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REVIEW PAPER

3D printing of phase change hydrogels

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Abstract. Phase change materials (PCM) can change from solid to liquid at a constant temperature, allowing them to retain more heat than other materials. 3D printing of PCM with hydrogels opens up new manufacturing possibilities by increasing productivity and functional diversity. This article reviews recent advances in phase change hydrogel 3D printing, focusing on metal-infused hydrogels, graphene-based, bio-based PCM, and chitosan-based materials. In particular, a new 3D printing method was developed, in which rheology was modulated using carbomers to produce a variety of functional hydrogels, including magnetic, dual-network, thermosensitive, and other types. Highly loaded PCM/cellulose nanofiber (CNF) ink and phase change organic hydrogel (PCOH) are new options for such encapsulation and thermal stability. This article discusses how hydrogels resemble the extracellular matrix and can be used in bioprinting to support basic biological processes. Despite all the progress, more research is needed to address issues such as the flexibility of PCM and achieving high energy storage density. This review article highlights how hydrogels and 3D printing can have a revolutionary impact. Future directions and potential applications in the field of flexible electronics, responsive drug delivery systems, and tissue engineering are covered in this article.

Keywords: 3D printing, phase change materials, hydrogels, applications.

Classification numbers: 2.9.1, 4.10.4, 5.9.3.

TABLE OF ABBREVIATIONS

CAD	Computer-aided design	FDM	Fused deposition modeling
CNF	Cellulose nanofiber	FPCMs	Flexible phase change materials
DIW	Direct ink writing	GelMA	Gelatin methacrylate
DLP	Digital light processing	LDW	Laser direct writing
DPDH	Disodium phosphate dodecahydrate	PAM	Polyacrylamide
ECM	Extracellular matrix	PCH	Phase-change hydrogels

PCM	Phase change materials	PNIPAM	Poly *N*-isopropyl acrylamide
PCOH	Phase change organic hydrogel	PPO	Polypropylene oxide
PEG	Polyethylene glycol	SLA	Stereolithography
PEGDA	Poly(ethylene glycol) diacrylate	SLS	Selective laser sintering
PEGTA	Poly(ethylene glycol)-tetra-acrylate	STL	Standard tessellation language
PEO	Polyethylene oxide	TPP/2PP	Two-photon polymerization
PLED	Polymer light-emitting devices		

1. INTRODUCTION

Phase-change materials (PCMs) can change from solid to liquid and back at a consistent temperature, releasing or absorbing energy. They have a high heat storage capacity compared to traditional materials. 3D printing, a technology for creating 3D structures from computer designs, is versatile and promises to improve manufacturing efficiency. Current 3D printing materials include bio-based PCMs, polymeric PCMs, Chitosan, graphene-based materials, hydrogels, and metals [1, 2]. A new 3D printing method using carbomer as a rheology modifier has been developed for creating different types of functional hydrogels. This method is efficient and allows for the direct ink writing of hydrogels with various properties, such as double network hydrogels, magnetic hydrogels, temperature-sensitive hydrogels, ionogels, micro-nano gels, and bio-gels, poly *N*-isopropyl acrylamide (PNIPAM) and disodium phosphate dodecahydrate, (DPDH)-based light and thermoresponsive hydrogels [3-5]. A high-loading PCM/cellulose nanofibrils (CNF) ink, stabilized with CNF gel, enables 3D printing of a robust composite monolith with excellent thermal stability. Ice templating and freeze drying yield a tailored PCM/CNF structure [6]. Polymer hydrogels are water-rich 3D networks that can be used as ink materials to create 3D porous scaffolds. These hydrogels mimic the extracellular matrix (ECM) and influence the fate of cells. Additionally, they support processes like matrix remodeling, cell migration, and cell adhesion in a 3D environment, which are essential for the normal development of functional tissues [7]. In contrast, there have been notable achievements in creating hydrogels for 3D printing, like those made from alginate and gelatin. A key limitation is the need for more diverse printable hydrogel systems. This lack hinders the rapid progress of this technology [8]. Current 3D printing can use inks like chitosan-based hydrogels for tissue engineering and drug delivery applications. Chitosan hydrogels can be formed through physical or chemical crosslinking, with physical gels responding to stimuli like pH or temperature for the sol-gel transition [9, 10]. An anisotropic structure with homogeneous stimuli or a homogeneous structure with anisotropic stimuli is needed to produce actuation movements in hydrogels. The latter is preferred for convenience, often created through ion-inject printing, or sacrificial printing. 3D printing, or additive manufacturing, allows rapid design and fabrication of intricate 3D structures [11]. 4D printing integrates 3D printing with smart materials like shape memory polymers and hydrogels. In hydrogel-based 4D printing, internal stress triggered by stimuli induces shape transformation. This method creates pure or single-component hydrogels with internal stress through inhomogeneous reactions or crosslinking mechanisms [12]. Various hydrogel types, like UV cross-linkable and thermoresponsive, are crucial in bioprinting for achieving intricate 3D structures resembling natural environments. Thermoresponsive hydrogels vary in mechanical and biological properties based on origin, gelation mechanism, and polymer chain structure [13]. Hydrogels, particularly gelatin methacrylate (GelMA) nanofibrous hydrogels with fatty acid/aspirin (ASP) capsules, show promise for flexible phase change materials (FPCMs). Combining GelMA with temperature-

responsive phase change gels and polyacrylamide (PAM) glycerol hydrogels addresses challenges like solid rigidity and melt leakage in phase change materials. However, achieving flexibility and high energy storage density remains challenging [14]. A proof-of-concept study developed a 3D-printed thermo-responsive hydrogel with double networks for a thermal skin-like sensor. The material exhibited phase transition with temperature, enabling 3D printing of microstructures and capacitance-temperature responses, notably in grid structures with magnified geometric area changes [15].

This review aims to provide a thorough, authoritative, critical, and easily understandable examination that caters to the interests of the chemistry and materials science communities. Using 3D printing in hydrogels allows for the amalgamation of the benefits derived from additive manufacturing and hydrogel materials. The review explores various aspects, including an overview of 3D printing technologies, the integration of polymer-phase change materials, and the comprehensive characterization of PCM-polymer composites in terms of their thermal, light, and mechanical properties. Additionally, the review delves into the diverse applications of these technologies.

2. OVERVIEW OF 3D PRINTING TECHNOLOGIES

Additive manufacturing (AM), also called 3D printing, allows for the creation of products with complex geometries that are not achievable with traditional methods. A few areas where 3D printing has advanced recently to produce functional components are electronics, electrochemistry, energy storage, catalysts, thermal management, aerospace, healthcare monitoring, the food sector, sensors, and robots. Computer-aided design (CAD) generates a 3D model and transforms it into an STL (Standard Tessellation Language) file, making drawing custom parts simple [16, 17]. This STL format represents the component as a three-dimensional mesh divided into several bound 2D layers. A 3D printer is then used to create the component, automatically depositing ink onto a substrate layer by layer [10]. The conventional 3D printing technologies include inkjet printing material jetting, poly jet, binder jetting, sheet lamination, selective laser sintering (SLS), filament extrusion-based systems (fused deposition modeling (FDM), two-photon polymerization (2PP), laser direct writing (LDW); laser sintered powder materials, extruded colloidal suspension, and direct ink writing (DIW) [18, 19]. The three most widely used 3D printing technologies are light-based 3D printing, extrusion, and inkjet (Figure 1).

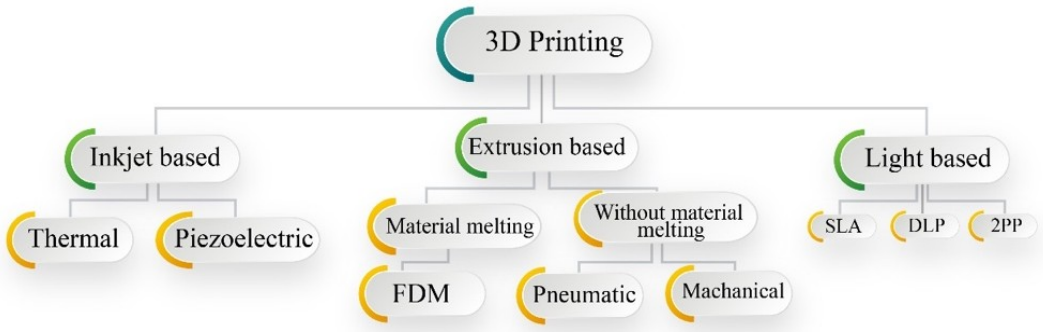

Figure 1. Schematic Overview of 3D printing technologies.

2.1. Inkjet based 3D printing

Thermal and piezoelectric inkjet 3D printing are the two main categories for inkjet-based powder-bed deposition techniques. Thermal inkjet 3D printing uses a heating element as a thin film resistor. In Table 1, the nearby ink is vaporized into bubbles using an electrical pulse that flows through this resistor [20]. The print head of the thermal printers is heated electrically, creating pressure pulses that push droplets out of the nozzle [21]. When piezoelectric inkjet 3D printing, the ink chamber's volume changes suddenly when an external voltage is applied to a piezoelectric transducer. Due to the pressure wave created by this volumetric change, ink droplets are ejected from the nozzle [22]. In addition to making it easier to deposit primary and/or stem cells at the necessary density, inkjet bioprinting preserves excellent cell viability and function even after printing. Although these characteristics show that inkjet-based 3D bioprinting can produce functional structures, resolving the above issues will turn this method into a priceless tissue engineering tool.

Table 1. Typical materials and examples of applications for 3D printing using inkjet technology [20].

Types	Materials	Applications
Monomers, Oligomers and Polymers	Hybrid polymers [e.g., poly(3,4-ethylene-dioxythiophene), poly(pyrrole), polyaniline, and poly(p-phenylene vinylene)	Transistors, displays, polymer light-emitting devices (PLED).
Metal and metal oxide, carbon materials	Graphene, carbon nanotubes, and carbon black; Silver and gold nanoparticle dispersions; Silver and gold precursor solutions	3D structures, radio-frequency identification (RFID), solar cells, and flexible electronics.
Biomaterials	Cells and biomolecules (such as proteins and DNA)	Regenerative medicine, biochips, biomarkers, biosensors, and immunoassay tests.

2.2. Extrusion based 3D printing

Extrusion-based 3D printing is one of the most widely used printing techniques. Although this technology requires materials with certain printability properties, it has the advantages of being widely adopted, being simple to use, and providing precise printing of complicated shapes through CAD and solidification procedures [23]. Extrusion-based 3D printing has moved from simple single-component or material printing to more intricate multi-material printing within the last ten years. The change has been made easier by printers, custom and open-source software, and the continuous advancement of 3D printing technology, which includes the capacity to use several print heads together [24]. Extrusion printing is a process that involves pushing material through a nozzle and onto a surface to create structures. Three-dimensional structures are made by continuously depositing material layer by layer, depending on whether the nozzle is moved above or below the stage, which directs the extruded material. It is possible to print photo-curable hydrogel-forming polymers onto an illuminated surface, and when light strikes them, they will solidify into hydrogels. Certain polymers can undergo direct photocuring, provided the right photoinitiator is used [25].

2.3. Light-based 3D printing

Technologies for 3D printing offer a high degree of material structure control. Thermoplastic is extruded from a nozzle manipulated in the xy-plane during FDM printing to construct a three-dimensional object layer by layer. Hydrogels have been printed using modified iterations of these extrusion-based printers; however, their resolution and speed are constrained [26]. An object may be formed using light-based 3D printing techniques more quickly and with finer spatial resolution than FDM printers. These printers use photopolymerization to initiate the process of a liquid polymerizing into a solid. This is achieved through digital light processing, or DLP, which projects a two-dimensional light pattern into a photo cross-linkable liquid or by raster-scanning a laser beam (stereolithography, Figure 2(b)) [27]. Still, few formulas and techniques are available for printing hydrogels using light-based printers, even though they hold great promise [28]. With light-based 3D printing, as opposed to inkjet and extrusion 3D printing, photopolymerization is induced, a photosensitive polymer ink is hardened, and layer-by-layer printing is made possible by an irradiation light source. Nozzles are not necessary with this method. The three light-based 3D printing technologies that are most commonly used are DLP, stereolithography (SLA), and 2PP (Figure 2(f)) [29]. The main differences between these 3D printing technologies are the light source and imaging technology, but the control and stepping systems remain the same [30]. The fundamentals and characteristics of DLP printing technology are introduced. Subsequently, Figure 2(c) illustrates the main applications of DLP-based 3D printing in PCM [31, 32].

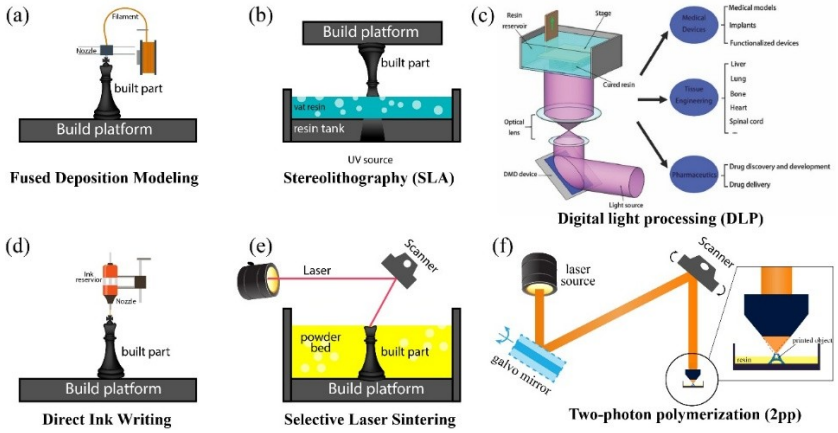

Figure 2. An overview of the most widely used 3D printing techniques based on phase change materials, (a) FDM, (b) SLA, (c) DLP [31], (d) DIW, (e) SLS [29], and (f) 2PP.

3. POLYMER PHASE CHANGE COMPOSITE HYDROGEL

PCMs show significant promise for controlling ambient temperature when they have the proper phase transition temperatures. In this review, a range of phase-change organohydrogels (PCOHs) for usage at room temperature by combining polyacrylamide (PAM) glycerol hydrogels with phase-change hydrated salts (DPDH) in an easy-to-follow one-step in situ polymerization process. Further, the economical and eco-friendly DPDH-hydrated salts (sodium dodecyl sulfate PCMs) can be incorporated into three-dimensional (3D) network structures to prevent drying [4]. PNIPAM [33], a crosslinker polyethylene glycol (PEG) dimethacrylate, poly(dimethyl siloxane) [34], and thermogelling chitosan-gelatin (CG) hydrogel, an extraordinary bioprinting ink, are smart hydrogels that serve as the main phase change material.

Figure 2 shows the steps involved in the synthesis and phase change of the PAM/DPDH organohydrogel [4]. The cellulose-graphene nanosheet composite phase change material can be made into a heating element, which has many advantages, such as realizing electrical control and ultra-long-distance optical control, easy secondary processing of the shape, energy saving, and long constant temperature time after cutting off the control circuit. The PAM/DPDH glycerol hydrogels' exceptional encapsulation capability and enhanced anti-drying qualities are significant. On the other hand, by adopting a similar process, the PAM/DPDH and poly(N,N-dimethylacrylamide) matrix with physical cross-links yields reversible phase-change hydrogels (PCHs); however, the PAM/DPDH hydrogel is prone to water loss, and the hydrated salts precipitate to the gel's surface quickly [35, 36]. The printing of components for various applications, including the food, aerospace, medicinal, and electrical industries, as well as soft robotics and tissue engineering, has brought attention to 3D printing technology in recent years. When exposed to various environmental stimuli such as temperature changes, pH variations, UV radiation, electric fields, etc., PCMs can change in size, shape, or other characteristics. Better possibilities are created in environments where human access is nearly impossible using 3D printing and PCMs [19]. The primary process is a shift in the hydration state that promotes intra- and intermolecular hydrogen bonding, which eventually causes a temperature change to either increase or reduce hydrogel solubility. Table 2 lists the interactions that affect the thermoresponsive hydrogels' phase change [37]. PCM enables system designs that are unrestricted in terms of degree of freedom. Improvements to smart systems will come from various novel and environmentally friendly ways to incorporate electronics. The creation of a biocompatible PCM system has the potential to transform the area of medicine and improve the efficacy of healing [38]. 3D printing, an anisotropic gel-phase ink that can thin shear. The ink comprises a gel-phase fugitive carrier and a hydrogel precursor solution. Triblock copolymers are an excellent option for 3D printing because of their shear-thinning and thermally reversible gelation properties. The copolymer comprises polyethylene oxide (PEO) and polypropylene oxide (PPO) [39, 40]. Anisotropy is essential to living organisms' shape changes and motions [41]. Here, we describe a bioinspired method for constructing temperature-responsive 3D structures with preprogrammed motions using linear hydrogel actuators comparable to biological linear contractile elements (Figure 3) [42].

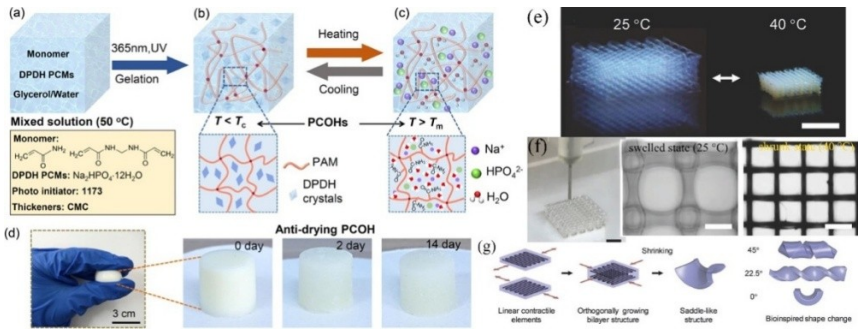

Figure 3. Preparation process and phase-change mechanism of the PAM/DPDH PCOHs. (a) A homogeneous solution of raw materials was dissolved at 50 °C and then gelatinized by photoinitiated polymerization under UV irradiation at 365 nm, (b) PAM/DPDH PCOHs in the crystallization state, (c) PAM/DPDH PCOHs in the melting state, (d) images of the PCOHs both during their preparation and after being kept in the air for two and fourteen days at 22 °C and 61 % relative humidity [4], (e) Reversible volume change of a PNIPAM structure that is temperature-responsive, (f) Top view optical microscope photos of a 3D printed structure at the swelled state (25 ºC) and the shrunk state (40 ºC), and (g) Hydrogel bilayer structures grown orthogonally and with controlled movements printed in three dimensions [42].

Here, Wei *et al.* [43] demonstrate a simple process for creating and printing PCM-filled inks via DIW, utilizing spherical PCM beads to adjust the viscosity of a photocurable resin matrix. High-temperature emulsification generated PCM beads, which were then printed, UV-cured, and dispersed in commercially available acrylate resin. As seen in Figure 4(a), PCM beads serve two purposes: they alter the ink's rheology and provide thermal energy management properties. Since the formulation of the ink is independent of the chemistry of the PCMs, multiple PCMs can be included in a single ink. This expands the operating temperature window and improves the structure's capacity for thermal management in Figure 4(b).

Figure 4. Preparation, printing, and design idea for the polymer PCM composites: (a) The design idea illustration. The liquid resin elastically contains the PCM, and its beads modify the rheology. (b) Schematic showing how the thixotropic ink made of photopolymerizable resin and PCM particles is made and printed [43].

Table 2. The response of various phase-change hydrogel materials to environmental stimuli.

Materials	Important factors	Key properties	Ref
PAAm hydrogel layer with PNIPAM/rGO hydrogel layer attached.	-Light exposure time -Light intensity	-Response to NIR light and sunlight -Bending to a ring	[44]
PEG-PNIPAM hydrogel in a bilayer structure featuring distinct PEG patterns in every layer.	-Temperature -Geometry and orientation of PEG -Hydrogel width	-Bioinspired motions (e.g., bending, twisting)	[42]
PNIPAM-PAAm-PTCA monolayer with gradient PAAm distribution in the thickness of gelatin hydrogel.	-Temperature -pH -UV light -Irradiated domain	-Bending angle of 350° -On-off fluorescence synergistic with shape change	[45]
Gradient distribution of GO through the thickness of a monolayer PNIPAM/GO hydrogel.	-GO concentration -Light exposure time -Light intensity -Electric field intensity	-Response to NIR light	[46]
PNIPAM hydrogel layer and alternating PpMS/ZrO_2 layer create Bragg stacks.	-ZrO_2 volume fraction -Temperature	-Thermo-tunable intensity of reflection peak -Thermo-tunable color from reddish to greenish blue	[47]
PNIPAM/VO_2 composite hydrogel film sandwiched between two glass slides	-Temperature -VO_2 content -Hydrogel thickness	-Long-term operation of 20 heating–cooling cycle	[48]

4. CHARACTERIZATION OF PCM-POLYMER COMPOSITE HYDROGEL

Numerous external stimuli, including temperature [49], light [50], pH, and electric signals [51], can cause the hydrogels to change phases. Hydrogels possessing photosensitive characteristics often undergo form changes when exposed to light locally [52]. The exceptional versatility of 3D printing in creating intricate shapes has made it a desirable choice [53]. In 4D printing, stresses are added to a 2D structure instead of direct printing, as in 3D printing. Stress is released upon triggering (e.g., heating), and the structure continues to expand into three dimensions over time (the fourth dimension). These methods get over the usual layer-by-layer printing restriction [54]. Different exposure times, and consequently light doses, result in pixelated polymer networks with varying cross-linking densities and degrees of monomer conversion. The impact of 2D print design on the 3D shape before printing genuine custom 3D shapes. A concentric circular print arrangement is employed for this analysis. Light-absorbing nanoparticles, such as carbon nanotubes, graphene, and GO nanosheets, which can convert visible and, more frequently, infrared light into thermal energy, are added to thermoresponsive hydrogels to provide them photo-responsive capabilities for the latter hydrogel actuator. The inhomogeneous Poly *N*-isopropyl acrylamide (PNIPAM/GO) hydrogels can be heated and experience bending deformation upon IR irradiation (Figure 5) because the GO nanosheets can absorb IR light and convert it to thermal energy. PCM adsorption carriers can be derived from the phase transition behavior of porous biomaterials [55], the cross-linker PEG-based porous polymer materials [56], etc. As a third-generation aerogel, cellulose aerogel possesses the benefits of both organic polymer and inorganic aerogels, including easy processing and good form stability, while being renewable and environmentally friendly. It possesses both the qualities of a light-responsive, high-load capacity polymer with a strong degradable heat capacity [57]. Thus, solid PCMs could be used to change the viscosity of inks. They are a desirable option for 3D printing system optimization when printing with a UV light curing system because of their capacity to increase ink viscosity without impairing the mechanical characteristics of the created structures [58].

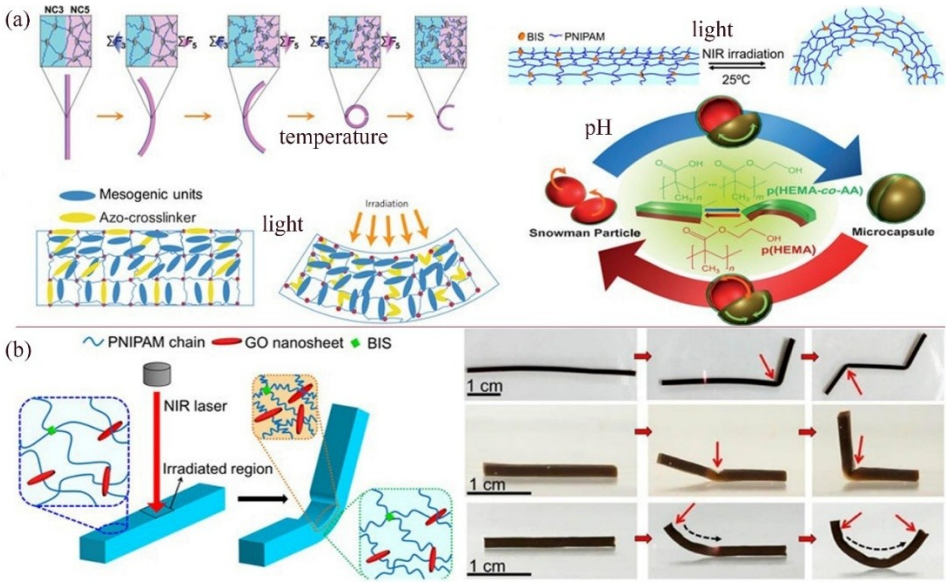

Figure 5. (a) PNIPAM/GO hydrogels' shape changes are activated by various external stimuli, such as temperature, local NIR irradiation, light, and pH; and (b) shape changes by local NIR irradiation [59].

Table 3. A summary of 3D printed hydrogels: Characterization of PMC polymer composites and their properties.

Response type(s)	Printing technique	Main material(s)	Proposed application(s)	Ref.
Light	MPI	PNIPAm, AuNRs	Microswimmer, artificial muscle	[60]
Light	DIW	PNIPAm, GO	Gripper	[61]
Light	DIW	Sodium alginate, GO		[62]
Light	DIW	Chitosan, GO	Smart curtain, remote control device	[63]
Thermal	PmSL	PNIPAm	Gripper, microfluidic	[59]
Thermal	DIW	PEG, α-CD		[64]
Thermal+ light	DIW	PNIPAm, laponite, CNT	Motion sensor	[65]
Thermal + pH	SLA	PNIPAm, PCEA	Gripper	[66]
Thermal + pH	DIW	Chitosan	Scaffold with antibacterial activity	[67]
Electric-field	PmSL	PEGDA, PAA	Artificial muscle	[68]
Magnetic-field	DIW	PAA, MNPs, PEG	Drug delivery systems, Magnetic soft robot	[69, 70]
Ion	SLA	PEGDA, PAA	Plug-and-play part, microfluidic	[71]

5. APPLICATIONS

In recent times, 3D printing design stimuli-responsive hydrogels have become popular in the manufacturing of advanced biomaterials. These hydrogels respond to environmental stimuli by exhibiting notable changes in their chemical, physical, and biological properties. Examples of 3D printed materials include tissue engineering, drug delivery, and smart biomedical devices made of PNIPAM, GelMa, DPDH, and PEG-based light and thermoresponsive hydrogels.

5.1. Tissue engineering

Hydrogel is a novel substance primarily utilized in tissue engineering, combining engineering and biology concepts to create viable replacements for diseased, missing, or injured tissues and organs. This process is known as 3R repair and regeneration. In response to transplant rejections and restrictions on donor tissue, this sector has been growing quickly. Because of its precision and customization capabilities, 3D printing is becoming an increasingly useful tool for scientists, engineers, and medical professionals. It has the potential to significantly accelerate the conversion of tissue engineering concepts into quick, affordable, and promising clinical applications [72].

5.1.1. Brain tissue

The ability to study the healthy neural dynamics and the alterations that lead to disease both in vitro and in vivo is made possible by advancements in neuronal cell technology, such as 3D brain organoids, neural progenitor cell bioprinting, and the homogeneity and specificity of neural cell types derived from induced pluripotent stem cells [73]. The brain's structural complexity limits the amount of functional 3D brain-like cortical tissue biomimicry, including

cerebral organoids and physiological processes [74]. 3D printed (by hand extrusion) cerebral cortex-like structures, which have been modified by RGD (Arginine-Glycine-Aspartic acid) and contain many layers of cortical neurons wrapped in gellan gum hydrogel and GelMa [75]. Signaling factor gradients, the ability to vascularize in later rounds, and the generation of brain tissue cell subtypes may be required for more sophisticated applications of these more complicated brain models.

5.1.2. Vascular tissue

Since isolated cells cannot survive in volumes less than 3 mm^3, the fabrication of circulatory systems is one of the primary problems in 3D printing. Vascular channels transfer nutrition, growth hormones, oxygen, and waste products away from live cells [76]. Shengjie *et al.* [77] printed vascular networks by combining adipose-derived stromal cells (ADSC) with gelatin, alginate, and chitosan (GAC) hydrogel composites. Numerous coaxial nozzles produced highly organized circulatory systems, as illustrated in Figure 6. The constructs were permanently fixed in shape by ionic crosslinking sodium alginate, and their morphologies were stabilized by combining gelatin methacryloyl (GelMA) with 4-arm poly(ethylene glycol)-tetra-acrylate (PEGTA). Compared to sECM hydrogels crosslinked with poly(ethylene glycol) diacrylate (PEGDA), the crosslinked hydrogel composites had better rheological characteristics, making them more appropriate for bioprinting. HepG2 C3A, Int407, and NIH3T3 cells were used in bioprinted hydrogel composites that showed (Figure 6) microcapillary tube structure and cell survival for up to 4 weeks [78].

5.1.3. Spinal cord regeneration

The tissues of the spinal cord are formed of several cell types that are very spatially distributed rather than structurally homogeneous. A traumatic spinal cord injury (SCI) has a dismal prognosis and might result in an instant loss of motor or sensory function below the site of injury [8]. It is assumed that this 3D bioprinting approach will make it possible to construct novel GelMa hydrogel-based bioscaffolds that mimic the complex architecture of CNS tissues, to cure specific neurological disorders, such as repairing or rebuilding the SCI [79]. It has been recently reported that complex CNS structures can be fabricated utilizing microscale continuous projection printing (μCPP) for tissue regeneration applications in spinal cord injury repair. Biomimetic 3D hydrogel scaffolds can be printed using the μCPP technique; the scaffold's characteristics can be adjusted to match the size of the human spinal cord and the real lesion geometries [80].

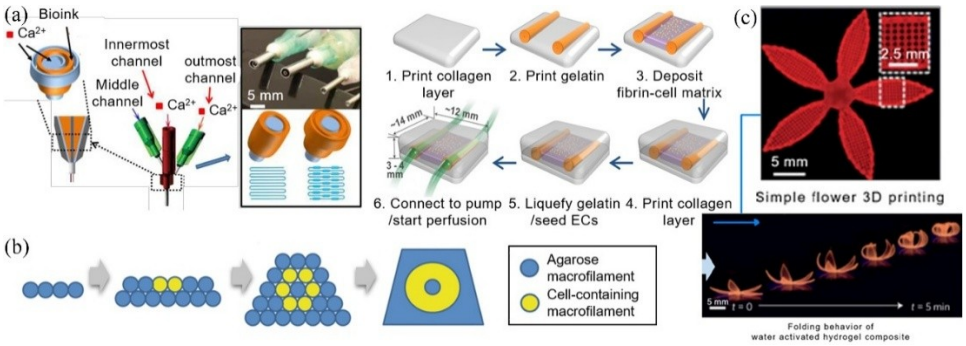

Figure 6. (a) Multiple coaxial nozzle techniques with alginate, GelMA, and 4-arm PEGTA construct the vascular system, and by bioprinting collagen, fibrin-cell combination, and sacrificial gelatin layer by layer; (b) Stacking hydrogel macro filaments to create a cellularized tubular structure; and (c) Effect of water-activated 4D printing [78].

5.2. Biomedical devices

Hydrogel-based soft electronics improve tailored gadgets and human-machine interactions. Conductive hydrogels are ideal options for strain sensors [81], artificial muscles [82], soft robotics [83], and artificial muscles because of their customizable elasticity and electrochemical characteristics. The excellent biocompatibility and stimuli-responsiveness of conductive hydrogel-based flexible electronics are pushing the boundaries of bioelectronics and could even enable implanted devices to repair injured peripheral nerves [84]. In particular, 3D printing offers more possibilities for conductive hydrogel patterning in a programmable way, improving the device's overall functionality. This section discusses the 3D printing of hydrogels for flexible electronics, such as electrochemical biosensors, soft robotics, shape-morphing actuators, flexible sensors, and electroluminescent devices in Figure 7 [85].

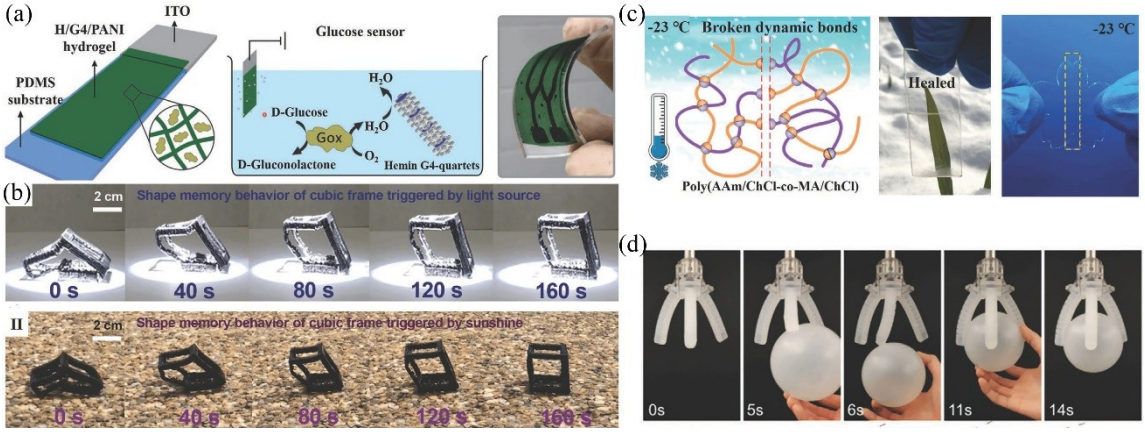

Figure 7. (a) Hydrogel-based glucose sensor and its three-dimensional sensing technique; (b) Shape-morphing actuators manufactured in 4D recovering the shape of the printed actuator with light intensity; (c) Mechanisms for self-healing and the conductor's characteristics at freezing temperatures; (d) Somatosensory feedback of the soft robotic grasper when the ball is being held and released [85].

5.3 Drug delivery

A thermo-responsive hydrogel was employed for its unique ability to gel exclusively at 37 °C across different polymer concentrations [13, 86]. An oral drug delivery system, various multilayered drug delivery devices, synthetic extracellular matrices, antibiotic-printed micropatterns, microcapsules, nanosuspensions, and mesoporous bioactive glass scaffolds were printed using 3D printing technology [87]. This allowed precise control over parameters like size, shape, release rate, and tailored formulation. Generally speaking, thermogels depend on the presence of a phase transition between room temperatures and physiological temperatures, such as a volume phase transition temperature, which occurs after injection, or a lower critical solution temperature, which allows for the in situ gelation of the material and is therefore used as a depot formulation in the case of PNIPAM or poly(oligoethylene glycol methacrylate) [88].

6. CONCLUSIONS AND FUTURE RESEARCH DIRECTION

PCMs and novel 3D printing techniques can be used to study phase transitions in hydrogels. These hydrogels' enhanced thermal storage results from the reversible phase

transition between liquid and solid phases. With their precision and usefulness, PCMs have new opportunities because of 3D printing technology. It is now possible to create complex and practical hydrogel structures. Various phase change materials such as metal hydrogels, bio-based hydrogels, polymer hydrogels, chitosan hydrogels, and graphene hydrogels have been successfully used recently. Functional hydrogels with various properties, such as dual network, magnetic, temperature sensitive, and biogels can now be fabricated using direct ink writing technology and rheology modifiers such as carbomer. For example, mixing cellulose nanofibrils with high-load PCM inks produces a strong composite monolith with excellent thermal stability. Hydrogels are suitable for tissue engineering and regenerative medicine applications utilizing 3D printing due to their inherent qualities, including the ability to mimic the extracellular matrix; even with great progress, several challenges remain, such as expanding the range of printable hydrogel systems and improving flexibility and density for energy conservation. Phase change hydrogel 3D printing holds great promise if these issues are addressed and the technology's application scope is expanded. Technological advances like smart materials and 4D printing with stimuli-sensing capabilities bring new possibilities for more dynamic and adaptive designs. New technologies such as thermoresponsive hydrogels and UV-crosslinkable hydrogels are promising for bioprinting applications as they can create complex 3D structures that closely mimic the natural environment. Spinal cord regeneration, vascular tissue development, and brain tissue generation are potential medical uses of 3D-printed hydrogels. Advanced bioprinting techniques, such as continuous micro projection printing, can precisely manipulate scaffold properties and accurately reproduce complex central nervous system architectures for effective tissue regeneration. Flexible electronics, biosensors, and soft robotics can see revolutionary advances in the versatility of 3D printing in the fabrication of hydrogel-based soft electronics. Since their electrochemical properties can be tuned, conductive hydrogels are an excellent choice for applications that require high biocompatibility and stimulus responsiveness. In summary, phase change hydrogels and 3D printing technology have great application potential in various fields. By overcoming current challenges and encouraging creativity, this holistic approach is expected to transform materials science, biomedical engineering, and other related scientific fields, paving the way for more efficient, adaptable, and valuable applications in the future.

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Declaration of competing interest. The authors declare that they have no known financial or interpersonal conflicts that would have appeared to impact the research presented in this study.

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