system, and optical transparency, phase transition hydrogel temperature sensors provide optical or electrical signals. An Agar/PSBMA hybrid hydrogel-based wearable temperature sensor was suggested by Yang et al. that can measure temperature on its own across a broad range of 10–40 °C [198]. As the temperature rose, this phase transition hydrogel showed clear UCST behaviour, with reversible changes in optical transparency. The temperature was directly converted into electrical signals, which made signal processing easier and increased interference resistance. P(HPA-co-AM)/HPC hydrogel was used by Zhou et al. [199] to create a wearable sensor for human mobility and health monitoring, which could be combined with hardware circuits to achieve the fever alarm function. Temperature coefficient of resistance (TCR) values for the sensor were −0.89%, 0.59%, and 0.39% in the 20–30, 30–50, and 50–72 °C temperature ranges, respectively, indicating a negative reaction to temperature. Its minimal detection limit for body temperature was as low as 0.1 °C, and it also showed outstanding real-time detection capabilities. Because of their advantageous flexibility, wearable strain sensors based on phase transition hydrogels can detect motions of human joints. This improves skin contact, lowers motion artefacts, and increases the device's signal-to-noise ratio. A CMC-P(AA-AM)-Al^{3+} hydrogel with high sensitivity and quick motion detection responsiveness was suggested by Chen et al. [200], allowing for real-time finger and leg bending angle monitoring. The hydrogel's unique composition has excellent mechanical properties, high conductivity and swelling behavior to track dynamic body movements in real-time. Moreover, it has an excellent biocompatibility, which enhances its application potential in wearable motion sensors for health care.

Huang et al. [65] developed a strain sensor and photothermal therapy (PTT) based on a single material, a phase change gel (PCG), which was fabricated using sodium sulfate decahydrate (Na2SO4·10H2O), polyacrylamide (PAM) and polydopamine (PDA) modified MXene (MXene). The PCGs have a high enthalpy value (128.8 J/g) and a suitable phase transition temperature that offers sufficient thermal comfort. The PAM hydrogel was effective in preventing the liquid leakage from the SSD and also gave flexibility. In addition to giving the PCGs a high photothermal conversion efficiency (93.83%), PDA@MXene guaranteed robust adhesive qualities for accurate motion monitoring. It is noteworthy that the PCGs showed that they could exhibit temperature-sensitive adhesion properties by triggering the SSD phase transition, which led to both high skin adhesion and non-painful removal at the same time. There are several potential uses for temperature-induced adhesive PCGs in portable medical equipment (**Figure 11A**).

With its exceptional flexibility and adjustable electrical conductivity, flexible gel electrodes, which are made up of a flexible substrate and gel electrolyte, find extensive use in biomedical applications, wearable technology, and flexible sensors [201] Another significant advantage of gel electrode is the excellent biocompatibility, which reduces the pressure and friction on the surface, reduces allergic reactions, and makes the wearing more comfortable compared with the stiff electrode made of Ag, Au or Pt. Further, the electrochemical characteristics of gel electrodes can be optimized by changing the composition and structure of the gel, thus enabling them to be used for various applications. Traditionally, however, gel electrodes are not flexible and tend to have a fixed shape and function. This limits their applications in areas where they need to be responsive and dynamic, such as smart wearable technology and adaptive medical monitoring.

The next generation of wearable electronics for Internet-of-things systems, health monitoring, "interface-on-invisible," and green energy harvesting needs electrically conductive materials that are extremely stretchy, industrially viable, negligibly hysteretic, and superior in transparency. Due to relative delamination and viscoelastic performance, the ionic hydrogel's practical potential is hindered by evident hysteresis and a restricted sensing range. A new liquid conductor, called egg white liquid, was created by self-liquidating egg white hydrogel [202]. It showed ultrahigh transparency (up to 99.8%) and conductivity (20.4 S m^{-1}) that was

View Article Online
DOI: 10.1039/D6MA00167J

comparable to that of ionic hydrogel, in addition to inheriting the hydrogel predecessor's designed architecture. Additionally, the 3D-printed liquid-elastomer hybrid exhibited outstanding sensitivity with very little hysteresis (0.77%), outstanding conformability, and the ability to track dynamic moduli and human movements. A gesture-controlled touchless user interface for front-end electrical devices is inspired by the liquid nature. Furthermore, by using this liquid conductor to create a triboelectric nanogenerator, mechanical energy harvesting and pressure sensing are shown. Notably, a new type of wearable electronics, human–machine interface, and renewable energy may be fueled by the as-prepared liquid via subsequent phase change, which has better transparency, ultralow hysteresis, economic advantage, and a unique liquid phase. The egg white–Ecoflex (EWL–Eco) sensor was shown to have dual electromechanical sensitivity to tensile strain and pressure, high flexibility, and low hysteresis for a variety of applications (**Figure 11B a**). It successfully differentiated between wrist motion via perpendicular sensor placement (**Figure 11B c**), finger bending (**Figure 11B b**), and nonplanar geometries such as artificial blood vessel expansion and balloon inflation (**Figure 11B d**). Through Morse code signalling, the gadget made it possible to interface between humans and electronics (**Figure 11B e**). Before and after exercise, wrist pulse analysis was used to confirm physiological monitoring, demonstrating variations in heart rate, pulse amplitude, augmentation index (AIr), and pulse transit time (PTT) (**Figure 11B f**). **Figure 11B g** showed the detection of forehead muscle contractions, **Figure 11B h** showed noncontact vibration sensing, and **Figure 11B i, j** showed the identification of friction and roughness across glass, paper, and sandpaper. Ultimately, by combining light-emitting diodes (LEDs) on Ecoflex with egg white hydrogel (EWH) conductive traces, a three-dimensional (3D) visual pressure sensor was created that displays spatially resolved pressure feedback by selective dimming (**Figure 11B k**).

View Article Online
DOI: 10.1039/D6MA00167J

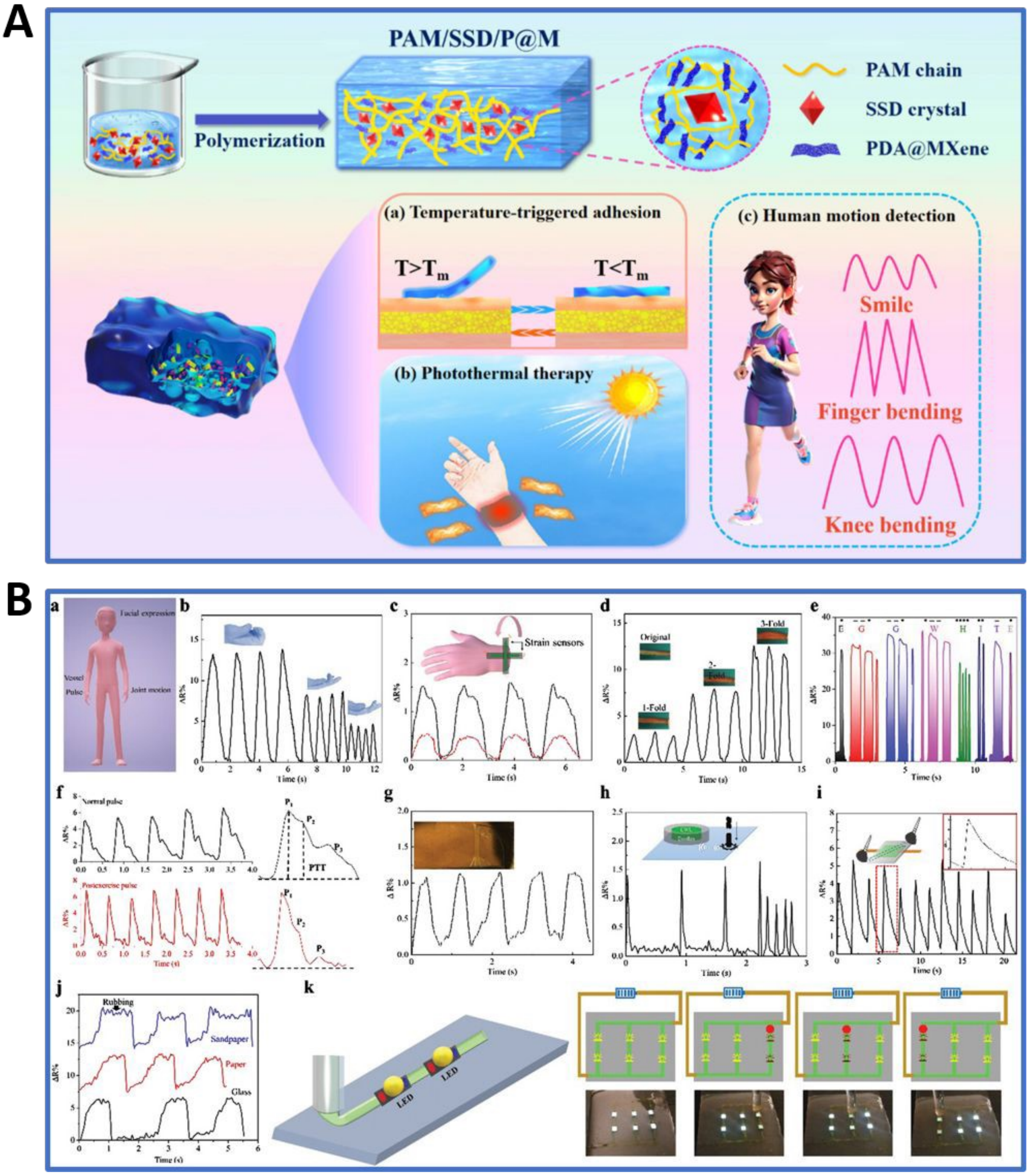


Figure 11: (A) Synthesis and various applications: ((a) Temperature-induced adhesion. (b) Photothermal therapy. (c) Human motion detection.) of PAM/SSD/P@M phase change hydrogel. Reproduced with permission from ref. [65]. Copyright 2025 Elsevier. **(B)** Low hysteresis and very dependable the EWL-Eco sensor detects a range of subtle and strong activities in the actual environment. a) Human motion sensing schematic. b) Variations in relative resistance to finger bending angles. c) Using biaxial fibrous sensors to capture wrist motion. d) Illustration of the lateral pressure exerted on the vessel-like tube by injecting water. e) A human-electronic interaction interface that uses a click-and-hold gesture on the sensor

surface to identify the Morse code for "EGG WHITE." f) The EWL-Eco sensor monitors wrist pulses and determines health status for inactive and postexercise settings. g) Motion detection of forehead wrinkles. h) The EWL–Eco sensor detects sound vibration in a noncontact mode. i) Shear force evaluation using a soft brush; inset: typical resistance change when the sensor surface is gently moved. j) How the EWL-Eco sensor reacts to various rough surfaces in terms of resistance. k) Diagram and images of a visual pressure sensor array using 3D printing EWL trace to detect the precise location of an external force; pressure was indicated by red dots. Reproduced with permission from ref. [202]. Copyright 2020 Wiley.

5.3 Soft robotics and optical applications

Compared to conventional rigid robots, soft robotics provides superior flexibility, adaptability, and safety, which makes them more appropriate for complicated situations including automated production, human–robotic interaction, environmental monitoring, and search and rescue [203]. Phase transition hydrogel-based soft robots may change their shape in response to external inputs, allowing them to carry out precise movements like walking and grabbing [204]. Because the laminated UCST-type gel layer and the LCST-type gel layer have opposite responsiveness to temperature changes, they can bend in different directions in response to heat. This is because the motion of soft robotics primarily depends on the differences in thermoresponsivity between various components. Soft robotics with various motion patterns under heat may be produced by varying their thicknesses and ratios. Hydrogel soft robotics requires fast bending and recovery properties. Li et al. combined an (NIPAM)-Laponite nano composite layer with LCST behaviour and a (NAGA) layer with UCST behaviour to simultaneously achieve a large bending angle range and fast thermally responsive bending and recovery capabilities [205]. The UCST and LCST layers showed entirely different expansion-contraction behaviour when the temperature was changed. This led to the hydrogel's reversible large-angle bending (+360° to −360°) in a brief amount of time (190 s), which made it the perfect carrier for ensnaring and moving small target objects. A wider variety of applications is available for hydrogel-based soft robotics that can operate in nonaqueous settings. The movement of a mimosa leaf served as the inspiration for Zheng et al.'s [206] hydrogel brake, which worked well in open air, water, and oil. Water molecules shifted between the PNIPAM layer (LCST-type) and the P(AAc-co-AAm) layer (UCST-type) in response to temperature changes, changing their dissolved states and enabling the hydrogel to retain or liberate target items. Another interesting technique for clamping objects is to alter the hydrogel's inherent stiffness. With the help of temperature regulation, Zhao et al. developed a

View Article Online
DOI: 10.1039/D6MA00167J

phase transition hydrogel soft robot based on PAA-PEG that has changeable stiffness and form. This ensures a dependable clamping force [207]. The outstanding contact that this hydrogel-based clamping system maintained with objects of different forms further increased its potential for a variety of applications.

The capacity of poly (N-isopropylacrylamide) (NIPAM) to reversibly swell and contract in response to temperature changes makes it one of the most popular hydrogels in soft robotics. When heated over its lower critical solution temperature, it expels water, reducing its volume by up to 90%. Not all hydrogels, in contrast to NIPAM, go from a hydrated, swollen polymer to a dehydrated, shrunken state when the temperature rises. Methylcellulose, a biocompatible hydrogel, for example, behaves in the opposite way, swelling when the temperature rises from a dehydrated condition [208]. Therefore, the chemistry, structure, and interaction of the polymer with water determine how a hydrogel reacts to external stimuli [209].

Choi et al. [210] showed that composite fibers made of a thermosensitive hydrogel and a shape memory polymer might be arranged to function as tensile, torsional, or flexural actuators. Additionally, the same fiber might be reprogrammed into a new kind of actuator by thermally erasing the preset mode of actuation. Poly(N-isopropylacrylamide) and polycaprolactone were used to create the fibers, and the fiber composition was adjusted to maximize the degree of actuation and form fixity. Heating the fibers to 60 °C and then cooling them under tensile, flexural, or torsional strain is one way to program them. After that, the fiber was conditioned by submerging it in room-temperature water, which caused the hydrogel phase to expand and distort its shape. The fiber was then heated at 35 °C in water to de-swell the thermosensitive poly(N-isopropylacrylamide) hydrogel, restoring the fiber's pre-programmed form. Several cycles of heating and cooling were used to demonstrate reversible actuation. Tensile strokes of up to 92%, bending curvature changes of 0.4 mm^{-1}, and torsional strokes of 10 turn/m were produced by the improved fibers. For soft robotics, the composite fibers provide a practical way to create a range of actuation kinds.

Phase transition hydrogels are frequently used in industries including information storage, encryption, and smart glass technology because of their transparency, light-absorbing qualities, and sensitivity to external stimuli like temperature, pH, and magnetic fields. Intrinsic storage and light-blocking storage are two categories for phase transition hydrogel-based information storage and decryption devices. Information is stored by the intrinsic storage type hydrogel, which modifies the system's composition and pH at certain points. The optical characteristics at these sites drastically alter in comparison to the backdrop when subjected to outside stimuli, allowing for the encryption and decryption of data [211]. The light-blocking

View Article Online
DOI: 10.1039/D6MA00167J

storage-type hydrogel, on the other hand, maintains consistent overall characteristics. Information is stored and decrypted by altering the hydrogel's overall transparency in response to outside stimuli.

Thermal responsiveness characterizes the great majority of the most popular and suggested phase transition hydrogel information storage methods. A phase transition hydrogel with LCST response was created by Zhou et al. using P(HPA-co-AM)/HPC/LiCl. When the temperature rose over the LCST, the hydrogel transitioned from a transparent to a turbid state [212]. The LCST of each pixel may be independently controlled by filling various locations on the display matrix with varying ratios of hydrogel precursor liquids, producing particular information at low temperatures and total turbidity at high temperatures. Yu et al. [213] created a bi-directional optically responsive hydrogel based on sodium dodecyl sulfate/potassium tartrate micelles and PNIPAM because shaded storage-type systems are generally more secure. They then combined it with a QR code to encapsulate it in a sandwich-style encrypted digital label. Only when the temperature fell within a certain range was the data shown. Based on this, a low-cost smart temperature monitoring system was created that made it possible to intuitively acquire an early warning by using the change in transparency when the temperature is beyond the predetermined range. Chen et al.[214] suggested magnetic field-responsive phase transition hydrogel information storage devices, in which thermal PNIPAM and magnetic Fe_3O_4 nanoparticles were inserted in hydrogel cubes to create stimulus-responsive units and attached to the surface of MoS_2 nanosheets. Magnetic field-assisted QR code programming was made achievable by the local control of the thermally induced phase transition of a single hydrogel cube with the application of a magnetic field and NIR light irradiation.

Stiff microchips, stretchy electrical interconnects, and flexible substrates must be robustly integrated to advance soft electronics. Conventional rigid-island designs move stress concentrations to island-substrate interfaces while reducing stress at chip-conductor connections. A gradient stiffness-programmed printed circuit board (GS-PCB) made of laser-patterned liquid metal conductors and a phase-transition-mediated hydrogel substrate was created by Kim et al. [215] to get around this. Strong soft-rigid adhesion and the introduction of spatial stiffness gradients during NaAc crystallization are provided by the hydrogel enhanced by polymer-sodium acetate (NaAc) interactions. The hydrogel is initially soft and homogeneous, as seen in **Figure 12a-i**; but, when it undergoes a phase transition, spatially variable stiffness appears (**Figure 12a-ii**). A stiffness gradient with high-stiffness areas in navy and low-stiffness areas in sky-blue is created when stretched, as rigid islands with attached chips stay unaltered while the soft areas around them extend (**Figure 12a-iii**). In contrast to

View Article Online
DOI: 10.1039/D6MA00167J

traditional rigid-island systems, this gradient transition reduces delamination and improves electromechanical stability (**Figure 12a-v**). By creating a finger-sensor device that could distinguish between objects under various mechanical disturbances, the method was verified.

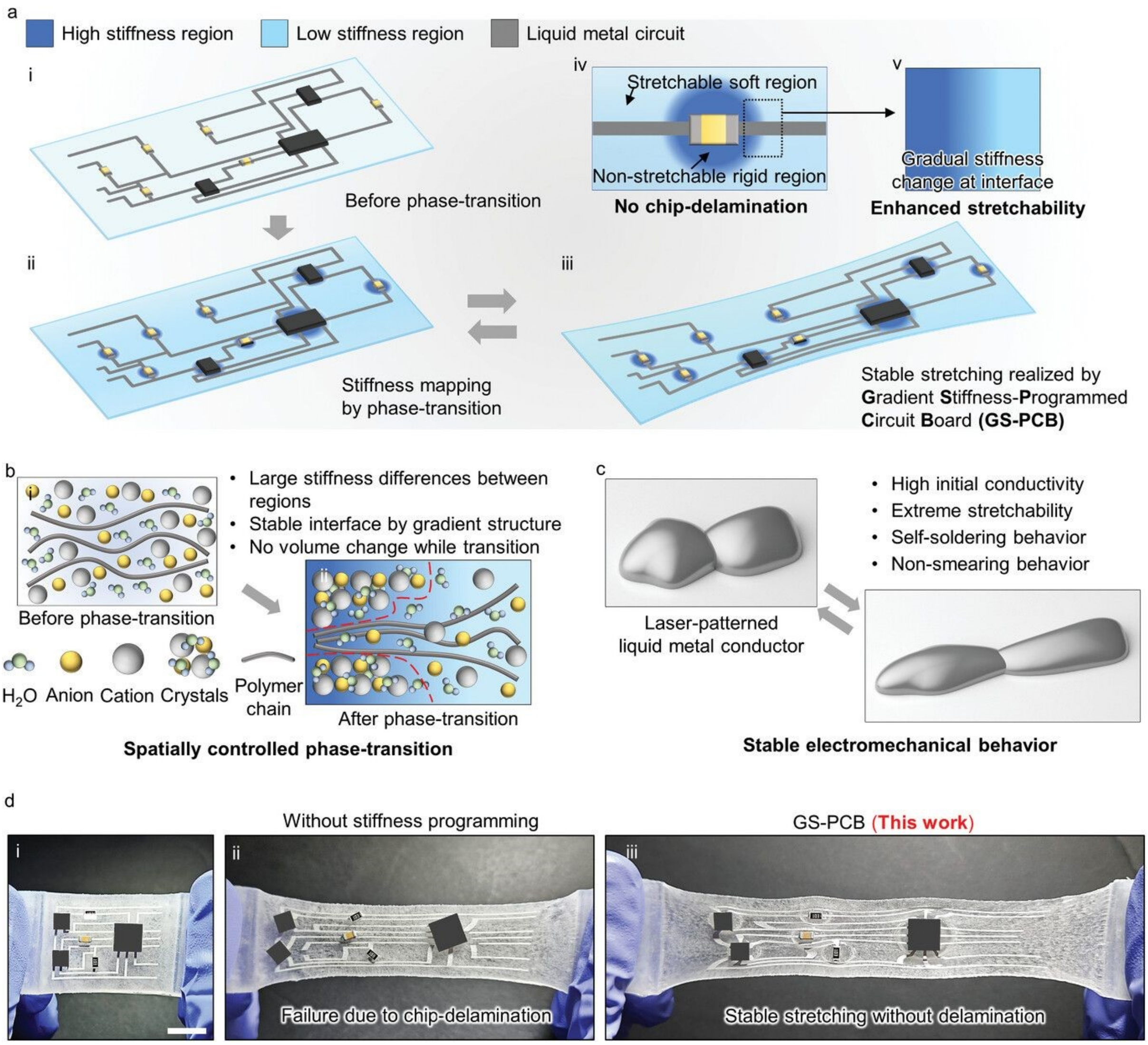


Figure 12: The characteristics of a gradient stiffness-programmed circuit board (GS-PCB). (a) GS-PCB schematic overview. i) GS-PCB prior to phase changeover. ii) Phase transition is used to accomplish stiffness mapping. iii) The GS-PCB exhibits stable stretching behaviour. iv) Avoiding delamination of chips. v) The gradient stiffness design's improved stretchability. (b) Diagram showing a spatially controlled phase transition and the characteristics that go along with it. (c) Diagram demonstrating the laser-patterned liquid metal conductor's steady electromechanical performance. (d) Stretching tests that contrast GS-PCB with a traditional substrate that does not have stiffness mapping: i) circuit with chips installed, ii) conventional

substrate that delaminates chips when stretched, and iii) GS-PCB that remains stable throughout. 1 mm is the scale bar. Reproduced with permission from ref. [215]. Copyright 2024 Wiley.

5.4 Thermal management

Phase transition materials are very promising for thermal management in electronic devices because of their high latent heat and capacity to absorb significant amounts of heat during the solid–liquid phase transition [216]. Phase transition hydrogels are especially useful for wearable electronics because they have inherent flexibility and stretchability in contrast to conventional phase transition materials like paraffin and metal alloys, and because of their composition of natural or biodegradable materials, which offers superior biocompatibility and recyclability [217]. Furthermore, these hydrogels' reversible phase transition qualities allow them to self-heal after mechanical injury, extending the life of wearable technology.

Numerous commercial heat sinks based on inorganic materials have been created to address the issue of heat dissipation in electronic devices; nevertheless, their limited application scenarios and single physical state restrict the heat dissipation impact. By injecting the phase transition gel between two layers of copper sheets, Zhao et al. [218] created a PAA-PEG-based phase transition hydrogel composite thermal spacer (**Figure 13a**). The gap between the hydrogel and the copper sheets was closed by completing a heating–cooling cycle, which greatly increased the device's overall thermal conductivity (**Figure 13b)**. The way phase transition hydrogels lose water is similar to how electronic devices release heat.To absorb a significant amount of heat produced by electronic components, Cheng et al. proposed a heat sink device based on a metal–organic framework/PVA/$CaCl_2 \cdot 6H_2O$ (MOF@PVA/CH) bilayer hydrogel. This device was able to effectively evaporate water molecules at high temperatures with a rate of ≈ 0.90 kg m^{-2} h^{-1} [219]. The extreme heat accumulation close to the CPU was much reduced when they tested this device on a smartphone (**Figure 13c**). Composite thermal materials, which combine phase transition hydrogels with conventional thermal conductive materials, have also attracted a lot of interest. To construct a wax–graphite–hydrogel (W–G–H) composite thermal sheet, Paramparambath et al. mixed graphite and hydrogel with paraffin wax. This resulted in a 12x increase in heat conductivity when compared to pure paraffin wax. When the W-G-H coating was applied to the back of a solar panel, the paraffin and graphite flakes functioned as heat conductors, transferring heat from the panel to the hydrogel. This caused the hydrogel to absorb heat and lose water, thereby reducing the panel's surface temperature (**Figure 13d**). These phase transition hydrogels will provide more application

View Article Online
DOI: 10.1039/D6MA00167J

value in the thermal control of wearable electronics because of their inherent flexibility.

The flexibility, form stability, and self-healing qualities of hydrogels are combined with the high latent heat and limited phase-transition temperature range of solid-liquid phase change materials to create heat-transfer hydrogels, which are appealing for use in chemistry, biomedicine, thermophysics, and other fields. By enclosing the phase-change material in a three-dimensional hydrogel network, PCHs solve the leakage and instability problems of traditional PCMs in heat control. For instance, Shen et al. [220] achieved efficient building cooling by creating a shape-stabilized organohydrogel by the polymerization of PCM-in-water emulsions catalyzed by horseradish peroxidase (HRP) (**Figure 13e**). Yang et al. [221] created thermally conductive channels that improved heat transmission by encapsulating polyethylene glycol (PEG) in polyacrylic acid (PAA) hydrogels containing graphene nanosheets (**Figure 13f-g**).

Since buildings use more than 41% of the world's energy, PCHs are very beneficial for building thermal control. PCHs can stop PCM leakage and save energy [222]. For passive cooling, Feng et al. [223] created a bilayer hydrogel structure modelled after human skin (**Figure 13h**). Sang et al. [224] achieved enhanced thermal conductivity and anti-leakage performance during 300 temperature cycles by using a eutectic hydrated salt/polymer hydrogel (**Figure 13i**). Paraffin wax and sodium acetate trihydrate were combined by Shao et al. [225] using Pickering emulsions to create a shape-stable emulsion gel with high latent heat and little loss during many cycles (**Figure 13j-m**). Although PCHs have a lot of potential for controlling building temperatures, improving their long-term stability is still a significant obstacle.

Beyond buildings, PCHs and PTHs are used in textile thermal management. In this context, solid-liquid PCMs store and release heat during phase transitions to provide thermophysiological comfort under a variety of environmental circumstances. PCHs are promising in personal thermal management (PTM) because of their biocompatibility and latent heat storage capability, which provide people with localized thermal comfort and safety. PCMs are also becoming more popular in thermal camouflage and infrared stealth, where they absorb target heat, lessen detection risk, and eliminate hotspots when restricted in hydrogel networks.

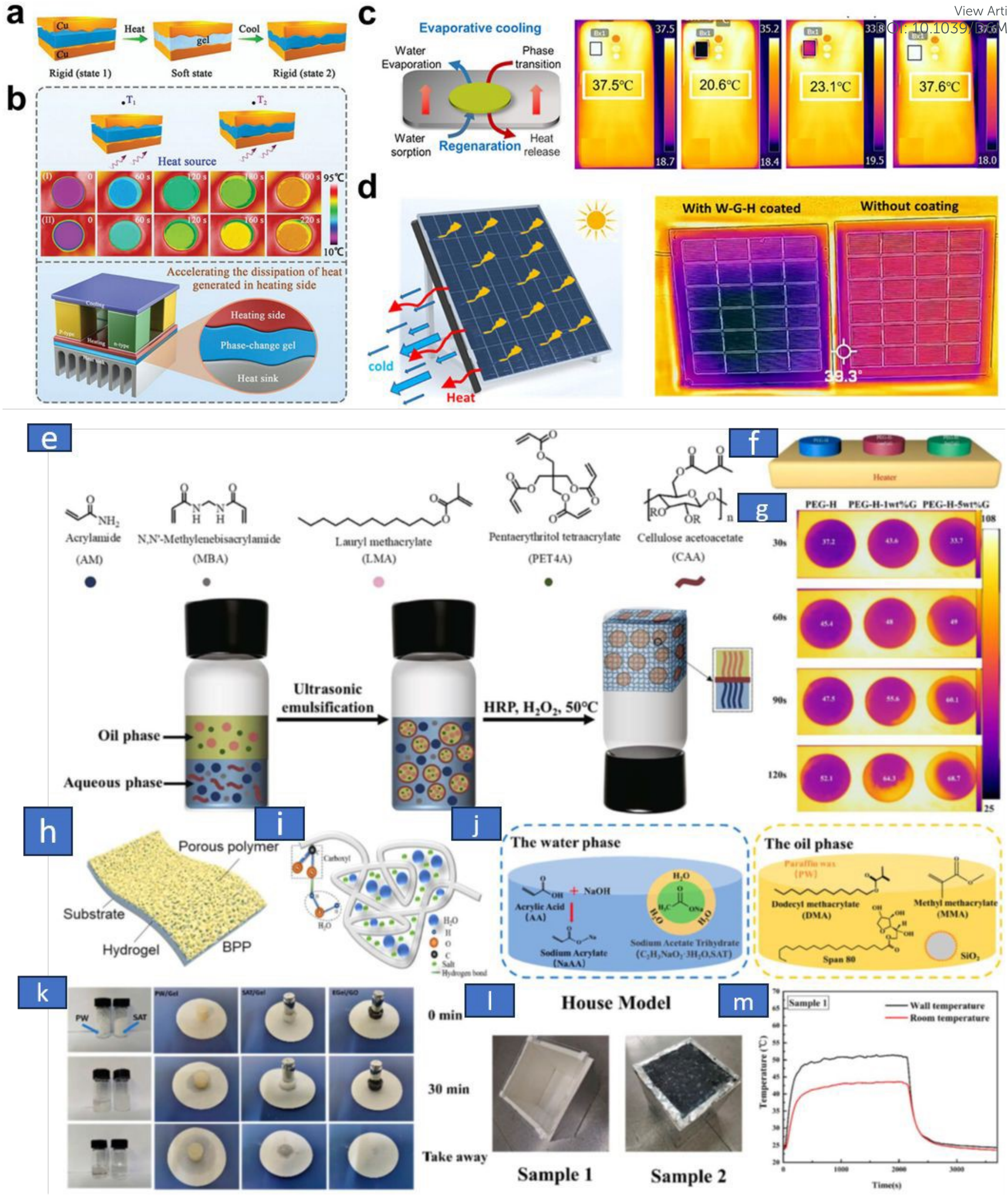


Figure 13: a) Diagrammatic representations showing how the phase transition gel is embedded between two stiff components. b) A comparison of two distinct states for heat conduction. An example of the completed thermoelectric cooling system, in which the phase transition gel acts as a gasket at the thermal interface. c) Diagram illustrating the operation of a self-sufficient high-enthalpy thermal management system that combines phase transition hydrogel with MOF, as well as infrared (IR) pictures of the smartphone's back. d) Diagram showing the heat sink

caused by the W-G-H coating that is applied to the solar panel's rear. This thermal picture shows the surface temperature mapping of solar panels, both coated and uncoated, with W-G-H composite. Reproduced with permission from ref. [7]. Copyright 2025 Wiley. Thermal control of buildings. The synthesis procedures of (e) oil-in-water emulsion-templated organohydrogel are shown schematically. (f) Experimental heating setup; (g) infrared thermal pictures showing PCHs' top surface temperature. (h) A picture of the bilayer porous polymer film's structure. (i) The eutectic hydrated salt/polymer hydrogel encapsulation concept. Oil-water biphasic PCH synthesis schematic diagram (j), PCH leakage tests (k), home models (l), and their photothermal temperature curves (m). Reproduced with permission from ref. [9]. Copyright 2025 Royal Society of Chemistry.

Table 5 summarizes the many functions of phase transition hydrogels (PTHs) and phase change hydrogels (PCHs), as well as their multipurpose uses in robotics, optics, wearable electronics, medicine, and thermal management. Their distinct phase transition-driven behaviors, along with attributes like biocompatibility, self-healing, flexibility, and reactivity to external stimuli, allow for widespread use in both established and developing domains, as this tabular depiction illustrates. The promise of PTHs and PCHs as next-generation smart materials for energy storage, healthcare, soft robotics, and sustainable electronics is highlighted by their incorporation into cutting-edge technology.

Table 5: A brief summary of the optical systems, wearable electronics, soft robotics, thermal management and medical applications of PTHs and PCHs.

Area of Application	**Particular Use**	**Important attributes or benefits**	**Type of Stimuli**	**Examples**	**Ref.**
Biomedical	Systems for delivering drugs	Targeted administration for cancer and chronic illnesses; high biocompatibility; controlled, localized, and stimuli-	Light, pH, and temperature	Injectable PTHs for targeted treatments; thermoresponsive PCHs for anticancer medication release	[226]

View Article Online
DOI: 10.1039/D6MA00167J

		responsive release			
	Wound dressings and skin care	Self-healing, bacteriostatic action, and moisture retention; promotes healing and creates a barrier of defense	pH and temperature	Antimicrobial PCH dressings; hydrogel burn and skin healing patches	[25,195]
	Devices for photothermal treatment	For targeted tumor ablation, convert light to heat; stability is improved by a reversible phase shift.	NIR and light irradiation	PTHs for cancer treatment using graphene oxide and gold nanorods	[65]
Wearable sensors and electrodes	Monitoring of the environment and health (temperature, humidity, breathing, joint mobility, sleep, and voice recovery)	High sensitivity, biocompatibility, wearability, and flexibility; adjustable conductivity and swelling	Bioelectric signals, strain, temperature, and humidity	Smart wearable health patches; PCH electrodes for ECG/EMG; strain sensors based on hydrogel	[196,227]
	Soft robots with shape-morphing actuators for precise	Safety, flexibility, and adaptability; motion is made	Light, magnetic field, electric	Bidirectional bending using UCST–LCST layered gels;	[228]

View Article Online
DOI: 10.1039/D6MA00167J

Soft robotics and optical applications	movements (bending, walking, and grasping)	possible by differential swelling and temperature response.	field, and temperature	hydrogel-based gripping actuators	
	Optical apparatus	Devices with adjustable refractive index and color/structural changes in response to light	Temperature and light	Photonic hydrogel crystals and optical switches based on PTH	[229]
Thermal management	Heat storage/dissipation in electronics, wearables, and buildings	High latent heat, reversible solid–liquid transition, leakage-proof, stretchable, self-healing	Temperature	PCH-based thermal interface materials; organohydrogel heat sinks	[30]
	Heat dissipation and storage in buildings, wearable technology, and electronics	High latent heat, self-healing, stretchy, leak-proof, and reversible solid-liquid transition	Temperature	Organohydrogel heat sinks and PCH-based thermal interface materials	[220]
	Individual thermal control	Localized temperature control for people's comfort and safety	Light and temperature	Wearable thermal pads and patches based on PCH	[200]

	Stealth and concealment using infrared	Reduce IR signatures and conceal hotspots by absorbing or redistributing heat.	Infrared radiation and temperature	Wearable camouflage and defense applications using PCH composites	[230]

6. Safety assessment, challenges, and future perspectives

Before being implemented in real-world applications, a comprehensive evaluation of the safety, biocompatibility and environmental impact of nanoparticle-reinforced phase-transition hydrogels (PTHs) and phase-change hydrogels (PCHs) is needed. The materials possess high potential in biomedicine, energy and soft electronics applications and need further studies to confirm the long-term safety properties of these materials. Biomedically, it is a significant problem, especially in the presence of inorganic nanoparticles such as metal oxides, carbon-based nanomaterials, and metallic nanostructures in the system [231]. Depending on the concentration, surface chemistry, and size of the particles, these nanomaterials can interact with the biological environment to cause oxidative stress, disruption of membranes and/or interference with cellular signalling pathways. While numerous of the hydrogel matrices are benign, the addition of nanoparticles could affect the biological response, and the dose and composition of the hydrogel must be carefully optimized.

Factors of importance for in vivo use are immunogenicity and inflammatory reactions. The immune system may become activated if foreign substances enter the body, causing fibrosis and encapsulation or chronic inflammation. If the PTHs and PCHs are intended to be used in tissue engineering, for medication administration, or for implantable devices, it is important to ensure that the nanostructures embedded in the polymer network and the polymer network itself are not immunogenic [232,233]. Surface functionalization, bioinert coatings and introduction of biocompatible polymers like polysaccharides and synthetic biodegradable networks can help mitigate these risks. A long-term issue is the accumulation of nanoparticles in biological tissues and their subsequent elimination. In the long term, non-biodegradable or slow-biodegradable nanoparticles can accumulate in vital organs and pose potential risks. This is particularly relevant when using the product for a long time or repeatedly, because of the possibility of chronic exposure. In order to address this issue, approaches such as using biodegradable nanofillers, renal clearance of size-controlled particles and stimuli-responsive degradation processes are being explored increasingly.

Biological safety is also crucial in the context of environmental sustainability, particularly with regard to its wide-ranging applicability in the energy and environmental fields [234]. Chemical reagents, solvents, and energy-intensive procedures that may not be in line with green chemistry principles are often used in the production of hydrogels reinforced with nanoparticles. Hence, solvent-free, aqueous-based and renewable and bio-derived polymers like cellulose, alginate and chitosan have been gaining increasing interest in the development of environmentally friendly synthesis pathways [235,236]. To reduce the effect on the environment, these materials' recyclability and end-of-life disposal must also be taken into account. Ecological toxicity is also a concern with potential environmental release after use or degradation of nanoparticles. The leached nanoparticles can have unforeseen effects when they are in contact with soil ecosystems, microbes and aquatic systems. Therefore, it is essential to create robust polymer–nanoparticle interfaces and leakage-prevention encapsulation techniques for the environment and for device function. In this industry, standardizing safety evaluation procedures continues to be a major difficulty. Information on toxicity and biocompatibility is often presented in a complex and contradictory way due to the differences in the composition of materials, manufacturing processes and testing conditions. To enable trustworthy comparison and regulatory approval of these materials, it will be crucial to establish uniform assessment frameworks, encompassing in vitro and in vivo testing criteria. In general, a holistic strategy is needed, involving materials design, toxicological evaluation and environmental aspects for the safe and sustainable development of nanoparticle-reinforced PTHs and PCHs. It will be critical to solve these problems to bring these systems from the prototype stage to a commercial and clinically viable solution. Although there is some progress, there are still a number of challenges that need to be overcome before commercialization:

- Mechanical-functional trade-off: Higher crosslink density results in higher strength but lower phase transition; Higher PCM results in higher latent heat but lower form stability. This can be addressed with hierarchical designs, dynamic crosslinks and dual-network structures.

- Slow phase transitions and limited temperature control cause real-time response to be limited in micro-actuators and bioelectronics. Transition kinetics have to be carefully tuned to ensure consistent performance.

View Article Online
DOI: 10.1039/D6MA00167J

- Long-term reliability is decreased due to network fatigue, microcracks, crosslink degradation and leakage of PCM. Gradient designs, dynamic covalent crosslinking and nanoparticle reinforcement have the potential to solve these issues.
- Electronic integration: Moisture content and thermal expansion and conductivity differences between hydrogels and silicon-based electronics can lead to instability at the interface. For reliable integration, development of hybrid materials, encapsulation and hydrophobic coatings are required.
- It may not be as cost-efficient and as industry friendly to be synthesized using multi-step processes and special additives. Fabrication methods should be eco-friendly, streamlined and scalable.

A number of research paths are anticipated to help accelerate the implementation of PTHs and PCHs in practice:

- Hybrid and multifunctional designs: Nanoparticles, nanofibers, 2D materials or conductive polymers can be combined to enhance thermal, electrical and optical properties for next-generation bioelectronics, multifunctional robotics and self-powered wearables.
- Hierarchical and gradient structures (e.g., cartilage or skin) can provide superior toughness, adaptability and responsiveness via bioinspired and adaptive architectures. The use of smart hydrogels that respond to external stimuli or that contain AI sensors is promising.
- Optimizing the polymer composition, network structures and fillers for faster and more precise transition kinetics and temperature control can provide rapid and accurate device performance.
- Sustainable and scalable fabrication methods: Hydrogels can be fabricated in a cost-effective and reproducible manner on a large scale by sustainable and scalable fabrication methods like Additive manufacturing, 3D printing and freeze-casting.
- Biocompatibility, Non Toxicity, Fatigue resistance and Self healing are important for wearable electronics, healthcare and clinical applications for long term reliability and safety.

- In addition to biomedical and energy systems, PTHs and PCHs can be used in other industrial sectors, including renewable energy, space technologies, environmental monitoring, smart infrastructures, etc., which will enhance sustainability and societal value.

7. Conclusions

Phase transition hydrogels (PTHs) and phase change hydrogels (PCHs) have come a long way and are now used in biomedical engineering, wearable electronics, soft robotics, flexible bioelectronics, and energy storage. The “smart” soft materials are made of phase change materials (PCMs) and polymers that have tunable, stimuli-responsive behaviour. The reversible transitions of PTHs and PCHs in response to temperature, pH, light, magnetic and electric stimuli, as well as their mechanical flexibility and nanoparticle reinforcement, make them ideal for multifunctional devices like thermal camouflage, drug delivery, and adaptive soft systems. The following are some of the main findings:

- In bioelectronics and medicine, tunable, reversible phase transitions, stretchability, biocompatibility and self-healing properties enable applications such as wearable sensors, flexible electrodes, soft actuators, drug delivery systems, wound dressings and photothermal therapy (PTT) for precise and minimally invasive treatment.
- Thermal energy storage and multifunctional composites: 3D hydrogel networks are used to encapsulate PCMs, preventing leakage and adding mechanical flexibility. Functional fillers can be used in infrared stealth, textile, personal thermal management (PTM), building thermal management, flame retardancy, photothermal conversion and thermoelectric energy production applications.
- Cross-disciplinary integration: Both PTHs and PCHs are suitable for optical devices, soft robotics, and flexible electronics, and can be dynamically modulated for sensing, adaptive systems, and camouflage.
- Overall significance: PTHs and PCHs link soft matter chemistry and functional device engineering and provide long-lasting, versatile and multifunctional platforms for future technologies.

Therefore, PTHs and PCHs are emerging as new soft materials, which are polymer chemistry and applied device engineering combined. Although mechanical–functional trade-offs,

View Article Online
DOI: 10.1039/D6MA00167J

integration problems, and long-term stability are still present, current advances in nanomaterial reinforcement, dynamic crosslinking, and bioinspired designs are overcoming these challenges. Interdisciplinary research is expected to continue in the future, leading to the commercialization of PTHs and PCHs for multifunctional, adaptive and sustainable solutions in biomedical, electronic and energy applications.

References

[1] H. Cao, L. Duan, Y. Zhang, J. Cao, K. Zhang, Current hydrogel advances in physicochemical and biological response-driven biomedical application diversity, Signal Transduct. Target. Ther. 6 (2021) 426. https://doi.org/10.1038/s41392-021-00830-x.

[2] S.R. Caliari, J.A. Burdick, A practical guide to hydrogels for cell culture, Nat. Methods 13 (2016) 405–414. https://doi.org/10.1038/nmeth.3839.

[3] P. Contessotto, D. Orbanić, M. Da Costa, C. Jin, P. Owens, S. Chantepie, C. Chinello, J. Newell, F. Magni, D. Papy-Garcia, N.G. Karlsson, M. Kilcoyne, P. Dockery, J.C. Rodríguez-Cabello, A. Pandit, Elastin-like recombinamers-based hydrogel modulates post-ischemic remodeling in a non-transmural myocardial infarction in sheep, Sci. Transl. Med. 13 (2021). https://doi.org/10.1126/scitranslmed.aaz5380.

[4] S. Awasthi, In Vitro and In Vivo Therapeutics of Double-Layered Hydrogels, Adv. Ther. (Weinh). 8 (2025). https://doi.org/10.1002/adtp.202400266.

[5] P. Lu, D. Ruan, M. Huang, M. Tian, K. Zhu, Z. Gan, Z. Xiao, Harnessing the potential of hydrogels for advanced therapeutic applications: current achievements and future directions, Signal Transduct. Target. Ther. 9 (2024) 166. https://doi.org/10.1038/s41392-024-01852-x.

[6] J. Ma, J. Zhong, F. Sun, B. Liu, Z. Peng, J. Lian, X. Wu, L. Li, M. Hao, T. Zhang, Hydrogel sensors for biomedical electronics, Chemical Engineering Journal 481 (2024) 148317. https://doi.org/10.1016/j.cej.2023.148317.

[7] Z. Zhu, Y. Gu, X. Wang, W. Fan, J. He, Y. Li, Multifunctional Bioelectronics Achieved by Empowering Hydrogels with Phase-Transition Ability, Adv. Funct. Mater. (2025). https://doi.org/10.1002/adfm.202507013.

[8] L. Yu, K. She, R. He, Q. Xu, Application and progress of temperature-sensitive hydrogels in cartilage injury repair, Front. Bioeng. Biotechnol. 13 (2025). https://doi.org/10.3389/fbioe.2025.1602303.

[9] H. Li, M. Sun, Z. Liu, C. Nie, C. Hu, D. Tang, Z. Rao, Solid–liquid phase change materials meet hydrogels: syntheses and multifunctional applications, J. Mater. Chem. A Mater. 13 (2025) 21245–21267. https://doi.org/10.1039/D5TA00346F.

[10] N.A. Shaibie, N.A. Ramli, N.D.F. Mohammad Faizal, T. Srichana, M.C.I. Mohd Amin, Poly(N-isopropylacrylamide)-Based Polymers: Recent Overview for the Development of

Temperature-Responsive Drug Delivery and Biomedical Applications, Macromol. Chem. Phys. 224 (2023). https://doi.org/10.1002/macp.202300157.

[11] J. Singh, P. Nayak, pH-responsive polymers for drug delivery: Trends and opportunities, Journal of Polymer Science 61 (2023) 2828–2850. https://doi.org/10.1002/pol.20230403.

[12] F. Xu, B.L. Feringa, Photoresponsive Supramolecular Polymers: From Light-Controlled Small Molecules to Smart Materials, Advanced Materials 35 (2023). https://doi.org/10.1002/adma.202204413.

[13] Z. Jiang, J. Tang, C. Zhang, Y. Xie, J. Chen, Thermoresponsive hydrogels incorporating phase-change energy storage materials for advanced thermal management, Polym. Chem. 16 (2025) 3379–3395. https://doi.org/10.1039/D5PY00318K.

[14] M. Zare, K.S. Mikkonen, Phase Change Materials for Life Science Applications, Adv. Funct. Mater. 33 (2023). https://doi.org/10.1002/adfm.202213455.

[15] J. Bao, H. Tu, J. Li, Y. Li, S. Yu, J. Gao, K. Lei, F. Zhang, J. Li, Applications of phase change materials in smart drug delivery for cancer treatment, Front. Bioeng. Biotechnol. 10 (2022). https://doi.org/10.3389/fbioe.2022.991005.

[16] R. Fan, Y. Cheng, R. Wang, T. Zhang, H. Zhang, J. Li, S. Song, A. Zheng, Thermosensitive Hydrogels and Advances in Their Application in Disease Therapy, Polymers (Basel). 14 (2022) 2379. https://doi.org/10.3390/polym14122379.

[17] R. Fan, Y. Cheng, R. Wang, T. Zhang, H. Zhang, J. Li, S. Song, A. Zheng, Thermosensitive Hydrogels and Advances in Their Application in Disease Therapy, Polymers (Basel). 14 (2022) 2379. https://doi.org/10.3390/polym14122379.

[18] X. Li, J.P. Gong, Design principles for strong and tough hydrogels, Nat. Rev. Mater. 9 (2024) 380–398. https://doi.org/10.1038/s41578-024-00672-3.

[19] Z. Zhang, S. Yang, F. Mi, Y. Yang, Q. Song, Y. Gao, C. Wu, W. Wen, Nanoparticle-Reinforced Hydrogel with a Well-Defined Pore Structure for Sustainable Drug Release and Effective Wound Healing, ACS Appl. Bio Mater. 8 (2025) 1406–1417. https://doi.org/10.1021/acsabm.4c01659.

[20] S. Awasthi, J.K. Gaur, M.S. Bobji, C. Srivastava, Nanoparticle-reinforced polyacrylamide hydrogel composites for clinical applications: a review, J. Mater. Sci. 57 (2022) 8041–8063.

[21] M.A. Campea, M.J. Majcher, A. Lofts, T. Hoare, A Review of Design and Fabrication Methods for Nanoparticle Network Hydrogels for Biomedical, Environmental, and Industrial Applications, Adv. Funct. Mater. 31 (2021). https://doi.org/10.1002/adfm.202102355.

[22] A. Bin Rashid, M. Haque, S.M.M. Islam, K.M.R. Uddin Labib, Nanotechnology-enhanced fiber-reinforced polymer composites: Recent advancements on processing techniques and applications, Heliyon 10 (2024) e24692. https://doi.org/10.1016/j.heliyon.2024.e24692.

View Article Online
DOI: 10.1039/D6MA00167J

[23] S. Awasthi, S. De, S.K. Pandey, Electrodeposited carbon nanostructured nickel composite coatings: A review, Heliyon 10 (2024) e26051. https://doi.org/10.1016/j.heliyon.2024.e26051.

[24] H. Wang, J. Du, Y. Mao, Hydrogel-Based Continuum Soft Robots, Gels 11 (2025) 254. https://doi.org/10.3390/gels11040254.

[25] M. Yun, L. Langford, L. Russell, N. Ndiforamang, A. Zhang, W. Bai, Emerging Stimuli-Responsive Hydrogels to Facilitate Healing of Chronic Wounds, RSC Applied Polymers (2025). https://doi.org/10.1039/D5LP00092K.

[26] I.S. Protsak, Y.M. Morozov, Fundamentals and Advances in Stimuli-Responsive Hydrogels and Their Applications: A Review, Gels 11 (2025) 30. https://doi.org/10.3390/gels11010030.

[27] T. Lv, Y. Chen, N. Li, X. Liao, Y. Heng, Y. Guo, K. Hu, A Comprehensive Review of Thermosensitive Hydrogels: Mechanism, Optimization Strategies, and Applications, Gels 11 (2025) 544. https://doi.org/10.3390/gels11070544.

[28] H. Kojima, Studies on the phase transition of hydrogels and aqueous solutions of thermosensitive polymers, Polym. J. 50 (2018) 411–418. https://doi.org/10.1038/s41428-018-0035-9.

[29] M. Chen, R. Liu, Y. Yang, Q. Bai, L. Xie, H. Zhu, A review on regulating the mechanical properties and phase transition temperature of UCST hydrogels, Eur. Polym. J. 243 (2026) 114482. https://doi.org/10.1016/j.eurpolymj.2025.114482.

[30] C. He, H. Chen, Z. Huang, Z. Song, X. Liu, S. Gao, J. Zhang, X. Zhang, Y. Jiang, H. Gao, Review on recent advances in shape-stable phase change hydrogels for thermal energy storage applications, J. Energy Storage 128 (2025) 117245. https://doi.org/10.1016/j.est.2025.117245.

[31] H. Kojima, Studies on the phase transition of hydrogels and aqueous solutions of thermosensitive polymers, Polym. J. 50 (2018) 411–418. https://doi.org/10.1038/s41428-018-0035-9.

[32] Y. Akiyama, S. Osawa, K. Masamune, Y. Muragaki, Alteration of the upper critical solution temperature (UCST) behavior of the nonionic polymer poly(N-acryloyl-nipecotamide) by its terminal group, Eur. Polym. J. 220 (2024) 113454. https://doi.org/10.1016/j.eurpolymj.2024.113454.

[33] L. Klouda, A.G. Mikos, Thermoresponsive hydrogels in biomedical applications, European Journal of Pharmaceutics and Biopharmaceutics 68 (2008) 34–45. https://doi.org/10.1016/j.ejpb.2007.02.025.

[34] S. Ashraf, H.-K. Park, H. Park, S.-H. Lee, Snapshot of phase transition in thermoresponsive hydrogel PNIPAM: Role in drug delivery and tissue engineering, Macromol. Res. 24 (2016) 297–304. https://doi.org/10.1007/s13233-016-4052-2.

[35] M. Levin, N. Cohen, Thermo-Mechanics of PNIPAM Gels: from a Single Chain to a Network Response, Macromolecules 58 (2025) 5187–5200. https://doi.org/10.1021/acs.macromol.4c03233.

[36] X. Meng, Y. Qiao, C. Do, W. Bras, C. He, Y. Ke, T.P. Russell, D. Qiu, Hysteresis-Free Nanoparticle-Reinforced Hydrogels, Advanced Materials 34 (2022). https://doi.org/10.1002/adma.202108243.

[37] L. Navarro, L.E. Theune, M. Calderón, Effect of crosslinking density on thermoresponsive nanogels: A study on the size control and the kinetics release of biomacromolecules, Eur. Polym. J. 124 (2020) 109478. https://doi.org/10.1016/j.eurpolymj.2020.109478.

[38] E. Turan, T. Çaykara, Swelling and network parameters of pH-sensitive poly(acrylamide-co-acrylic acid) hydrogels, J. Appl. Polym. Sci. 106 (2007) 2000–2007. https://doi.org/10.1002/app.26848.

[39] G. Elyashevich, E. Rosova, Z. Zoolshoev, N. Saprykina, I. Kuryndin, Reversibility of Swelling, pH Sensitivity, Electroconductivity, and Mechanical Properties of Composites Based on Polyacrylic Acid Hydrogels and Conducting Polymers, Journal of Composites Science 7 (2023) 261. https://doi.org/10.3390/jcs7060261.

[40] G. Patroklou, E. Triantafyllopoulou, P.-E. Goula, V. Karali, M. Chountoulesi, G. Valsami, S. Pispas, N. Pippa, pH-Responsive Hydrogels: Recent Advances in Pharmaceutical Applications, Polymers (Basel). 17 (2025) 1451. https://doi.org/10.3390/polym17111451.

[41] K. Gao, K. Xu, Advancements and Prospects of pH-Responsive Hydrogels in Biomedicine, Gels 11 (2025) 293. https://doi.org/10.3390/gels11040293.

[42] T. Pandey, V. Pandey, A mechanistic understanding to photophysical phenomenon in development of near-infrared (NIR) responsive hydrogels: Advancements in precision drug delivery, J. Drug Deliv. Sci. Technol. 106 (2025) 106682. https://doi.org/10.1016/j.jddst.2025.106682.

[43] A. Raza, U. Hayat, T. Rasheed, M. Bilal, H.M.N. Iqbal, “Smart” materials-based near-infrared light-responsive drug delivery systems for cancer treatment: A review, Journal of Materials Research and Technology 8 (2019) 1497–1509. https://doi.org/10.1016/j.jmrt.2018.03.007.

[44] Y. Xing, B. Zeng, W. Yang, Light responsive hydrogels for controlled drug delivery, Front. Bioeng. Biotechnol. 10 (2022). https://doi.org/10.3389/fbioe.2022.1075670.

[45] T. V. Patil, D.K. Patel, S.D. Dutta, K. Ganguly, K.-T. Lim, Graphene Oxide-Based Stimuli-Responsive Platforms for Biomedical Applications, Molecules 26 (2021) 2797. https://doi.org/10.3390/molecules26092797.

[46] I. Carayon, A. Gaubert, Y. Mousli, B. Philippe, Electro-responsive hydrogels: macromolecular and supramolecular approaches in the biomedical field, Biomater. Sci. 8 (2020) 5589–5600. https://doi.org/10.1039/D0BM01268H.

View Article Online
DOI: 10.1039/D6MA00167J

[47] M. Suhail, Alamgir, A. Wahab, T. Eggers, Z. Ahmad, K. Shehzad, M.Z. Iqbal, Magnetically responsive hydrogel systems: Fundamental features, emerging applications, and future horizons, Coord. Chem. Rev. 543 (2025) 216916. https://doi.org/10.1016/j.ccr.2025.216916.

[48] Md.A. Shahid, Md.M. Rahman, Md.T. Hossain, I. Hossain, Md.S. Sheikh, Md.S. Rahman, N. Uddin, S.W. Donne, Md.I.U. Hoque, Advances in Conductive Polymer-Based Flexible Electronics for Multifunctional Applications, Journal of Composites Science 9 (2025) 42. https://doi.org/10.3390/jcs9010042.

[49] S. Meenakshy, J. Jesslyn, S. Anas, Development and Applications of Polypyrrole-Based Conductive Inks: An Overview, Adv. Mater. Technol. 10 (2025). https://doi.org/10.1002/admt.202401216.

[50] X. Fu, L. Hosta-Rigau, R. Chandrawati, J. Cui, Multi-Stimuli-Responsive Polymer Particles, Films, and Hydrogels for Drug Delivery, Chem 4 (2018) 2084–2107. https://doi.org/10.1016/j.chempr.2018.07.002.

[51] S. Rittikulsittichai, A.G. Kolhatkar, S. Sarangi, M.A. Vorontsova, P.G. Vekilov, A. Brazdeikis, T. Randall Lee, Multi-responsive hybrid particles: thermo-, pH-, photo-, and magneto-responsive magnetic hydrogel cores with gold nanorod optical triggers, Nanoscale 8 (2016) 11851–11861. https://doi.org/10.1039/C5NR09235C.

[52] S. Jiang, L. Xia, H. Ma, T. Yang, L. Qian, pH and temperature dual-responsive hydrogel actuator with bidirectional bending behavior and ultra large bending angle, Eur. Polym. J. 197 (2023) 112296. https://doi.org/10.1016/j.eurpolymj.2023.112296.

[53] G. Ciarleglio, E. Toto, M.G. Santonicola, Conductive and Thermo-Responsive Composite Hydrogels with Poly(N-isopropylacrylamide) and Carbon Nanotubes Fabricated by Two-Step Photopolymerization, Polymers (Basel). 15 (2023) 1022–1038.

[54] J. Ren, Y. Hu, C.-H. Lu, W. Guo, M.A. Aleman-Garcia, F. Ricci, I. Willner, pH-responsive and switchable triplex-based DNA hydrogels, Chem. Sci. 6 (2015) 4190–4195. https://doi.org/10.1039/C5SC00594A.

[55] Y. Lu, C. Chen, H. Li, P. Zhao, Y. Zhao, B. Li, W. Zhou, G. Fan, D. Guan, Y. Zheng, Visible light-responsive hydrogels for cellular dynamics and spatiotemporal viscoelastic regulation, Nat. Commun. 16 (2025) 1365. https://doi.org/10.1038/s41467-024-54880-0.

[56] S. Awasthi, A. Srivastava, A. Goyal, Design Strategies of Magneto-Responsive Polyacrylamide Ferrogels, J. Clust. Sci. 36 (2025) 214. https://doi.org/10.1007/s10876-025-02945-5.

[57] P. Schütz, S.B. Lemich, M. Weißpflog, P. Körner, V. Abetz, B. Hankiewicz, Multi-responsive hydrogels based on magneto-plasmonic nanoparticles in a thermo-responsive polymer matrix, Nano Trends 10 (2025) 100113. https://doi.org/10.1016/j.nwnano.2025.100113.

[58] B. Jeong, S.W. Kim, Y.H. Bae, Thermosensitive sol–gel reversible hydrogels, Adv. Drug Deliv. Rev. 54 (2002) 37–51. https://doi.org/10.1016/S0169-409X(01)00242-3.

View Article Online
DOI: 10.1039/D6MA00167J

[59] S. Narayana, B.H.J. Gowda, U. Hani, M.G. Ahmed, Z.A. Asiri, K. Paul, Smart Poly(N-isopropylacrylamide)-Based Hydrogels: A Tour D'horizon of Biomedical Applications, Gels 11 (2025) 207. https://doi.org/10.3390/gels11030207.

[60] M. Boruah, A. Sarma, Dry Gels: Concept, Current Trends, and New Avenues in Drug Delivery and Biomedical Application, Adv. Healthc. Mater. (2025). https://doi.org/10.1002/adhm.202500863.

[61] P. Yapa, I. Munaweera, Functionalized nanoporous architectures derived from sol–gel processes for advanced biomedical applications, J. Mater. Chem. B (2025). https://doi.org/10.1039/D5TB00958H.

[62] A. Dolai, K. Manna, S. Samanta, G. Roymahapatra, S. Pal, Reversible Light-Controlled Sol–Gel Transition of an Ionic Hydrogel, Langmuir (2025). https://doi.org/10.1021/acs.langmuir.5c02433.

[63] G.J. Owens, R.K. Singh, F. Foroutan, M. Alqaysi, C.-M. Han, C. Mahapatra, H.-W. Kim, J.C. Knowles, Sol–gel based materials for biomedical applications, Prog. Mater. Sci. 77 (2016) 1–79. https://doi.org/10.1016/j.pmatsci.2015.12.001.

[64] C.Y. Gong, P.W. Dong, S. Shi, S.Z. Fu, J.L. Yang, G. Guo, X. Zhao, Y.Q. Wei, Z.Y. Qian, Thermosensitive PEG–PCL–PEG Hydrogel Controlled Drug Delivery System: Sol–Gel–Sol Transition and In Vitro Drug Release Study, J. Pharm. Sci. 98 (2009) 3707–3717. https://doi.org/10.1002/jps.21694.

[65] Z. Huang, Y. Liu, T. Zhu, W. Jiang, D. Sun, J. Yang, X. Qi, Y. Wang, Phase change hydrogels with tunable adhesion for wearable thermal management and intelligent healthcare, J. Energy Storage 98 (2024) 113043. https://doi.org/10.1016/j.est.2024.113043.

[66] M. Song, L. Wang, F. Shao, H. Xie, H. Xu, W. Yu, Thermally induced flexible phase change hydrogels for solar thermal storage and human thermal management, Chemical Engineering Journal 464 (2023) 142682. https://doi.org/10.1016/j.cej.2023.142682.

[67] M. Popescu, F. Sava, A. Velea, A. Lőrinczi, Crystalline–amorphous and amorphous–amorphous transitions in phase-change materials, J. Non. Cryst. Solids 355 (2009) 1820–1823. https://doi.org/10.1016/j.jnoncrysol.2009.04.053.

[68] S. Rao, S. Huang, X. Liu, S. Lin, C. Glynn, K. Felix, A. Sahasrabudhe, C. Maley, J. Xu, W. Chen, E. Hong, A. Crosby, Q. Wang, Control of Polymers' Amorphous-crystalline Transition for Hydrogel Bioelectronics Miniaturization and Multifunctional Integration, (2023). https://doi.org/10.21203/rs.3.rs-2864872/v1.

[69] R. Islam, S. Maparathne, P. Chinwangso, T.R. Lee, Review of Shape-Memory Polymer Nanocomposites and Their Applications, Applied Sciences 15 (2025) 2419. https://doi.org/10.3390/app15052419.

[70] B.T. Qin, L.L. Zhang, Y. Lu, Preparation and Inhibition Characteristic of Multi-Phase Foamed Gel for Preventing <i>Spontaneous</i> Combustion of Coal, Adv. Mat. Res. 634–638 (2013) 3678–3682. https://doi.org/10.4028/www.scientific.net/AMR.634-638.3678.

[71] Z. Li, C. Ji, F. Guo, H. Mi, X. Zhu, J. Qiu, A multi-interface CoNi-SP/C heterostructure for quasi-solid-state hybrid supercapacitors with a graphene oxide-containing hydrogel electrolyte, J. Mater. Chem. A Mater. 10 (2022) 4671–4682. https://doi.org/10.1039/D1TA10138B.

[72] F. Liu, M.W. Urban, Recent advances and challenges in designing stimuli-responsive polymers, Prog. Polym. Sci. 35 (2010) 3–23. https://doi.org/10.1016/j.progpolymsci.2009.10.002.

[73] P. Ilg, Stimuli-responsive hydrogels cross-linked by magnetic nanoparticles, Soft Matter 9 (2013) 3465. https://doi.org/10.1039/c3sm27809c.

[74] M. Bustamante-Torres, D. Romero-Fierro, B. Arcentales-Vera, K. Palomino, H. Magaña, E. Bucio, Hydrogels Classification According to the Physical or Chemical Interactions and as Stimuli-Sensitive Materials, Gels 7 (2021) 182. https://doi.org/10.3390/gels7040182.

[75] W. Kong, Y. Yang, A. Yuan, L. Jiang, X. Fu, Y. Wang, H. Xu, Z. Liu, J. Lei, Processable and recyclable crosslinking solid-solid phase change materials based on dynamic disulfide covalent adaptable networks for thermal energy storage, Energy 232 (2021) 121070. https://doi.org/10.1016/j.energy.2021.121070.

[76] Y. Gao, K. Peng, S. Mitragotri, Covalently Crosslinked Hydrogels via Step-Growth Reactions: Crosslinking Chemistries, Polymers, and Clinical Impact, Advanced Materials 33 (2021). https://doi.org/10.1002/adma.202006362.

[77] M.S. Rehmann, K.M. Skeens, P.M. Kharkar, E.M. Ford, E. Maverakis, K.H. Lee, A.M. Kloxin, Tuning and Predicting Mesh Size and Protein Release from Step Growth Hydrogels, Biomacromolecules 18 (2017) 3131–3142. https://doi.org/10.1021/acs.biomac.7b00781.

[78] G.S. Sheridan, C.M. Evans, Understanding the Roles of Mesh Size, Tg, and Segmental Dynamics on Probe Diffusion in Dense Polymer Networks, Macromolecules 54 (2021) 11198–11208. https://doi.org/10.1021/acs.macromol.1c01767.

[79] Y. Zhang, W.G. Chapman, Modeling Lower Critical Solution Temperature Behavior of Associating Dendrimers Using Density Functional Theory, Langmuir 35 (2019) 10808–10817. https://doi.org/10.1021/acs.langmuir.9b00514.

[80] Y. Yuan, K. Raheja, N.B. Milbrandt, S. Beilharz, S. Tene, S. Oshabaheebwa, U.A. Gurkan, A.C.S. Samia, M. Karayilan, Thermoresponsive polymers with LCST transition: synthesis, characterization, and their impact on biomedical frontiers, RSC Applied Polymers 1 (2023) 158–189. https://doi.org/10.1039/D3LP00114H.

[81] Z. Ghassemi, J.B. Leach, Impact of Confinement within a Hydrogel Mesh on Protein Thermodynamic Stability and Aggregation Kinetics, Mol. Pharm. 21 (2024) 1137–1148. https://doi.org/10.1021/acs.molpharmaceut.3c00677.

[82] Y. Meng, A Sustainable Approach to Fabricating Ag Nanoparticles/PVA Hybrid Nanofiber and Its Catalytic Activity, Nanomaterials 5 (2015) 1124–1135. https://doi.org/10.3390/nano5021124.

[83] H. Yoshitake, K. Sugimura, Y. Teramoto, Y. Nishio, Magnetic property of oriented films of cellulose nanocrystal/carrageenan composites containing iron oxide nanoparticles: Effect of anisotropic aggregation of the nanoparticles, Polymer (Guildf). 99 (2016) 147–156. https://doi.org/10.1016/j.polymer.2016.07.004.

[84] J. Tan, S. Luo, W. Ji, Y. Li, L. Li, X. Cheng, Phase-changing hydrogels incorporated with copper sulfide-carbon nanotubes for smart thermal management and solar energy storage, J. Energy Storage 50 (2022) 104653. https://doi.org/10.1016/j.est.2022.104653.

[85] L.S. Ribeiro, R.L. Sala, L.A.O. de Jesus, S.A. Cruz, E.R. Camargo, Analyzing the Effects of Silica Nanospheres on the Sol–Gel Transition Profile of Thermosensitive Hydrogels, Langmuir 37 (2021) 7373–7379. https://doi.org/10.1021/acs.langmuir.1c00723.

[86] S. Awasthi, Ferrogels towards nanotheranostics, Mater. Today Chem. 35 (2024) 101877.

[87] S. Sturm, M. Siglreitmeier, D. Wolf, K. Vogel, M. Gratz, D. Faivre, A. Lubk, B. Büchner, E. V. Sturm (née Rosseeva), H. Cölfen, Magnetic Nanoparticle Chains in Gelatin Ferrogels: Bioinspiration from Magnetotactic Bacteria, Adv. Funct. Mater. 29 (2019) 1905996. https://doi.org/10.1002/adfm.201905996.

[88] M. Liang, W. Cao, Y. Huang, Reversible heat-set four-phase transitions of gel1-to-sol1-to-gel2-to-sol2 in binary hydrogels, Front. Chem. Sci. Eng. 18 (2024) 155. https://doi.org/10.1007/s11705-024-2501-6.

[89] A.C. Marques, P.J. Costa, S. Velho, M.H. Amaral, Stimuli-responsive hydrogels for intratumoral drug delivery, Drug Discov. Today 26 (2021) 2397–2405. https://doi.org/10.1016/j.drudis.2021.04.012.

[90] L. Montero De Espinosa, W. Meesorn, D. Moatsou, C. Weder, Bioinspired Polymer Systems with Stimuli-Responsive Mechanical Properties, Chem. Rev. 117 (2017) 12851–12892.

[91] P. Wang, Y. Lv, J. Duan, G. Sun, C. Meng, Y. Li, S. Guo, T. Zhang, A thermally responsive phase-change hydrogel for skin-mountable multifunctional sensors, Nano Energy 136 (2025) 110722. https://doi.org/10.1016/j.nanoen.2025.110722.

[92] D. Hofmann, D. Sychev, Z. Zagradska-Paromova, E. Bittrich, G.K. Auernhammer, J. Gaitzsch, Surface Topology of Redox- and Thermoresponsive Nanogel Droplets, Macromol. Rapid Commun. 45 (2024). https://doi.org/10.1002/marc.202400049.

[93] L. Ruiz-Virgen, M.A. Hernandez-Martinez, G. Martínez-Mejía, R. Caro-Briones, E. Herbert-Pucheta, J.M. del Río, M. Corea, Analysis of Structural Changes of pH–Thermo-Responsive Nanoparticles in Polymeric Hydrogels, Gels 10 (2024) 541. https://doi.org/10.3390/gels10080541.

[94] S. Chatterjee, P.C. Hui, C. Kan, Thermoresponsive Hydrogels and Their Biomedical Applications: Special Insight into Their Applications in Textile Based Transdermal Therapy, Polymers (Basel). 10 (2018) 480. https://doi.org/10.3390/polym10050480.

[95] N. Majstorović, J. Pechtold, S. Agarwal, Upper Critical Solution Temperature Type Thermoresponsive Reactive Copolymers for Enzyme Immobilization, ACS Appl. Polym. Mater. 4 (2022) 5395–5403. https://doi.org/10.1021/acsapm.2c00448.

[96] Y. Chen, B. Gu, X. Hao, Z. Lu, D. Wang, Nanofibrous membrane/thermoresponsive hydrogel composites with temperature-controlled capability for enhancing infected wounds healing, J. Colloid Interface Sci. 680 (2025) 172–180. https://doi.org/10.1016/j.jcis.2024.10.170.

[97] X. Zhou, W. Zhang, P. Yuan, Z. Liu, J. Zhao, Y. Gu, H. Chu, Synthesis of Thermo-Responsive Hydrogel Stabilizer and Its Impact on the Performance of Ecological Soil, Applied Sciences 15 (2025) 8279. https://doi.org/10.3390/app15158279.

[98] M. Hao, B. Qin, Q. Shi, S. Wang, The preparation and characteristics of mining waste-derived thermoresponsive hydrogel used for coal fire suppression, Colloids Surf. A Physicochem. Eng. Asp. 725 (2025) 137644. https://doi.org/10.1016/j.colsurfa.2025.137644.

[99] P.A. Parvathy, V.A. Ajisafe, A.M. Raichur, S.K. Sahoo, Dual responsive curcumin loaded N-isopropylacrylamide/acrylated linseed oil copolymeric hydrogel films: tuned LCST with value-added properties, New Journal of Chemistry 48 (2024) 16065–16079. https://doi.org/10.1039/D4NJ03560G.

[100] S.H. Moon, S.J. Park, Y.W. Lee, Y.J. Yang, LCST/UCST behavior of polysaccharides for hydrogel fabrication, RSC Adv. 14 (2024) 35754–35768. https://doi.org/10.1039/D4RA06240J.

[101] F. Chen, G. Lu, H. Yuan, R. Li, J. Nie, Y. Zhao, X. Shu, X. Zhu, Mechanism and regulation of LCST behavior in poly(hydroxypropyl acrylate)-based temperature-sensitive hydrogels, J. Mater. Chem. A Mater. 10 (2022) 18235–18247. https://doi.org/10.1039/D2TA04271A.

[102] K. Belal, F. Stoffelbach, J. Lyskawa, M. Fumagalli, D. Hourdet, A. Marcellan, L. De Smet, V.R. de la Rosa, G. Cooke, R. Hoogenboom, P. Woisel, Recognition-Mediated Hydrogel Swelling Controlled by Interaction with a Negative Thermoresponsive LCST Polymer, Angewandte Chemie 128 (2016) 14180–14184. https://doi.org/10.1002/ange.201605630.

[103] E. Suljovrujic, M. Krstic, Z. Rogic Miladinovic, S. Petrovic, A. Leskovac, G. Stamboliev, Optimization of thermoresponsive hydrogels based on oligomers with lower critical solution temperature (LCST) far below/above physiological temperatures for biomedical applications, React. Funct. Polym. 189 (2023) 105612. https://doi.org/10.1016/j.reactfunctpolym.2023.105612.

[104] Y. Ling, L. Chen, M. Huang, C. Zhou, L. Yang, H. Niu, L. Su, Y. Yang, R.P. Pirraco, R.L. Reis, J. Chen, A Novel Method for the Preparation of Poly (Acrylamide-co-Acrylonitrile) Upper Critical Solution Temperature Thermosensitive Hydrogel by the Partial Dehydration of Acrylamide Grafted Polypropylene Sheets, Gels 8 (2022) 345. https://doi.org/10.3390/gels8060345.

[105] Z. Zheng, L. Zhang, Y. Ling, H. Tang, Triblock copolymers containing UCST polypeptide and poly(propylene glycol): Synthesis, thermoresponsive properties, and modification of PVA hydrogel, Eur. Polym. J. 115 (2019) 244–250. https://doi.org/10.1016/j.eurpolymj.2019.03.034.

View Article Online
DOI: 10.1039/D6MA00167J

[106] M. Xia, Y. Cheng, Z. Meng, X. Jiang, Z. Chen, P. Theato, M. Zhu, A Novel Nanocomposite Hydrogel with Precisely Tunable UCST and LCST, Macromol. Rapid Commun. 36 (2015) 477–482. https://doi.org/10.1002/marc.201400665.

[107] H. Huang, X. Qi, Y. Chen, Z. Wu, Thermo-sensitive hydrogels for delivering biotherapeutic molecules: A review, Saudi Pharmaceutical Journal 27 (2019) 990–999. https://doi.org/10.1016/j.jsps.2019.08.001.

[108] F. Andrade, M.M. Roca-Melendres, E.F. Durán-Lara, D. Rafael, S. Schwartz, Stimuli-Responsive Hydrogels for Cancer Treatment: The Role of pH, Light, Ionic Strength and Magnetic Field, Cancers (Basel). 13 (2021) 1164. https://doi.org/10.3390/cancers13051164.

[109] A. Pitto-Barry, N.P.E. Barry, Pluronic® block-copolymers in medicine: from chemical and biological versatility to rationalisation and clinical advances, Polym. Chem. 5 (2014) 3291–3297. https://doi.org/10.1039/C4PY00039K.

[110] H. Liaqat, S.F. Badshah, M.U. Minhas, K. Barkat, S.A. Khan, M.D. Hussain, M. Kazi, pH-Sensitive Hydrogels Fabricated with Hyaluronic Acid as a Polymer for Site-Specific Delivery of Mesalamine, ACS Omega 9 (2024) 28827–28840. https://doi.org/10.1021/acsomega.4c03240.

[111] Y. Wang, C. Yang, W. Zhang, X. Wang, Z. Zhao, Z. Wang, L. Zhang, Multifunctional self-healing and pH-responsive hydrogel dressing based on cationic guar gum and hyaluronic acid for on-demand drug release, Int. J. Biol. Macromol. 301 (2025) 140326. https://doi.org/10.1016/j.ijbiomac.2025.140326.

[112] I. Iftikhar, K. Barkat, S.F. Badshah, M.U. Ashraf, Y. Mehmood, I. Anjum, G.A. Shazly, A. Metouekel, Y.A. Younous, M. Bourhia, Formulation of pH responsive polymeric hydrogels for prolonged delivery of famciclovir with biosafety evaluation, Sci. Rep. 15 (2025) 18686. https://doi.org/10.1038/s41598-025-96739-4.

[113] J. He, Q. Zhou, Z. Ge, S. Jiang, J. Li, W. Feng, H. Yang, pH-Gated Switch of LCST-UCST Phase Transition of Hydrogels, Adv. Funct. Mater. 34 (2024). https://doi.org/10.1002/adfm.202404341.

[114] A.M. Rosales, S.L. Vega, F.W. DelRio, J.A. Burdick, K.S. Anseth, Hydrogels with Reversible Mechanics to Probe Dynamic Cell Microenvironments, Angew. Chem. Int. Ed. 56 (2017) 12132–12136. https://doi.org/10.1002/anie.201705684.

[115] M.P. Lutolf, J.L. Lauer-Fields, H.G. Schmoekel, A.T. Metters, F.E. Weber, G.B. Fields, J.A. Hubbell, Synthetic matrix metalloproteinase-sensitive hydrogels for the conduction of tissue regeneration: Engineering cell-invasion characteristics, Proceedings of the National Academy of Sciences 100 (2003) 5413–5418. https://doi.org/10.1073/pnas.0737381100.

[116] I.A. Marozas, J.J. Cooper-White, K.S. Anseth, Photo-induced viscoelasticity in cytocompatible hydrogel substrates, New J. Phys. 21 (2019) 045004. https://doi.org/10.1088/1367-2630/ab1309.

[117] H. Zhu, X. Yang, G.M. Genin, T.J. Lu, F. Xu, M. Lin, The relationship between thiol-acrylate photopolymerization kinetics and hydrogel mechanics: An improved model incorporating photobleaching and thiol-Michael addition, J. Mech. Behav. Biomed. Mater. 88 (2018) 160–169. https://doi.org/10.1016/j.jmbbm.2018.08.013.

[118] B.J. Carberry, V. V. Rao, K.S. Anseth, Phototunable Viscoelasticity in Hydrogels Through Thioester Exchange, Ann. Biomed. Eng. 48 (2020) 2053–2063. https://doi.org/10.1007/s10439-020-02460-w.

[119] A. Ansari, S. Bhowmik, K. Zhang, C.L. Reeves, D. Vahala, Y.S. Choi, A. Gelmi, A.N. Combes, R.S. Tuan, V.X. Truong, J.S. Forsythe, J.E. Frith, A Visible Light-Responsive Hydrogel to Study the Effect of Dynamic Tissue Stiffness on Cellular Mechanosensing, Adv. Funct. Mater. 35 (2025). https://doi.org/10.1002/adfm.202501585.

[120] M. Sheikhi, S. Vakili, N. Karimi, F. Rafiemanzelat, A. Maleki, A. Taheri, Z. Mohamadnia, A. Ramazani, 4D Printing of Self-Healing, Thermally, and Near-Infrared Light-Responsive Granular Hydrogels with Segmental Directed Movement for Soft Robotic, ACS Appl. Polym. Mater. 7 (2025) 1717–1728. https://doi.org/10.1021/acsapm.4c03518.

[121] S. Panja, D.J. Adams, Gel to gel transitions by dynamic self-assembly, Chemical Communications 55 (2019) 10154–10157. https://doi.org/10.1039/C9CC05154F.

[122] X. Li, P. Yang, Z. Zhu, Z. You, W. Zhang, T. Zhang, M. Chen, X. Zhou, Sol-Gel condensation of temperature sensitive and shape stabilized phase change materials for thermal energy storage, Thermochim. Acta 693 (2020) 178758. https://doi.org/10.1016/j.tca.2020.178758.

[123] A.D. Drozdov, J. deClaville Christiansen, Modulation of the volume phase transition temperature for multi-stimuli-responsive copolymer hydrogels, Int. J. Mech. Sci. 211 (2021) 106753. https://doi.org/10.1016/j.ijmecsci.2021.106753.

[124] V. Castrejón-Comas, N. Mataró, L. Resina, D. Zanuy, Q. Nuñez-Aulina, J. Sánchez-Morán, H. Enshaei, M. Arnau, H. Muñoz-Galán, J.C. Worch, A.P. Dove, C. Alemán, M.M. Pérez-Madrigal, Electro-responsive hyaluronic acid-based click-hydrogels for wound healing, Carbohydr. Polym. 348 (2025) 122941. https://doi.org/10.1016/j.carbpol.2024.122941.

[125] H. Zhang, S. Hua, C. He, M. Yin, J. Qin, H. Liu, H. Zhou, S. Wu, X. Yu, H. Jiang, Y. Wang, Y. Qian, Application of 4D-Printed Magnetoresponsive FOGS Hydrogel Scaffolds in Auricular Cartilage Regeneration, Adv. Healthc. Mater. 14 (2025). https://doi.org/10.1002/adhm.202404488.

[126] P. Ruidas, S.K. Dubey, S.A. Hafiz, J. Mandal, S. Mukherjee, N.N. Ghosh, R. Midya, D. Roy, D. Das, S. Singh, P. Neogi, S. Saha, U.K. Roy, S. Bhattacharyya, A. Ghosh, S. Bhattacharjee, Chiral Self-Assembly of a Pyrene-Appended Glutamylalanine Dipeptide and Its Charge Transfer Complex: Fabrication of Magneto-Responsive Hydrogels and Human Cell Imaging, Macromol. Rapid Commun. 46 (2025). https://doi.org/10.1002/marc.202400672.

[127] Q. Wang, L. Zhang, T. Wu, H. Song, W. Li, Electrohydrodynamic-printed dual-anionic hydrogel soft actuator with enhanced ionic synergy, Chem. Eng. Sci. 320 (2026) 122523. https://doi.org/10.1016/j.ces.2025.122523.

View Article Online
DOI: 10.1039/D6MA00167J

[128] K. Werengowska-Ciećwierz, M. Wiśniewski, A.P. Terzyk, S. Furmaniak, The Chemistry of Bioconjugation in Nanoparticles-Based Drug Delivery System, Advances in Condensed Matter Physics 2015 (2015) 1–27. https://doi.org/10.1155/2015/198175.

[129] R.A. Frimpong, J.Z. Hilt, Magnetic Nanoparticles in Biomedicine: Synthesis, Functionalization and Applications, Nanomedicine 5 (2010) 1401–1414. https://doi.org/10.2217/nnm.10.114.

[130] K. Saha, S.S. Agasti, C. Kim, X. Li, V.M. Rotello, Gold Nanoparticles in Chemical and Biological Sensing, Chem. Rev. 112 (2012) 2739–2779. https://doi.org/10.1021/cr2001178.

[131] M.A. Ward, T.K. Georgiou, Thermoresponsive Polymers for Biomedical Applications, Polymers (Basel). 3 (2011) 1215–1242. https://doi.org/10.3390/polym3031215.

[132] A. Galperin, T.J. Long, B.D. Ratner, Degradable, Thermo-Sensitive Poly(N-isopropyl acrylamide)-Based Scaffolds with Controlled Porosity for Tissue Engineering Applications, Biomacromolecules 11 (2010) 2583–2592. https://doi.org/10.1021/bm100521x.

[133] B.R. Twaites, C. de las Heras Alarcón, M. Lavigne, A. Saulnier, S.S. Pennadam, D. Cunliffe, D.C. Górecki, C. Alexander, Thermoresponsive polymers as gene delivery vectors: Cell viability, DNA transport and transfection studies, Journal of Controlled Release 108 (2005) 472–483. https://doi.org/10.1016/j.jconrel.2005.08.009.

[134] Y. Okuyama, R. Yoshida, K. Sakai, T. Okano, Y. Sakurai, Swelling controlled zero order and sigmoidal drug release from thermo-responsive poly(N-isopropylacrylamide-co-butyl methacrylate) hydrogel, J. Biomater. Sci. Polym. Ed. 4 (1993) 545–556. https://doi.org/10.1163/156856293X00195.

[135] J.A. Galicia, F. Cousin, E. Dubois, O. Sandre, V. Cabuil, R. Perzynski, Local structure of polymeric ferrogels, J. Magn. Magn. Mater. 323 (2011) 1211–1215.

[136] A.H. Monfared, A. Zamanian, I. Sharifi, M. Mozafari, Reversible multistimuli-responsive manganese–zinc ferrite/P(NIPAAM-AAc-AAm) core-shell nanoparticles: A programmed ferrogel system, Mater. Chem. Phys. 226 (2019) 44–50. https://doi.org/10.1016/j.matchemphys.2019.01.016.

[137] M. Häring, J. Schiller, J. Mayr, S. Grijalvo, R. Eritja, D. Díaz, Magnetic Gel Composites for Hyperthermia Cancer Therapy, Gels 1 (2015) 135–161. https://doi.org/10.3390/gels1020135.

[138] B. Li, T. Lin, Y. Lai, T. Chiu, Y. Yeh, Engineering Multiresponsive Alginate/PNIPAM/Carbon Nanotube Nanocomposite Hydrogels as On-Demand Drug Delivery Platforms, Small 21 (2025). https://doi.org/10.1002/smll.202407420.

[139] H. Iresha, T. Kobayashi, Ultrasound-triggered nicotine release from nicotine-loaded cellulose hydrogel, Ultrason. Sonochem. 78 (2021) 105710. https://doi.org/10.1016/j.ultsonch.2021.105710.

[140] Z. Li, X. Liang, Z. Qiu, Z. Liu, S. Wang, Y. Zhou, N. Li, Ion-interferential cell cycle arrest for melanoma treatment based on magnetocaloric bimetallic-ion sustained release hydrogel, Chinese Chemical Letters 35 (2024) 109592. https://doi.org/10.1016/j.cclet.2024.109592.

[141] S. Ekici, P. Ilgin, S. Yilmaz, N. Aktas, N. Sahiner, Temperature and magnetic field responsive hyaluronic acid particles with tunable physical and chemical properties, Appl. Surf. Sci. 257 (2011) 2669–2676. https://doi.org/10.1016/j.apsusc.2010.10.040.

[142] H. Chen, X. Zhang, L. Shang, Z. Su, Programmable Anisotropic Hydrogels with Localized Photothermal/Magnetic Responsive Properties, Advanced Science 9 (2022). https://doi.org/10.1002/advs.202202173.

[143] X. Ma, L. Lin, K. Peng, Q. Zheng, Y. Feng, Y. Chen, Construction and Performance Study of an Injectable Dual-Network Hydrogel Dressing with Inherent Drainage Function, ACS Appl. Mater. Interfaces 16 (2024) 59143–59155. https://doi.org/10.1021/acsami.4c09483.

[144] R. Parhi, Cross-linked hydrogel for pharmaceutical applications: A review, Adv. Pharm. Bull. 7 (2017) 515–530.

[145] K. Xu, X. Liang, P. Li, Y. Deng, X. Pei, Y. Tan, K. Zhai, P. Wang, Tough, stretchable chemically cross-linked hydrogel using core – shell polymer microspheres as cross-linking junctions, Polymer (Guildf). 118 (2017) 58–67. https://doi.org/10.1016/j.polymer.2017.04.055.

[146] E. Karada, O. Omer Barıs ß, U. Uz, D. Saraydin, Swelling equilibria and dye adsorption studies of chemically crosslinked superabsorbent acrylamide/maleic acid hydrogels, Eur. Polym. J. 38 (2002) 2133–2141.

[147] S.C. Kim, D. Klempner, K.C. Frisch, H.L. Frisch, Polyurethane interpenetrating polymer networks. V. Engineering properties of polyurethane–poly(methyl methacrylate) IPN's, J. Appl. Polym. Sci. 21 (1977) 1289–1295.

[148] Y.S. Lipatov, Polymer blends and interpenetrating polymer networks at the interface with solids, Progress in Polymer Science (Oxford) 27 (2002) 1721–1801.

[149] G. Li, J. Chen, Z. Yan, S. Wang, Y. Ke, W. Luo, H. Ma, J. Guan, Y. Long, Physical crosslinked hydrogel-derived smart windows: anti-freezing and fast thermal responsive performance, Mater. Horiz. 10 (2023) 2004–2012. https://doi.org/10.1039/D3MH00057E.

[150] C. Yin, Z.-X. Fei, J. Sun, L. Weng, X. Wang, K.-K. Yang, L.-Y. Shi, High-enthalpy biphasic phase change organogels with shape memory function based on hydrophobic association and H-bonding interaction, Chemical Engineering Journal 468 (2023) 143495. https://doi.org/10.1016/j.cej.2023.143495.

[151] Y. Fang, X. Xiong, L. Yang, W. Yang, H. Wang, Q. Wu, Q. Liu, J. Cui, Phase Change Hydrogels for Bio-Inspired Adhesion and Energy Exchange Applications, Adv. Funct. Mater. 33 (2023). https://doi.org/10.1002/adfm.202301505.

[152] T. Hu, Z. Chen, S. Zhang, J. Niu, Y. Fang, W. Yuan, W. Zhang, L. Li, Shape-stable hydrated salt phase change hydrogels for solar energy storage and conversion, J. Energy Storage 92 (2024) 112051. https://doi.org/10.1016/j.est.2024.112051.

[153] T. Zhu, W. Jiang, X. Shi, D. Sun, J. Yang, X. Qi, Y. Wang, MXene/Ag doped hydrated-salt hydrogels with excellent thermal/light energy storage, strain sensing and photothermal

View Article Online
DOI: 10.1039/D6MA00167J

antibacterial performances for intelligent human healthcare, Compos. Part A Appl. Sci. Manuf. 170 (2023) 107526. https://doi.org/10.1016/j.compositesa.2023.107526.

[154] K. Peng, J. Zhang, J. Yang, L. Lin, Q. Gan, Z. Yang, Y. Chen, C. Feng, Green Conductive Hydrogel Electrolyte with Self-Healing Ability and Temperature Adaptability for Flexible Supercapacitors, ACS Appl. Mater. Interfaces 14 (2022) 39404–39419. https://doi.org/10.1021/acsami.2c11973.

[155] L. Wu, W. Niu, Q. Wang, Q. Yang, J. Song, Q. Guo, W. Gong, X. Kang, K. Nishinari, M. Zhao, Microstructure determined the gel properties of gelatin/dextran more than the macrophase separation, Food Hydrocoll. 135 (2023) 108116. https://doi.org/10.1016/j.foodhyd.2022.108116.

[156] S. Zheng, Y. Wang, M. Li, Y. Li, Y. Zhang, Research progress on preparation and fluorescence sensing performance of layered double hydroxides-based inorganic/organic luminescent nanocomposites, Mater. Today Commun. 38 (2024) 108466. https://doi.org/10.1016/j.mtcomm.2024.108466.

[157] Y. Fang, Z. Bai, L. Yang, Q. Liu, W. Xu, J. Wei, K. Yang, Q. Wang, J. Cui, Reversible Phase Change-Induced Hardening and Softening for Conditions-Adaptive and Mechanics-Reconfigurable Applications, Adv. Funct. Mater. 34 (2024). https://doi.org/10.1002/adfm.202314353.

[158] Y. Luo, L. Zou, J. Qiao, J. Zhang, K. Liu, H. Wu, P. Lin, Y. Chen, Boron Nitride-Doped Inorganic Hydrated Salt Gels Demonstrating Superior Thermal Energy Storage and Wearability Toward High-Performance Personal Thermal Management, ACS Appl. Energy Mater. 5 (2022) 11591–11603. https://doi.org/10.1021/acsaem.2c02070.

[159] P. Cheng, Z. Tang, X. Chen, J. Xu, P. Liu, X. Zhang, G. Wang, Advanced phase change hydrogel integrating metal-organic framework for self-powered thermal management, Nano Energy 105 (2023) 108009. https://doi.org/10.1016/j.nanoen.2022.108009.

[160] M. Song, L. Wang, F. Shao, H. Xie, H. Xu, W. Yu, Thermally induced flexible phase change hydrogels for solar thermal storage and human thermal management, Chemical Engineering Journal 464 (2023) 142682. https://doi.org/10.1016/j.cej.2023.142682.

[161] K. Wang, G. Chen, S. Weng, L. Hou, D. Ye, X. Jiang, Thermo-Responsive Poly(N-isopropylacrylamide)/Hydroxypropylmethyl Cellulose Hydrogel with High Luminous Transmittance and Solar Modulation for Smart Windows, ACS Appl. Mater. Interfaces 15 (2023) 4385–4397. https://doi.org/10.1021/acsami.2c15367.

[162] A. Garcia-Garcia, S. Muñana-González, S. Lanceros-Mendez, L. Ruiz-Rubio, L.P. Alvarez, J.L. Vilas-Vilela, Biodegradable Natural Hydrogels for Tissue Engineering, Controlled Release, and Soil Remediation, Polymers (Basel). 16 (2024) 2599. https://doi.org/10.3390/polym16182599.

[163] T.A. Adjuik, S.E. Nokes, M.D. Montross, Biodegradability of bio-based and synthetic hydrogels as sustainable soil amendments: A review, J. Appl. Polym. Sci. 140 (2023). https://doi.org/10.1002/app.53655.

View Article Online
DOI: 10.1039/D6MA00167J

[164] A. Yarahmadi, B. Dousti, M. Karami-Khorramabadi, H. Afkhami, Materials based on biodegradable polymers chitosan/gelatin: a review of potential applications, Front. Bioeng. Biotechnol. 12 (2024). https://doi.org/10.3389/fbioe.2024.1397668.

[165] L. Lujan, M.L. Goñi, R.E. Martini, Cellulose–Chitosan Biodegradable Materials for Insulating Applications, ACS Sustain. Chem. Eng. 10 (2022) 12000–12008. https://doi.org/10.1021/acssuschemeng.2c03538.

[166] J. Lin, G. Jiao, A.J. Scott, C.C. Xu, G. Gagnon, A. Kermanshahi-pour, Green synthesis of self-assembly, self-healing, and injectable polyelectrolyte complex hydrogels using chitosan, sulphated polysaccharides, hydrolyzed collagen and nanocellulose, Int. J. Biol. Macromol. 288 (2025) 138566. https://doi.org/10.1016/j.ijbiomac.2024.138566.

[167] N. Srikhao, S. Theerakulpisut, P. Chindaprasirt, M. Okhawilai, R. Narain, P. Kasemsiri, Green synthesis of nano silver-embedded carboxymethyl starch waste/poly vinyl alcohol hydrogel with photothermal sterilization and pH-responsive behavior, Int. J. Biol. Macromol. 242 (2023) 125118. https://doi.org/10.1016/j.ijbiomac.2023.125118.

[168] M.J. Ansari, R.R. Rajendran, S. Mohanto, U. Agarwal, K. Panda, K. Dhotre, R. Manne, A. Deepak, A. Zafar, M. Yasir, S. Pramanik, Poly(N-isopropylacrylamide)-Based Hydrogels for Biomedical Applications: A Review of the State-of-the-Art, Gels 8 (2022) 454. https://doi.org/10.3390/gels8070454.

[169] M.A. Haq, Y. Su, D. Wang, Mechanical properties of PNIPAM based hydrogels: A review, Materials Science and Engineering: C 70 (2017) 842–855. https://doi.org/10.1016/j.msec.2016.09.081.

[170] Y. Li, J. Luo, G. Xie, D. Zhu, C. Zhao, X. Zhang, M. Liu, Y. Wu, Y. Guo, W. Yu, Recent Progress on Regulating the LCST of PNIPAM-Based Thermochromic Materials, ACS Appl. Polym. Mater. 7 (2025) 1–11. https://doi.org/10.1021/acsapm.4c03406.

[171] C. He, H. Chen, Z. Huang, Z. Song, X. Liu, S. Gao, J. Zhang, X. Zhang, Y. Jiang, H. Gao, Review on recent advances in shape-stable phase change hydrogels for thermal energy storage applications, J. Energy Storage 128 (2025) 117245. https://doi.org/10.1016/j.est.2025.117245.

[172] I. Krakovský, L. Hanyková, J. Šťastná, Phase transition in polymer hydrogels investigated by swelling, DSC, FTIR and NMR, J. Therm. Anal. Calorim. 150 (2025) 1245–1262. https://doi.org/10.1007/s10973-024-13380-5.

[173] A. Okudan, A. Altay, Investigation of the Effects of Different Hydrophilic and Hydrophobic Comonomers on the Volume Phase Transition Temperatures and Thermal Properties of N-Isopropylacrylamide-Based Hydrogels, Int. J. Polym. Sci. 2019 (2019) 1–12. https://doi.org/10.1155/2019/7324181.

[174] M. Salmerón Sánchez, G.G. Ferrer, M. Monleón Pradas, J.L. Gómez Ribelles, Influence of the Hydrophobic Phase on the Thermal Transitions of Water Sorbed in a Polymer Hydrogel Based

on Interpenetration of a Hydrophilic and a Hydrophobic Network, Macromolecules 36 (2003) 860–866. https://doi.org/10.1021/ma021069p.

[175] C. Yin, Z.-X. Fei, J. Sun, L. Weng, X. Wang, K.-K. Yang, L.-Y. Shi, High-enthalpy biphasic phase change organogels with shape memory function based on hydrophobic association and H-bonding interaction, Chemical Engineering Journal 468 (2023) 143495. https://doi.org/10.1016/j.cej.2023.143495.

[176] D. Tan, F. Meng, Y. Ni, W. Sun, Q. Liu, X. Wang, Z. Shi, Q. Zhao, Y. Lei, S. Luan, L. Xue, Robust and smart underwater adhesion of hydrophobic hydrogel by phase change, Chemical Engineering Journal 471 (2023) 144625. https://doi.org/10.1016/j.cej.2023.144625.

[177] Md.H. Zahir, S.A. Mohamed, R. Saidur, F.A. Al-Sulaiman, Supercooling of phase-change materials and the techniques used to mitigate the phenomenon, Appl. Energy 240 (2019) 793–817. https://doi.org/10.1016/j.apenergy.2019.02.045.

[178] B. Adhikari, A. Banerjee, Short peptide based hydrogels: incorporation of graphene into the hydrogel, Soft Matter 7 (2011) 9259. https://doi.org/10.1039/c1sm06330h.

[179] J.G. Um, S. Habibpour, Y.-S. Jun, A. Elkamel, A. Yu, Development of π–π Interaction-Induced Functionalized Graphene Oxide on Mechanical and Anticorrosive Properties of Reinforced Polyurethane Composites, Ind. Eng. Chem. Res. 59 (2020) 3617–3628. https://doi.org/10.1021/acs.iecr.9b06755.

[180] F. Li, Y. Zhu, B. You, D. Zhao, Q. Ruan, Y. Zeng, C. Ding, Smart Hydrogels Co-switched by Hydrogen Bonds and π–π Stacking for Continuously Regulated Controlled-Release System, Adv. Funct. Mater. 20 (2010) 669–676. https://doi.org/10.1002/adfm.200901245.

[181] D. Lu, Y. Mo, S. Sun, Q. Wang, Z. Wu, W. Wang, M. Zhu, Mechanically Reinforced Nanocomposite Hydrogels and Advanced Applications in Biosensing and Bioelectronics, Chemistry of Materials 37 (2025) 3871–3902. https://doi.org/10.1021/acs.chemmater.5c00498.

[182] H. He, X. Shen, Z. Nie, Engineering interactions between nanoparticles using polymers, Prog. Polym. Sci. 143 (2023) 101710. https://doi.org/10.1016/j.progpolymsci.2023.101710.

[183] T. Lo Wong, C. Vallés, A. Nasser, C. Abeykoon, Effects of boron-nitride-based nanomaterials on the thermal properties of composite organic phase change materials: A state-of-the-art review, Renewable and Sustainable Energy Reviews 187 (2023) 113730. https://doi.org/10.1016/j.rser.2023.113730.

[184] S.Y. Zheng, H. Ding, J. Qian, J. Yin, Z.L. Wu, Y. Song, Q. Zheng, Metal-Coordination Complexes Mediated Physical Hydrogels with High Toughness, Stick–Slip Tearing Behavior, and Good Processability, Macromolecules 49 (2016) 9637–9646. https://doi.org/10.1021/acs.macromol.6b02150.

[185] H. Xu, L. Jiang, A. Yuan, Y. Lei, Z. Wei, Y. Wang, J. Lei, Thermally-stable, solid-solid phase change materials based on dynamic metal-ligand coordination for efficient thermal energy

View Article Online
DOI: 10.1039/D6MA00167J

storage, Chemical Engineering Journal 421 (2021) 129833. https://doi.org/10.1016/j.cej.2021.129833.

[186] J. Qiu, D. Huo, J. Xue, G. Zhu, H. Liu, Y. Xia, Encapsulation of a Phase-Change Material in Nanocapsules with a Well-Defined Hole in the Wall for the Controlled Release of Drugs, Angew. Chem. Int. Ed. 58 (2019) 10606–10611. https://doi.org/10.1002/anie.201904549.

[187] P. Suryavanshi, S. Mahajan, S.K. Banerjee, K. Seth, S. Banerjee, Synthesis and characterization of a pH/temperature-dual responsive hydrogel with promising biocompatibility features for stimuli-responsive 5-FU delivery, J. Mater. Chem. B 12 (2024) 5098–5110. https://doi.org/10.1039/D4TB00168K.

[188] C. Liu, C. Ruan, R. Shi, B.-P. Jiang, S. Ji, X.-C. Shen, A near infrared-modulated thermosensitive hydrogel for stabilization of indocyanine green and combinatorial anticancer phototherapy, Biomater. Sci. 7 (2019) 1705–1715. https://doi.org/10.1039/C8BM01541D.

[189] J. Zhang, W. Lin, L. Yang, A. Zhang, Y. Zhang, J. Liu, J. Liu, Injectable and pH-responsive self-assembled peptide hydrogel for promoted tumor cell uptake and enhanced cancer chemotherapy, Biomater. Sci. 10 (2022) 854–862. https://doi.org/10.1039/D1BM01788H.

[190] O. Goncharuk, Y. Samchenko, D. Sternik, L. Kernosenko, T. Poltorats'ka, N. Pasmurtseva, M. Abramov, E. Pakhlov, A. Derylo-Marczewska, Thermosensitive hydrogel nanocomposites with magnetic laponite nanoparticles, Appl. Nanosci. 10 (2020) 4559–4569. https://doi.org/10.1007/s13204-020-01388-w.

[191] D. Britton, D. Almanzar, Y. Xiao, H.-W. Shih, J. Legocki, P. Rabbani, J.K. Montclare, Exosome Loaded Protein Hydrogel for Enhanced Gelation Kinetics and Wound Healing, ACS Appl. Bio Mater. 7 (2024) 5992–6000. https://doi.org/10.1021/acsabm.4c00569.

[192] X. Ma, L. Lin, K. Peng, Q. Zheng, Y. Feng, Y. Chen, Construction and Performance Study of an Injectable Dual-Network Hydrogel Dressing with Inherent Drainage Function, ACS Appl. Mater. Interfaces 16 (2024) 59143–59155. https://doi.org/10.1021/acsami.4c09483.

[193] Y. Chen, L. Chang, Z. Zhang, M. Zhou, Y. Gao, Y. Wang, Y. Liu, J. Qin, Biodegradable pectin-based thermo-responsive composite GO/hydrogel with mussel inspired tissue adhesion for NIR enhanced burn wound healing, Chemical Engineering Journal 480 (2024) 148067. https://doi.org/10.1016/j.cej.2023.148067.

[194] Y. Li, X. Wu, Y. Liao, H. Shao, Y. Chen, W. Yang, P. Lv, Y. Zhou, C. Tang, Multifunctional inorganic-organic phase-change hydrogel with excellent light-to-thermal energy conversion and strain sensing capabilities for medical thermotherapy and monitoring, Polymer (Guildf). 305 (2024) 127171. https://doi.org/10.1016/j.polymer.2024.127171.

[195] H. Fu, K. Xue, Y. Zhang, M. Xiao, K. Wu, L. Shi, C. Zhu, Thermoresponsive Hydrogel-Enabled Thermostatic Photothermal Therapy for Enhanced Healing of Bacteria-Infected Wounds, Advanced Science 10 (2023). https://doi.org/10.1002/advs.202206865.

View Article Online
DOI: 10.1039/D6MA00167J

[196] M. Jia, J. Zhang, Thermoresponsive PEDOT:PSS/PNIPAM conductive hydrogels as wearable resistive sensors for breathing pattern detection, Polym. J. 54 (2022) 793–801. https://doi.org/10.1038/s41428-022-00626-y.

[197] Q. Pang, H. Hu, H. Zhang, B. Qiao, L. Ma, Temperature-Responsive Ionic Conductive Hydrogel for Strain and Temperature Sensors, ACS Appl. Mater. Interfaces 14 (2022) 26536–26547. https://doi.org/10.1021/acsami.2c06952.

[198] J. Yang, W. Huang, K. Peng, Z. Cheng, L. Lin, J. Yuan, Y. Sun, N. Cho, Y. Chen, Versatile Agar-Zwitterion Hybrid Hydrogels for Temperature Self-Sensing and Electro-Responsive Actuation, Adv. Funct. Mater. 34 (2024). https://doi.org/10.1002/adfm.202313725.

[199] B. Zhou, W. Yuan, Tunable thermoresponsive and stretchable hydrogel sensor based on hydroxypropyl cellulose for human motion/health detection, visual signal transmission and information encryption, Carbohydr. Polym. 343 (2024) 122497. https://doi.org/10.1016/j.carbpol.2024.122497.

[200] Z. Chen, J. Liu, Y. Chen, X. Zheng, H. Liu, H. Li, Multiple-Stimuli-Responsive and Cellulose Conductive Ionic Hydrogel for Smart Wearable Devices and Thermal Actuators, ACS Appl. Mater. Interfaces 13 (2021) 1353–1366. https://doi.org/10.1021/acsami.0c16719.

[201] Y. Li, Y. Gu, S. Qian, Y. Pang, A. Yu, S. Zheng, W. Xia, Y. Liao, B. Liu, S. Liu, Q. Zhao, A stretchable, ionic conductive, and adhesive patch electrode with ultra-low on-skin impedance for electrophysiological signal recording, Science China Information Sciences 68 (2025) 129402. https://doi.org/10.1007/s11432-024-4246-9.

[202] Q. Chang, Y. He, Y. Liu, W. Zhong, Q. Wang, F. Lu, M. Xing, Protein Gel Phase Transition: Toward Superiorly Transparent and Hysteresis-Free Wearable Electronics, Adv. Funct. Mater. 30 (2020). https://doi.org/10.1002/adfm.201910080.

[203] J. Li, J. Cao, R. Bian, R. Wan, X. Zhu, B. Lu, G. Gu, Multimaterial cryogenic printing of three-dimensional soft hydrogel machines, Nat. Commun. 16 (2025) 185. https://doi.org/10.1038/s41467-024-55323-6.

[204] D. Tan, F. Meng, Y. Ni, W. Sun, Q. Liu, X. Wang, Z. Shi, Q. Zhao, Y. Lei, S. Luan, L. Xue, Robust and smart underwater adhesion of hydrophobic hydrogel by phase change, Chemical Engineering Journal 471 (2023) 144625. https://doi.org/10.1016/j.cej.2023.144625.

[205] J. Li, Q. Ma, Y. Xu, M. Yang, Q. Wu, F. Wang, P. Sun, Highly Bidirectional Bendable Actuator Engineered by LCST–UCST Bilayer Hydrogel with Enhanced Interface, ACS Appl. Mater. Interfaces 12 (2020) 55290–55298. https://doi.org/10.1021/acsami.0c17085.

[206] J. Zheng, P. Xiao, X. Le, W. Lu, P. Théato, C. Ma, B. Du, J. Zhang, Y. Huang, T. Chen, Mimosa inspired bilayer hydrogel actuator functioning in multi-environments, J. Mater. Chem. C Mater. 6 (2018) 1320–1327. https://doi.org/10.1039/C7TC04879C.

[207] L.K. Hill, M. Meleties, P. Katyal, X. Xie, E. Delgado-Fukushima, T. Jihad, C.-F. Liu, S. O’Neill, R.S. Tu, P.D. Renfrew, R. Bonneau, Y.Z. Wadghiri, J.K. Montclare, Thermoresponsive Protein-

Engineered Coiled-Coil Hydrogel for Sustained Small Molecule Release, Biomacromolecules 20 (2019) 3340–3351. https://doi.org/10.1021/acs.biomac.9b00107.

[208] Y. Shirasaki, J. Tanaka, H. Makazu, K. Tashiro, S. Shoji, S. Tsukita, T. Funatsu, On-Chip Cell Sorting System Using Laser-Induced Heating of a Thermoreversible Gelation Polymer to Control Flow, Anal. Chem. 78 (2006) 695–701. https://doi.org/10.1021/ac0511041.

[209] T.R. Hoare, D.S. Kohane, Hydrogels in drug delivery: Progress and challenges, Polymer (Guildf). 49 (2008) 1993–2007. https://doi.org/10.1016/j.polymer.2008.01.027.

[210] J.G. Choi, G.M. Spinks, S.J. Kim, Mode shifting shape memory polymer and hydrogel composite fiber actuators for soft robots, Sens. Actuators A Phys. 342 (2022) 113619. https://doi.org/10.1016/j.sna.2022.113619.

[211] S. Qi, W. Yuan, Robust and adhesive double-solvent phase change hydrogel with reversible transparency and shape memory for thermal management and information encryption, Chemical Engineering Journal 473 (2023) 145329. https://doi.org/10.1016/j.cej.2023.145329.

[212] B. Zhou, W. Yuan, Tunable thermoresponsive and stretchable hydrogel sensor based on hydroxypropyl cellulose for human motion/health detection, visual signal transmission and information encryption, Carbohydr. Polym. 343 (2024) 122497. https://doi.org/10.1016/j.carbpol.2024.122497.

[213] Z. Yu, Y. Ma, L. Mao, Y. Lian, Y. Xiao, Z. Chen, Y. Zhang, Bidirectional optical response hydrogel with adjustable human comfort temperature for smart windows, Mater. Horiz. 11 (2024) 207–216. https://doi.org/10.1039/D3MH01376F.

[214] H. Chen, X. Zhang, L. Shang, Z. Su, Programmable Anisotropic Hydrogels with Localized Photothermal/Magnetic Responsive Properties, Advanced Science 9 (2022). https://doi.org/10.1002/advs.202202173.

[215] M. Kim, S. Hong, J.J. Park, Y. Jung, S.H. Choi, C. Cho, I. Ha, P. Won, C. Majidi, S.H. Ko, A Gradient Stiffness-Programmed Circuit Board by Spatially Controlled Phase-Transition of Supercooled Hydrogel for Stretchable Electronics Integration, Advanced Materials 36 (2024). https://doi.org/10.1002/adma.202313344.

[216] L. Zou, Y. Luo, J. Zhang, X. Sheng, Y. Chen, P. Lin, Phase change material gel particles with suitable size and superior thermophysical properties towards highly efficient thermal management of miniature electronic components, J. Energy Storage 60 (2023) 106590. https://doi.org/10.1016/j.est.2022.106590.

[217] J. Wang, J. Wang, Z. Sheng, R. Du, L. Yan, X. Zhang, Solid–Liquid–Vapor Triphase Gel, Langmuir 37 (2021) 13501–13511. https://doi.org/10.1021/acs.langmuir.1c02333.

[218] X. Zhao, L.-M. Peng, Y. Chen, X.-J. Zha, W.-D. Li, L. Bai, K. Ke, R.-Y. Bao, M.-B. Yang, W. Yang, Phase change mediated mechanically transformative dynamic gel for intelligent control of versatile devices, Mater. Horiz. 8 (2021) 1230–1241. https://doi.org/10.1039/D0MH02069A.

[219] P. Cheng, Z. Tang, X. Chen, J. Xu, P. Liu, X. Zhang, G. Wang, Advanced phase change hydrogel integrating metal-organic framework for self-powered thermal management, Nano Energy 105 (2023) 108009. https://doi.org/10.1016/j.nanoen.2022.108009.

[220] X. Shen, F. Wang, Z. Mao, H. Xu, B. Wang, X. Sui, X. Feng, Biphasic organohydrogels based on phase change materials with excellent thermostability for thermal management applications, Chemical Engineering Journal 416 (2021) 129181. https://doi.org/10.1016/j.cej.2021.129181.

[221] J. Yang, W. Yu, C. Liu, H. Xie, H. Xu, Phase change mediated graphene hydrogel-based thermal interface material with low thermal contact resistance for thermal management, Compos. Sci. Technol. 219 (2022) 109223. https://doi.org/10.1016/j.compscitech.2021.109223.

[222] M. Wu, T. Li, Q. He, R. Du, R. Wang, Thermally conductive and form-stable phase change composite for building thermal management, Energy 239 (2022) 121938. https://doi.org/10.1016/j.energy.2021.121938.

[223] C. Feng, P. Yang, H. Liu, M. Mao, Y. Liu, T. Xue, J. Fu, T. Cheng, X. Hu, H.J. Fan, K. Liu, Bilayer porous polymer for efficient passive building cooling, Nano Energy 85 (2021) 105971. https://doi.org/10.1016/j.nanoen.2021.105971.

[224] G. Sang, H. Zeng, Z. Guo, H. Cui, Y. Zhang, X. Cui, L. Zhang, W. Han, Studies of eutectic hydrated salt/polymer hydrogel composite as form-stable phase change material for building thermal energy storage, Journal of Building Engineering 59 (2022) 105010. https://doi.org/10.1016/j.jobe.2022.105010.

[225] F. Shao, L. Wang, R. Luo, W. Yu, H. Xie, Shape-Stable Hybrid Emulsion Gel with Sodium Acetate Trihydrate and Paraffin Wax for Efficient Solar Energy Storage and Building Thermal Management, ACS Appl. Mater. Interfaces 15 (2023) 38474–38484. https://doi.org/10.1021/acsami.3c07429.

[226] T.R. Hoare, D.S. Kohane, Hydrogels in drug delivery: Progress and challenges, Polymer (Guildf). 49 (2008) 1993–2007.

[227] K. Chen, F. Wang, Y. Hu, M. Liu, P. Liu, Y. Yu, Q. Feng, X. Xiao, Highly Stretchable, Sensitive, and Durable Ag/Tannic Acid@Graphene Oxide-Composite Hydrogel for Wearable Strain Sensors, ACS Appl. Polym. Mater. 4 (2022) 2036–2046.

[228] X. Li, R. Xu, C. Xie, Z. Ge, B. Gao, C.T. Lim, Microscale Architectures for Intelligent Soft Robotics: From Functional Microneedles to Biointegrated Wearable Systems, Nanomicro Lett. 18 (2026) 179. https://doi.org/10.1007/s40820-025-02026-2.

[229] A.K. Yetisen, N. Jiang, A. Fallahi, Y. Montelongo, G.U. Ruiz-Esparza, A. Tamayol, Y.S. Zhang, I. Mahmood, S. Yang, K.S. Kim, H. Butt, A. Khademhosseini, S. Yun, Glucose-Sensitive Hydrogel Optical Fibers Functionalized with Phenylboronic Acid, Advanced Materials 29 (2017). https://doi.org/10.1002/adma.201606380.

[230] F. Wang, D. Pang, X. Liu, M. Liu, W. Du, Y. Zhang, X. Cheng, Progress in application of phase-change materials to cooling clothing, J. Energy Storage 60 (2023) 106606. https://doi.org/10.1016/j.est.2023.106606.

View Article Online
DOI: 10.1039/D6MA00167J

[231] S. Awasthi, A. Srivastava, D. Kumar, S.K. Pandey, N.M. Mubarak, M.H. Dehghani, K. Ansari, An insight into the toxicological impacts of carbon nanotubes (CNTs) on human health: A review, Environmental Advances 18 (2024) 100601. https://doi.org/10.1016/j.envadv.2024.100601.

[232] K.M. Ainslie, T.A. Desai, Microfabricated implants for applications in therapeutic delivery, tissue engineering, and biosensing, Lab Chip 8 (2008) 1864. https://doi.org/10.1039/b806446f.

[233] S. Capuani, G. Malgir, C.Y.X. Chua, A. Grattoni, Advanced strategies to thwart foreign body response to implantable devices, Bioeng. Transl. Med. 7 (2022). https://doi.org/10.1002/btm2.10300.

[234] S. Khare, S. Kumar, P. Urmaliya, S. Yadav, Recent advancement and applications of green hydrogels: Revolutionizing biomedicine and environmental sustainability, Results Chem. 18 (2025) 102857. https://doi.org/10.1016/j.rechem.2025.102857.

[235] R.W. Nurhayati, R.D. Cahyo, K. Alawiyah, G. Pratama, E. Agustina, R.D. Antarianto, A.R. Prijanti, W. Mubarok, A.J. Rahyussalim, Development of double-layered alginate-chitosan hydrogels for human stem cell microencapsulation, in: 2019: p. 020004. https://doi.org/10.1063/1.5139324.

[236] T.A. Khan, A.K. Azad, S. Fuloria, A. Nawaz, V. Subramaniyan, M. Akhlaq, M. Safdar, K. V. Sathasivam, M. Sekar, O. Porwal, D.U. Meenakshi, R. Malviya, M.M. Miret, A. Mendiratta, N.K. Fuloria, Chitosan-Coated 5-Fluorouracil Incorporated Emulsions as Transdermal Drug Delivery Matrices, Polymers (Basel). 13 (2021) 3345. https://doi.org/10.3390/polym13193345.

View Article Online
DOI: 10.1039/D6MA00167J

Data Availability Statement

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