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Recent Advances in the Additive Manufacturing of Stimuli-Responsive Soft Polymers

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Stimuli-responsive polymers (SRPs) are special types of soft materials, which have been extensively used for developing flexible actuators, soft robots, wearable devices, sensors, self-expanding structures, and biomedical devices, thanks to their ability to change their shapes and functional properties in response to external stimuli including light, humidity, heat, pH, electric field, solvent, and magnetic field or combinations of two or more of these stimuli. In recent years, additive manufacturing (AM) aka 3D printing technology of these SRPs, also known as 4D printing, has gained phenomenal attention in different engineering fields, thanks to its unique ability to develop complex, personalized, and innovative structures, which undergo twisting, elongating, swelling, rolling, shrinking, bending, spiraling, and other complex morphological transformations. Herein, an effort has been made to provide insightful information about the AM techniques, type of SRPs, and their applications including, but not limited to tissue engineering, soft robots, bionics, actuators, sensors, construction, and smart textiles. This article also incorporates the current challenges and prospects, hoping to provide an insightful basis for the utilization of this technology in different engineering fields. It is expected that the amalgamation of 3D printing with SRPs would provide unparalleled advantages in different engineering arenas.

1. Introduction

Additive manufacturing (AM), alias 3D printing, refers to the process of creating solid 3D objects from a computer-aided design (CAD) digital model successively depositing layers, instead of removing the material as in the conventional manufacturing processes like machining.[1–4] A 3D printing process starts with designing a 3D model using CAD software, which is then converted into a standard triangle language (STL) file by dividing the model into small polygons.[5–7] These polygons, upon adding up, create the complete exact model. The STL file is then sliced into thin layers required for printing and each layer is printed one at a time to create the final object.[8–10] Every successive layer is deposited upon the previous layer according to the information of the CAD sliced model. The feedstock material could be solid, liquid, or powder depending upon the type of the AM process applied.[11–15] The AM technology has remarkably reduced the human interface, processing time, and cost associated with the product development process.[16] It offers the flexibility to create 3D objects from a range of inorganic and organic materials including plastics, metals, ceramics, and composites.[17–21] Inorganic materials have limitations such as brittle nature and high thermal properties. Compared to these materials, organic materials are abundant, soft, and flexible.[22] Traditional organic materials have received significant attraction and importance in the AM technology, thanks to their excellent mechanical, thermal, and electronic properties.[23–25] Over the years, polymer-based organic materials have shown tremendous scope in various applications like soft robotics, sensors, and digital technology.[26–29] However, these

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The ORCID identification number(s) for the author(s) of this article can be found under https://doi.org/10.1002/adem.202301074.

DOI: 10.1002/adem.202301074

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polymers have shown reluctance to environmental stimuli due to the presence of irreversible covalent bonds. Thus, these polymers require the fine-tuning of their intrinsic properties through external stimulations for biocomputing, artificial machines, and therapeutic applications.[30–32]

AM technology has laid the foundation for making complex structures with selective properties and has revolutionized the design, development, and manufacturing of objects, offering new possibilities for the textile, biomedical, aerospace, automotive, and construction industries.[33–39] The 3D printing has further categorized into material extrusion, vat photopolymerization, inkjet printing, powder bed fusion (PBF), directed energy deposition, binder jetting (BJ), and sheet lamination.[40–43] The AM process selection depends on feedstock material, complexity, product size, and the desired final properties.[44–47] However, the AM technology is not limited to create objects with permanent characteristics, rather it can fabricate the structures that respond to external stimuli such as heat, light, moisture, pH, mechanical force, etc.[48–50] This innovative process of creating smart and dynamic structures using the 3D printing technique is known as the 4D printing, which adds more flexibility to product development.[51] In 2013, a research group headed by Tibbits from the Massachusetts Institute of Technology (MIT) offered the novel concept of the 4D printing.[52–55] Additionally, the 4D printing applies the same procedures as the AM utilized but includes an extra feature of change or transformation in material characteristics over time.[56–58] The fourth dimension indicates the ability of stimuli-responsive materials to adjust and transform over time in response to an external stimulus or environmental condition.[59–61] Stimuli-responsive materials are engineered to respond in a specific and controlled manner under different inputs.[62] These materials can be classified into shape-memory polymers (SMPs), shape-changing polymers (SCPs), shape-memory alloys (SMAs), shape-memory hydrogels (SMHs), and liquid crystal elastomers (LCEs).[63–68] The incorporation of smart materials into 3D-printed structures gives rise to products with embedded memory that help the material to adjust and change its shape once activated.[69–71] 4D printing technology has enormous potential in the technological fields of construction, biomedical engineering, soft robotics, functional textiles, and electronic devices, due to the bending, spiraling, folding, and twisting mechanisms of stimuli-responsive materials,[72–77] as elaborated in **Figure 1**.

1.1. Scope of the Review

Stimuli-responsive polymers (SRPs) are extensively applied to develop dynamic structures using the 4D printing technology. The combination of 3D printing and stimuli-responsive behavior provided by smart polymers permits the development of complex and multifunctional materials for limitless applications.[78–80] Few reviews on the 4D printing of soft polymers can be found in the literature. For instance, Lang et al. have recently published a review article,[81] which contains advancement in 3D-printed SMP research only, with a prime focus on remote-actuation approaches and multimaterial responsive structures. Similarly, Shafranek et al.[82] provided insightful information about the

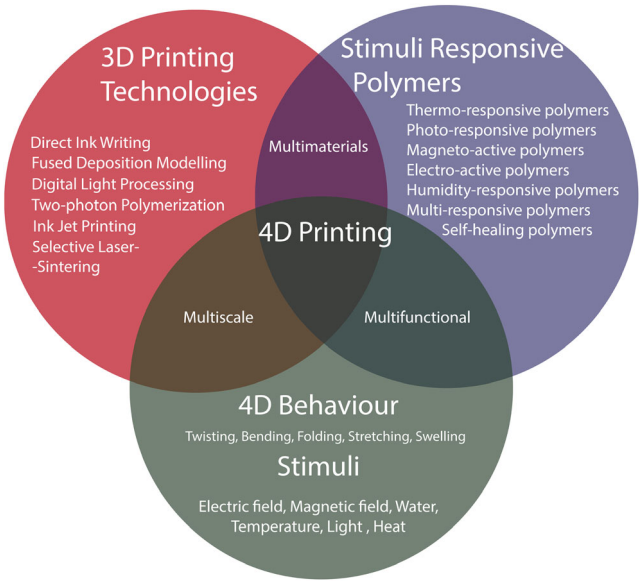

Figure 1. A brief overview of AM of SRPs.

3D printing of stimuli-responsive materials including SMHs, SMPs, and SMAs. However, the literature lacks a critical review only focusing on the 3D printing of SRPs. Herein, this review closes the gap by providing an overview of recent advancements in the AM of SRPs to fabricate shape-morphing and bioinspired structures for different engineering applications. Additionally, the current review also elucidates types of SRPs, 3D printing techniques, and their applications. Finally, it includes current challenges and outlook for future research directions.

1.2. Stimuli-Responsive Materials

Stimuli-responsive materials are programmable materials, which exhibit the potential to change their geometries from temporary states to original states in response to an external stimulation such as moisture, light, heat, pH, electric field, and magnetic field.[83–86] These materials exhibit a variety of features such as self-folding, self-assembly, and self-adaptability.[87] In addition, self-healing properties can be easily induced in soft responsible materials. These materials not only produce controlled and predefined changes in their shape but also transform the colors or functional properties.[88] Additionally, stimuli-responsive materials which change the material's properties such as thermal conductivity[89] and stiffness[90] as well as color[91–95] have also been garnering tremendous interest in arenas of tissue engineering, energy absorption materials, optical systems, and biosensors. Out of all stimuli-responsive materials, SRPs have gained more attention, due to their advantages of easy processability, low cost, large stretchability, adjustable properties, reversible deformability, and remarkable shape-memory effects.[96–99] Furthermore, these polymers offer excellent properties including biodegradability, biocompatibility, and adaptability, which are essential requirements for applications in complex areas such as the development of tissue scaffolds.[100–102] These SRPs induced changes in polymer

chain conformation by transforming polymer–solvent or polymer–polymer interactions.[103,104] However, the fabrication of novel SRP composites through 3D printing and inducing their properties via stimulations are still in their infancy that require adequate attention and further investigations.[105]

SRPs are classified into two main types: SMPs and SCPs.[106–108] SMPs, as a series of dual-shape materials with intrinsic properties, can realize the conversion between temporary and permanent geometries under the influence of particular stimulation,[109–111] as illustrated in **Figure 2**. These polymers can restore their shapes after being programmed at a high temperature and cooling at a low temperature.[112] By freezing the oriented crystallites of different domains, SMPs store their temporary shapes. However, these polymers retain their original shapes by erasing temporary shapes upon heating at an elevated temperature.[113–115] Conventionally, SMPs cannot shift back to temporary shape after cooling, unless additional programming is done to develop another oriented domain. These polymers with irreversible actuation are known as one-way (irreversible) SMPs.[116] On the other hand, two-way (reversible) SMPs do not require programming to recover their temporary geometry.[117–120]

In contrast to SMPs, SCPs gradually change their shapes after external stimulations and return to their basic shapes as soon as the stimulations are removed. Such actuating behavior is reversible and can be repeated multiple times.[121–125] SMPs and SCPs are different from each other based on the geometric movement of structures.[126–128] Both types of polymers offer attractive advantages including distinct mechanical properties, potential recyclability, low density, excellent shape recovery, and deformation capacity under a variety of stimuli, easy processing, recoverable strain, chemical stability and modification, biocompatibility, and biodegradability with adjustable degradation rate.[129–133] Compared to SMAs, these polymers are lightweight, cost effective, and easier to process. Thanks to these appealing properties, both SCPs and SMPs are considered promising

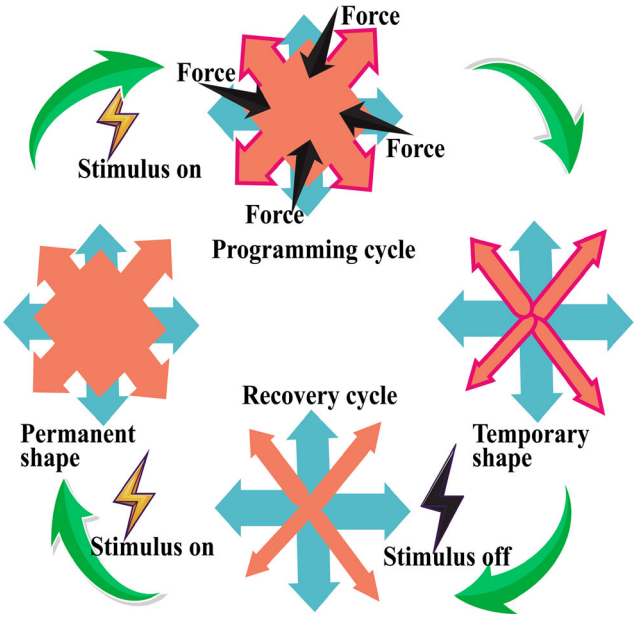

Figure 2. Schematic illustration of shape-memory cycle for SRPs.

SRPs for the biomedical sector like drug delivery, cancer treatment, tissue engineering, cardiovascular, and other surgical applications.[134–140]

Hydrogels are macromolecular structures, in which polymer molecules are bonded in a hydrophilic network.[141–144] These materials can absorb a large amount of water, due to void imperfections in their structures (such as hydroxyl and carboxyl groups).[145–148] Conventional hydrogels offer limited swelling response associated with water dependency. Hence, recently, stimuli-responsive functional hydrogels have been fabricated, which have shown dynamic behavior upon environmental changes such as light, heat, electric field, pH, magnetic field, etc.[149–158] Alongside SRPs, stimuli-responsive hydrogels are also extensively applied for different engineering applications.[159–166] These materials exhibit a great ability to self-folding upon external stimulations, which make them highly suitable for fabricating dynamic scaffolds.[167–171] Additionally, smart hydrogels are also attractive for developing drug delivery structures, thanks to their aqueous nature and excellent sensitivity to biological stimuli.[172–176]

In summary, stimuli-responsive hydrogels and SRPs have enabled the AM of functional structures for a variety of applications. **Table 1** contains a wide range of stimuli-responsive naturally derived and synthetic polymers as well as hydrogels and their advantages and disadvantages.

2. AM Techniques

AM is a unique process of fabricating solid full-scale 3D objects by depositing the material layer by layer. This technology has widely emerged as an alternative manufacturing approach compared to conventional manufacturing techniques thanks to its ability to create complicated geometries for multimaterials with intricate features in decreased processing time for manufacturing.[177–183] 3D printing of SRPs utilizes the same commercial AM technologies to develop smart structures for different engineering applications.[184] However, 3D printers should be compatible with these smart materials or exhibit multimaterial characteristics for the cases where deformation mismatch within the structure causes functional or shape change.[185–188] Different processes, each with their specific advantages and limitations (**Table 2**), have been developed depending on the types of feedstock materials, the relevant processes, and their working principles are discussed in the following section.

2.1. Extrusion-Based Printing

Extrusion-based AM is a flexible manufacturing technology used to fabricate smart and customized structures through the deposition of extrudate in a layer-by-layer manner.[189–197] This technology is highly suitable for developing a wide range of objects spanning from small prototypes to large-scale structures.[7,198–200] In extrusion-based printing, the successive printed layers bonded together by one of the two approaches: fused deposition modeling (FDM) and direct ink writing (DIW).[201–204] FDM utilizes temperature to liquefy the solid feedstock enabling the flow through the extrusion nozzle and

Table 1. Prominent stimuli-responsive materials for 3D printing.

Source	Materials	Prominent features	Common stimulus	Advantages	Disadvantages	References
Naturally derived	Chitosan	SMP composites	pH, ionic	i. Good biocompatibility ii. Osteoconductive iii. Excellent antimicrobial properties	i. Slow gelatin ii. Bad water solubility	[588–591]
	Hyaluronic acid	SMP composites	–	i. Excellent biocompatibility ii. Chemical modification is easy	i. Requires chemical modification for stable cross-linking	[592–595]
	Alginate	SMP composites	pH, ionic	i. Fast gelatin ii. Low cost	i. Slow degradation ii. Low cellular attachment	[596–599]
	Collagen	SMP composites	Heat, pH	i. High bioactivity ii. ECM structural protein	i. Weak mechanical strength ii. Slow gelatin	[600–603]
	Silk fibroin	SMP composites	Enzymes	i. High elasticity ii. Good mechanical strength iii. Excellent chemical stability	i. Poor cell attachment ii. Rapid degradation iii. Aggregate under shear stress	[604–609]
	Agarose	SMP composites	Heat	i. Mechanically robust ii. Gelatin temperature close to physiological temperature	i. Excessive water uptake ii. Low cellular attachment	[610–613]
	Gellan gum	SMP composites	Heat, ionic	i. Easy blending and excellent rheological modifier ii. Mechanically robust	i. Poor cell attachment	[614–617]
	Gelatin	SMP composites	Heat	i. Derivative of collagen ii. High bioactivity iii. Better solubility	i. Rapid degradation ii. Weak mechanical strength	[618–621]
Synthetic	PLA	SCP and SMP	Heat, light	i. Excellent biocompatibility ii. High flexibility	i. Low cellular adhesion ii. High inflammability	[622–624]
	PEG	SMP composites	–	i. High water solubility ii. Permits versatile chemical modifications	i. Poor cell attachment ii. Poor biodegradability	[625–627]
	PCL	SCP and SMP	Heat	i. High biocompatibility ii. Excellent mechanical properties	i. Low cell adhesion	[628–631]
	PNIPAm	SCP and SMP	Heat	i. Mechanically robust ii. Gelatin temperature close to physiological temperature	i. Poor cell attachment ii. Poor biodegradability	[632–635]
	PU	SMP composites	Heat	i. Tailorable sol–gel transition temperature ii. Excellent biocompatibility	i. Poor biodegradability ii. Weak cell attachment	[636–639]

deposition over the previously printed layer,[205–209] as illustrated in **Figure 3**A, which forms bonds during solidification. The combination of the FDM and thermo-responsive polymers (TRPs) is most commonly used to print dynamic structures. FDM is flexible and straightforward, but highly unsuitable for 4D printing, due to its incompatibility with SRPs.[210–216] Additionally, the lack of intelligent filaments has made the FDM process largely complex for the 4D printing.[217–220]

In contrast to FDM printing, DIW is a common printing approach in which the bonding and solidification of various layers are occurred by chemical changes through the cross-linking (curing) reactions and interlayer cross-linking.[221–226] In the DIW printing, viscoelastic polymer inks are extruded through a nozzle and are deposited as thin layers one upon another to create a solid object,[227–230] as illustrated in Figure 3B. This technique is suitable for gel-like materials which offer special rheological properties such as shear thinning.[231–235]

2.2. Photopolymerization

Photopolymerization is another widely used and emerging AM method that uses light to solidify a liquid polymer (resin) to create 3D objects. The basic principle involves the exposure of a photocurable resin to UV radiation in a predefined method.[236–240] Photopolymerization-based AM technology is broadly categorized into two main types: vat photopolymerization and material jetting (MJ).[241–243] Vat photopolymerization involves the selective curing of a liquid photopolymer resin with a light source to create a solid part.[244] It is further classified into stereolithography (SLA) and digital light processing (DLP).[245–249]

In SLA, a laser is used to selectively cure photopolymerizable resins like methacrylamides or acrylates to form a solid object,[250] as illustrated in Figure 3C. This printing approach offers quick printability, high fabrication speed, and high spatial resolution.[251–253] DLP involves the use of a projector with an optical mirror to selectively cure a liquid resin,[254–256] as shown in Figure 3D. Additionally, this process can be used for printing multimaterials.[257–259] MJ is another type of photopolymerization that involves the deposition of liquid material through an inkjet head onto a build platform,[260] as illustrated in Figure 3E. The deposited liquid material is then cured and solidified using a light source to develop a 3D object.[261] MJ process is suitable for printing intricate multimaterial parts with high precision and surface quality.[262]

Table 2. Different AM techniques, printed materials, their advantages, and disadvantages.

AM technology	Main processes	Resolution [μm]	Curing method	Key features and advantages	Disadvantages	Materials	References
Extrusion	DIW	100–600	Gel formation, evaporation, and temperature-assisted	i. Versatile process ii. Multimaterial printing iii. Thixotropic ink iv. Excellent shape morphing response	i. Lower resolution ii. Difficult to print intricate geometries	Viscoelastic materials, plastics, ceramic paste, and food	[640–643]
	FDM	100–150	Unaided cooling, extrusion	i. Affordable cost ii. High mechanical strength iii. Simple printing process iv. Good shape recovery behavior	i. Poor printing resolution ii. High postprocessing time iii. Relatively low dimensional precision	Thermoplastics, thermoset polymers	[644–647]
	Bioprinting	–	Gel formation	i. Cell-laden intricate structures ii. Assist in the development of complex structures	i. Low printing resolution ii. Complex operation iii. Relatively expensive	Hydrogels, living cells, and biomolecules	[648–654]
Photopolymerization	DLP	10–50	Photopolymerization, cross-linking	i. High printing speed ii. Higher resolution iii. Variety of printable materials iv. Projector light curing	i. Low mechanical strength ii. Slight distortion in printed objects iii. Postprocessing is mandatory iv. Difficult to induce sequential shape shifting	Photopolymers, metamaterials, and elastomers	[582, 655–657]
	SLA	50–100	Photopolymerization, cross-linking	i. Fast fabrication ii. High resolution iii. Variety of printable materials iv. Recycling of unused polymers v. Low material consumption	i. Low tensile properties ii. Not suitable for multimaterial printing iii. Cytotoxicity iv. Designing of 4D printable ink is difficult due to cross-link density	Photocurable resins	[658–661]
	TPP	0.1–5	Photopolymerization, cross-linking	i. Extremely high resolution ii. Excellent shape memory behavior of microstructures iii. Develop complex structures	i. Low production ii. Relatively higher cost	Photocurable resins	[662–667]
	IJP	50–300	UV photopolymerization	i. High gelatin speed ii. Good resolution iii. Noncontact nozzle iv. High accuracy	i. Low dimensional accuracy ii. Limited printable materials iii. Relatively low mechanical strength	Photopolymers, bioinks	[668–671]
Powder-based printing	SLS	50–100	Laser-assisted coalescence	i. No need of supporting structure ii. Excellent mechanical properties iii. Better printing time	i. Limited printable materials ii. Low resolution iii. Slow shape-memory behavior iv. Low reusability of un-sintered powders v. High printing cost	Plastic powder and metal powder	[672–674]

2.3. Powder-Based Printing

Powder-based AM processes use a powder bed as feedstock material and selectively melt and fuse the powder particles in a layer-by-layer fashion, to create the desired objects,[263] as illustrated in Figure 3F. These processes are extensively used to create parts with complex geometries, high strength, and low weight.[264] Metals and polymers are the most common 3D-printed materials used for these techniques.[265–267] Powder-based printing is broadly classified into PBF and BJ.[268] PBF involves fusing or melting the powder particles together using a laser beam and is further classified into selective laser sintering (SLS) and selective laser melting (SLM).[269,270] In SLS, the high laser beam is bombarded onto the powder bed through a predetermined pathway and is melted partially.[271] However, in SLM powder particles are melted completely by the striking beam. SLS is suitable for the 3D printing of polymeric composites, while SLM is best suited to fabricate complex metal parts.[272] BJ refers to the deposition of a liquid binder or a chemical agent onto a powder bed, to bind the powder particles and creates the desired component. This technique allows the fabrication of porous scaffolds and artificial bones.[273]

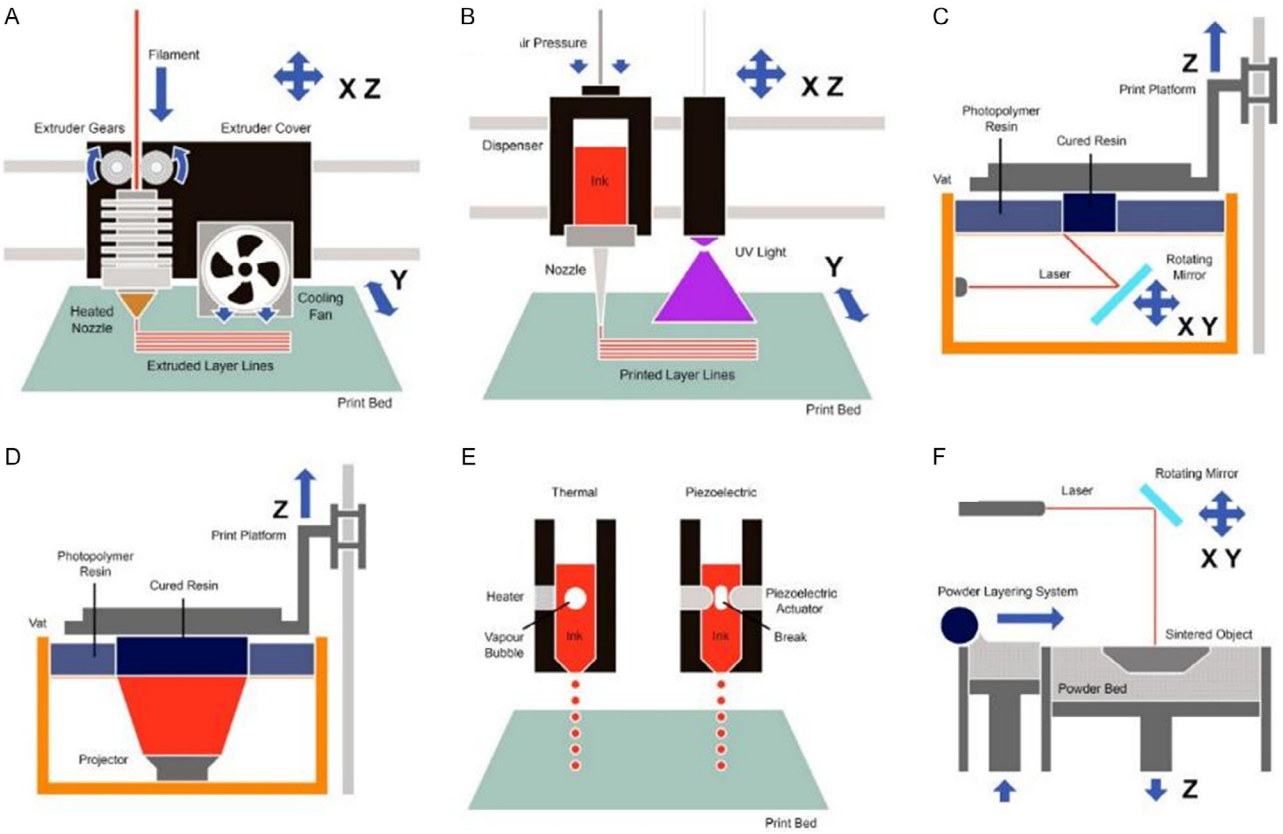

Figure 3. Schematic illustration of different AM techniques: A) FDM; B) DIW; C) SLA; D) DLP; E) inkjet printing; and F) SLS.[3]

3. Additively Manufactured SRPs

SRPs are intelligent materials that require external stimuli to induce changes in properties, shape, or color.[274–276] These soft polymers undergo twisting, bending, spiraling, and folding mechanisms upon exposure to temperature, light, humidity, pH, electric field, or magnetic field.[277–280] **Figure 4** depicts different stimuli, which change various macroscopic properties of SRP composites. The choice of a particular stimulus is dependent on the operating environment. In recent years, stimuli responses of different polymers have been widely investigated (**Figure 5**),[281] whereas **Figure 6** shows the classification of SRPs, based on the type of external stimulation. This section incorporates a critical discussion on different SRPs including temperature-, light-, humidity-, electro-, magneto-, pH-, and multi-responsive polymers.

3.1. TRPs

TRPs are an important subclass of soft smart materials, which depend on temperature change to induce gelification.[282–284] They respond to temperature fluctuations making them suitable materials in the biomedical sector for tissue engineering, drug delivery, and gene delivery applications.[285–288] For instance,

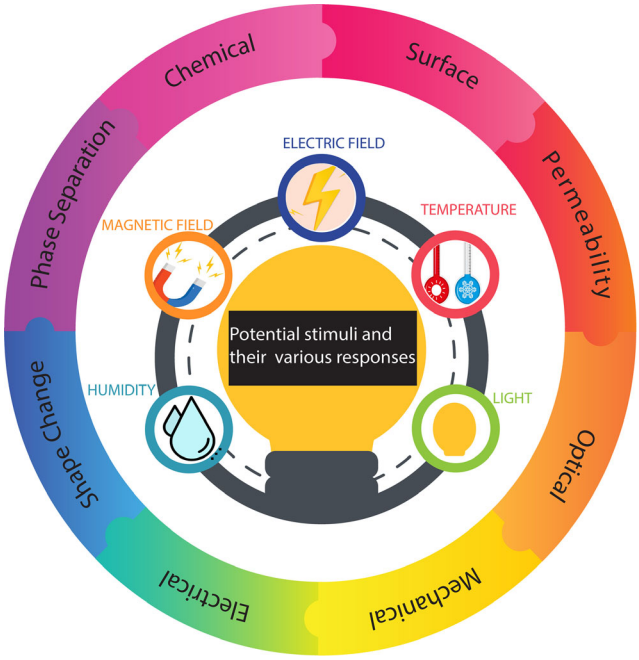

Figure 4. Potential stimuli and response of polymers.

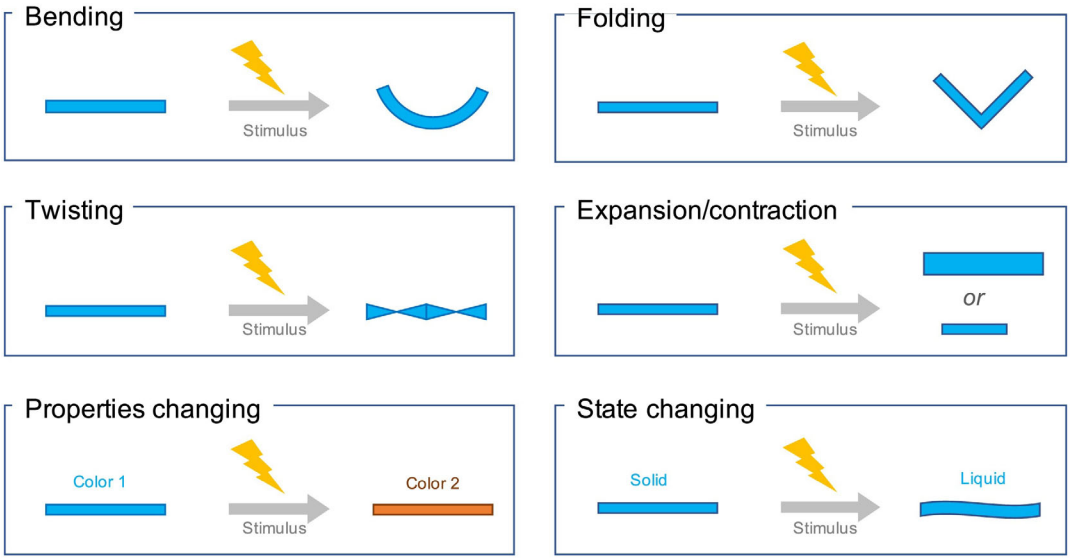

Figure 5. A variety of responses obtained when SRPs are exposed to external stimuli. Adapted with permission.[581] Copyright 2020, Elsevier B.V.

drug-laden TRPs exhibit great potentials for achieving the therapeutic effect in the living body, thanks to their excellent biochemical properties.[289–291] These polymers have become a significant research area of interest, due to their low cost, biocompatibility, thermal stability, and high flexibility.[292–294] TRPs are further divided based on an upper critical solution temperature (UCST) and a lower critical solution temperature (LCST). UCST and LCST are critical temperatures above and below polymers that get miscible, respectively.[295–299]

TRPs have shown broad application prospects in the AM technology, due to their shape-changing abilities at a specific temperature.[300–302] For instance, Wang et al.[301] used PCL/thermoplastic polyurethane (TPU)-based SMP composites to develop a bionic gripper, which responded to heat stimulus, as illustrated in **Figure 7**A The study revealed that PCL/TPU-based thermo-responsive composite exhibited excellent shape-memory recovery ratio (81.1 %), shape-memory fixity (100%), and stable shape-memory performance.

In another study, Nizioł et al.[302] developed a novel printable bioink focused to fabricate thermo-responsive structures, as illustrated in Figure 7B. In this study, printable inks were loaded with antimicrobial material for rendering therapeutic properties which can be used for wound healing applications. The study demonstrated that these 3D-printed structures exhibited excellent temperature-induced shape-morphing characteristics.

Transparent morphing polymers are an emerging class of soft TRPs, which exhibit great potential for developing imperceptible soft robotics, actuators, and wearable devices, thanks to their soft and stretchy nature and optically transparent properties.[303–305] For instance, Lee et al.[304] developed a highly transparent directional thin film actuator by incorporating a flexible and transparent silver nanowire percolation network heater. The inherent directional thermal expansion behavior of LPDE provided controlled bending without requiring structural modification of the actuator or additional complicated heater design. Additionally, these polymers can be used as a supportive component in camouflage skin, which aims to develop adaptable transparent systems.[306]

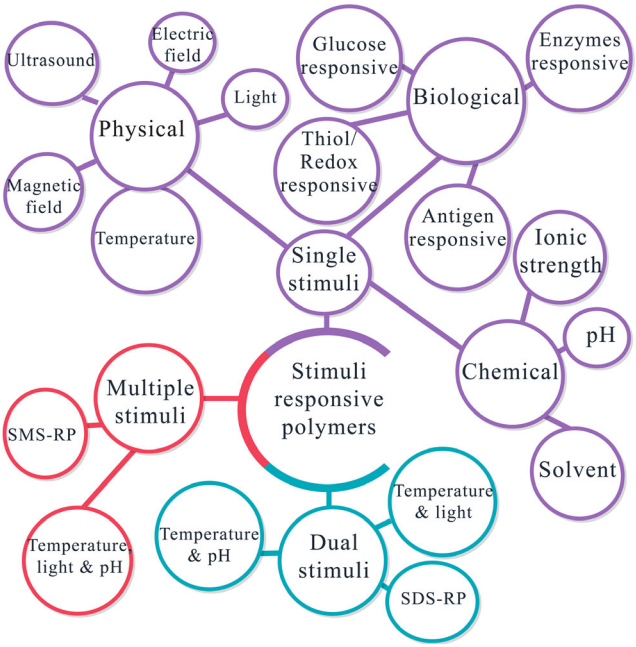

Figure 6. Classification of stimuli-responsive macromolecules.

3.2. Photo-Responsive Polymers

Photo-responsive polymers are a class of well-known stimuli-responsive materials that can change functional properties when exposed to light stimulus. These polymers offer good printability, suitability, and processability.[307–309] Near-infrared (NIR), a green, directionally controllable, remote, and clean energy resource, has been extensively applied to develop light-driven

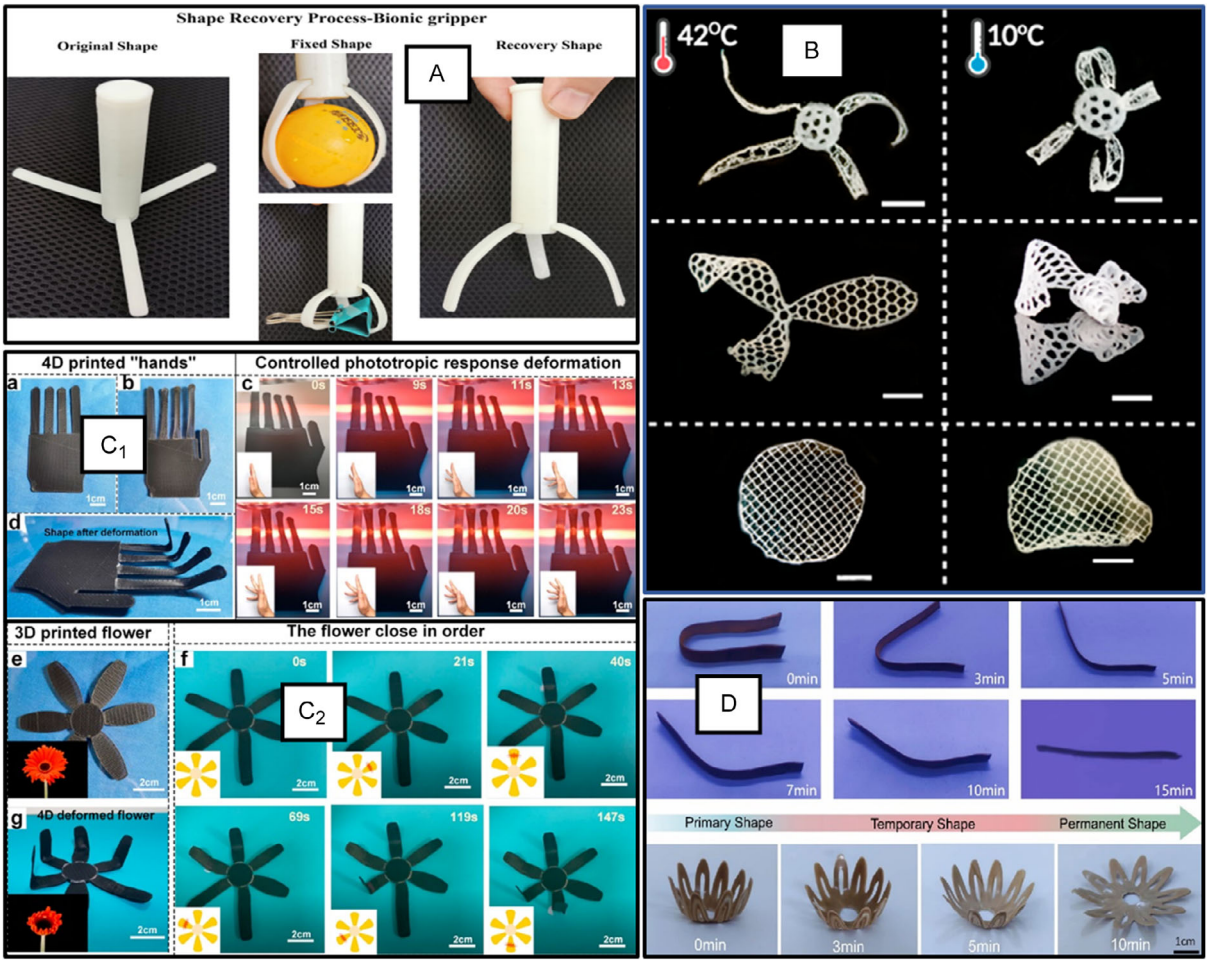

Figure 7. A) Shape recovery process of PCL/TPU-based bionic gripper developed through FDM. Reproduced with permission.[301] Copyright 2022, The Polymer Society, Taipei. B) 3D-printed thermo-responsive flower-like petals, propeller, and disc. Reproduced under the terms of the Creative Commons Attribution license.[302] Copyright 2021, The Authors, Published by MDPI. C) Sequential folding of C_1) 3D-printed hand-like structure and C_2) 3D-printed flower. Reproduced with permission.[326] Copyright 2023, Elsevier Ltd. D) Photo-responsive PU/LNT-based structures exhibiting excellent shape-memory performance. Reproduced under the terms of the CC-BY-NC-ND license.[327] Copyright 2023, The Authors, Published by Elsevier Ltd.

structures, due to its ability to minimize damage to the material and operator.[310] Light-actuated structures are usually fabricated by incorporating photo-responsive moieties (e.g., azobenzene derivatives) in the polymer.[311–313]

Light triggering exhibits adjustable irradiation parameters (wavelength and light intensity) and excellent programmable properties.[314] The choice of wavelength to induce photoreactions is extremely important for actuations and other biomedical applications.[315–317] UV radiations (low wavelength), which are applied to trigger either reversible or irreversible photoinduced reactions, are not suitable for the biomedical sector, due to their limited penetration depth and they might harm cells or tissues in in vivo applications. On the other hand, the wavelength range of NIR is adequate for biomedical applications thanks to their low energy absorption and scattering by biological media, thus ensuring deeper penetration.[318–320] However, the photoreaction induced by NIR light is usually slower and inefficient, due to low photon absorption cross sections of chromospheres. Additionally, decreasing the thickness of polymer films or increasing the light intensity increases the photochemical reactions.[321]

Light-based untethered stimulus is considered an attractive trigger to induce the shape-morphing behavior of 3D-printed devices.[322] Light-triggered actuators fabricated through 3D printing are highly effective, due to their rapid and reversible response, remote control, and precise movement.[323–325] Besides soft robotics, 3D printing has been extensively applied to develop tissue scaffolds, drug delivery, biomimetic and microfluidic devices.[326–328] For instance, Ren et al.[326] observed remotely controlled deformation in PLA/carbon black (CB)-based photo-responsive polymer composites. The authors endowed photothermal conversion features by introducing CB filler in the PLA matrix. The study demonstrated excellent phototropic bending responses of 3D-printed structures including hand-like and flower-shaped structures, as illustrated in Figure 7C_1,C_2. In another study, Wang et al.[327] developed light-responsive polyurethane (PU)/lignin nanotubes (LNTs)-based shape-memory

composites using DIW printing, as illustrated in Figure 7D. The study demonstrated that 3D-printed composites exhibited excellent shape-memory performance and mechanical properties, which can permit their utilization in the soft robotics field.

3.3. Magneto-Active Polymers

Magneto-active polymers (MAPs) are smart materials that are capable of changing shape as well as mechanical properties under magnetic field as a stimulus. These polymers contain two major constituents: a nonmagnetic matrix and magnetic particles (MPs).[329–331] Polymer matrices are doped with magnetic nanoparticles (MNPs) to achieve magneto-responsive behavior. In case of a nonmagnetic matrix, hard or soft magnetic fillers are accountable for the responsive behavior of MAPs.[332–334] In some cases, SMP can be used as a matrix material to generate functional structures. Magneto-active structures offer rapid response and noncontact control properties.[335] Each part of a smart structure is magnetized in different directions for achieving complex deformation under magnetic stimulus.[336]

Magneto-active structures can be developed by using 3D printing techniques including DLP, DIW, and FDM.[337] 3D-printed magneto-active products are fabricated by uniformly dispersing soft MPs or through anisotropic magnetization during the curing process.[338–340] The latter is done by patterning MPs or magnetizing hard MPs into the polymer matrix. These structures transform their shapes, upon the application of a magnetic field.[341] Additionally, 3D printing technology is highly diverse technology that promotes the development of magnetic products.[342] In comparison to the DLP printing, FDM printing offers advantages like wide adaptability, no particle sedimentation, and low cost. Furthermore, low initial modulus of ink may cause complete collapse of 3D-printed structure, during DIW printing process.[343]

For instance, Cao et al.[344] employed thermoplastic rubber (TPR) filament to develop biomimetic magnetoactuators by using a FDM technique, as illustrated in **Figure 8**A. The results revealed that the carbonyl iron-incorporated actuator exhibited fast response, good controllability, and a programmable integrated structure.

In another study, Guan et al.[345] developed a soft robotic gripper through the DIW process by using MAP, as illustrated in Figure 8B. The result indicated that the 3D-printed gripper exhibited an excellent fast response rate and clamping force. Likewise, Yue et al.[346] incorporated cellulose nanofibers (CNFs) and iron oxide particles into PCL and poly-hydroxybutyrate (PHB) to fabricate magneto-responsive SMP composites. The results indicated that PCL/PHB/CNFs (5 wt%)/Fe_3O_4 (10 wt%)-based specimen showed a high tensile strength of 60.67 MPa. Figure 8C depicts a snowflake model, which quickly restored its actual shape in the presence of a magnetic field. In another study, Zhu et al.[347] developed polydimethylsiloxane (PDMS)/Fe-based composite ink to develop a magneto-responsive smart gripper with controllable and rapid response, as illustrated in Figure 8D. Similarly, Liu et al.[348] developed magneto-responsive PLA/thermoplastic polyurethane (TPU)/Fe_3O_4-based honeycomb lattice structure by using the FDM process, as illustrated in Figure 8E. The results indicated that the lattice structure exhibited an excellent recovery ratio (>91%), shape fix ratio (≈100%), with a quick magnetic response.

3.4. Electro-Active Polymers

Among other smart polymers, electro-active polymers (EAPs) are promising stimuli-responsive soft materials which exhibit a considerable change in size or shape under an electrical stimulation.[349–351] These smart materials offer a large amount of deformation under an electric field. This special feature can help these polymers to use for developing muscle-like actuators and artificial robots.[352] These polymers can endure a large amount of deformation and forces, while the conjugated EAPs also show low energy optical transition, low ionization potential, and high electron affinities.[353–355] These materials are used to develop drug delivery systems, tissue engineering scaffolds, actuators, power generators, and other different biomedical devices, thanks to their ability to convert different signals including thermal, mechanical, and magnetic into electrical ones.[356–358] Additionally, EAP-based structures can convert mechanical strain into electric signals that are highly useful for energy harvesting and sensing applications.[81,359,360]

Based on activation principles, these polymers are divided into ionic or electric groups. Electronic EAPs use electrostatic forces of electrodes to induce actuation for contracting the polymers. Examples of these EAPs include ferroelectric, electrostrictive, and piezoelectric.[361–363] However, ionic EAPs displace the ions contained in the polymeric material to trigger actuation. Conducting polymers, polyelectrolyte gels, and polymer–metal composites are some common examples of ionic EAPs.[364–366]

Dielectric elastomers (DEs) are typical EAPs, which form transducer devices when laminated between two compliant electrodes. The DE layer offers in-plane expansion and compression along its thickness, under a high electric field.[367–369] 3D printing of DE-based EAPs has sparked tremendous attraction in different engineering applications.[370–374] For instance, Gonzalez et al.[375] used TPU-based DE to print electro-active structure via FDM technique, as illustrated in **Figure 9**A. The results indicated that the ideal elongation performance and high dielectric constant of elastomer make it suitable for an EAP actuator. Similarly, Haghiashtiani et al.[376] successfully printed soft actuators using elastomer-hydrogel material which was capable of generating different bending modes, as illustrated in Figure 9B. The authors used different strategies to improve the performance of the current devices for achieving high actuation rates with a low electric field. The results revealed that the device deflection could be enhanced by decreasing the DE layer thickness. However, it increased the probability of dielectric breakdown. Surface modification through barium titanate can be used to improve the electromechanical strength and dielectric properties. Furthermore, multiple stacking of DE layers can also improve the actuation ability of the 3D-printed structure.

3.5. Humidity-Responsive Polymers

Humidity-responsive polymers or moisture-responsive polymers are generally synthesized with hydrophilic groups like carboxyl, amide, pyrrolyl, or hydroxyl, which trigger

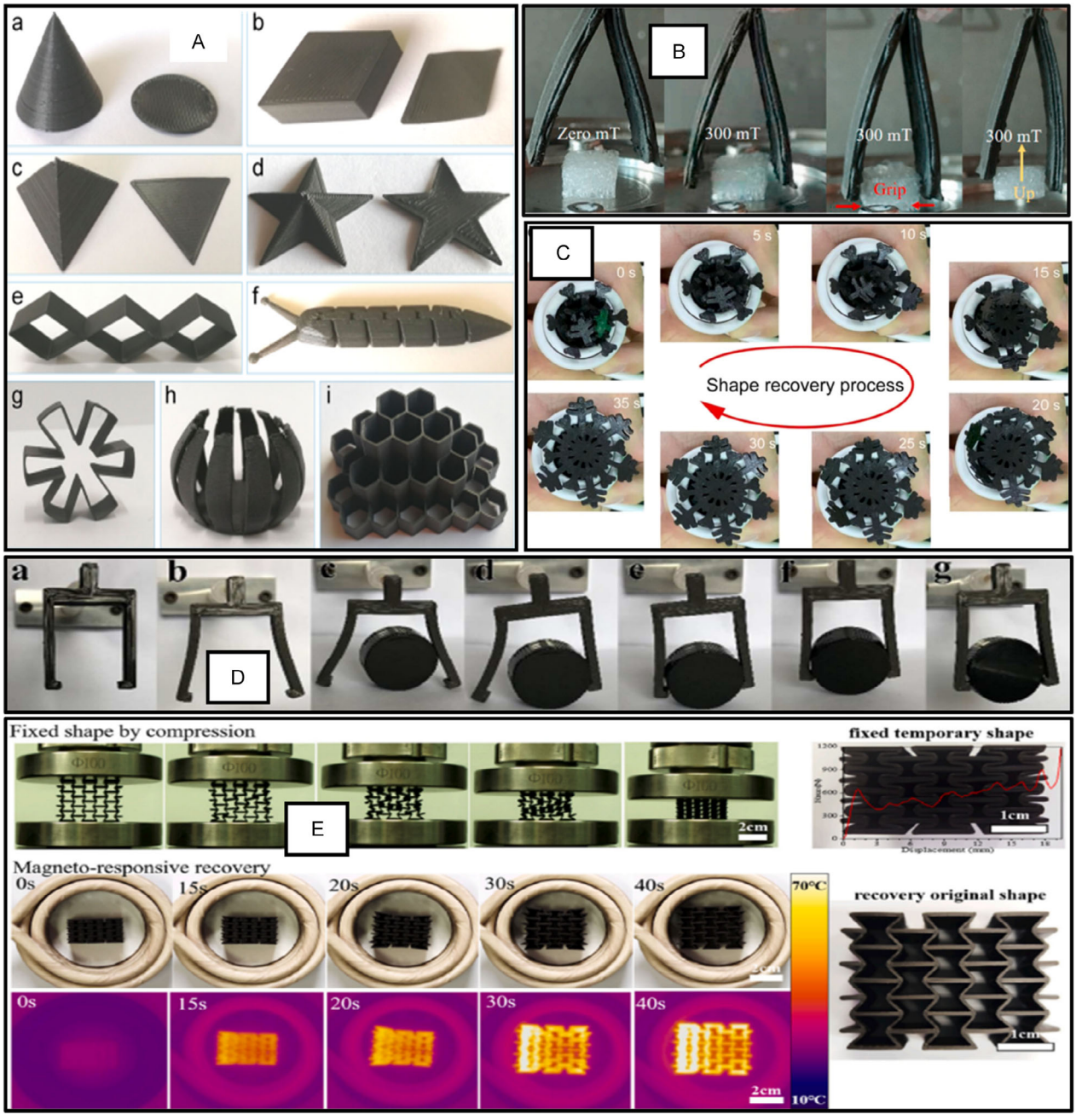

Figure 8. A) Different 3D-printed structures for biomimetic applications. Reproduced with permission.[344] Copyright 2021, American Chemical Society. B) Photographs of 3D-printed smart grippers performing grabbing upon exposure to magnetic stimulus. Reproduced with permission.[345] Copyright 2022, Elsevier Ltd. C) Magneto-responsive shape recovery behavior of 3D-printed snowflake model. Reproduced with permission.[346] Copyright 2021, Elsevier B.V. D) 3D-printed clamping jaw, which is responsive to external magnetic stimulus. Reproduced with permission.[347] Copyright 2018, American Chemical Society. E) 4D-printed PLA/TPU/Fe_3O_4-based honeycomb structure; E_1) fixed temporary shape during compression test; E_2) shape-memory behavior of structure upon magnetic stimulus. Reproduced with permission.[348] Copyright 2022, Elsevier Ltd.

humidity-driven reversible swelling for actuation by exhibiting shape-morphing behaviors like twisting, bending, locomotion, and spiraling.[377–379] These polymers offer fascinating dynamic and biomimetic responses, which permit them to develop structures for sensors, soft robots, artificial muscles, and smart electronic devices.[291,380–382]

3D printing of humidity-responsive polymers enables the development of intricate and programmable structures.[383–385] Sometimes, humidity-responsive polymers are designed to enhance the mechanical properties of 3D-printed structures.[386] For instance, Jiang et al.[384] presented an innovative approach for developing cross-linked poly(MAA-*co*-OEGMA)-based

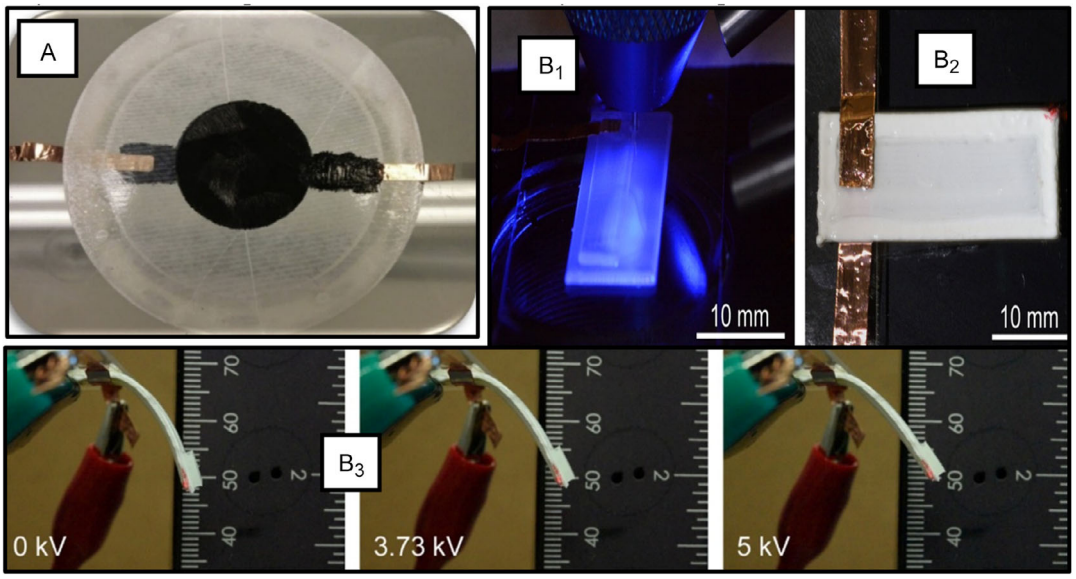

Figure 9. A) 3D-printed dielectric EAP actuator structure. Reproduced with permission.[375] Copyright 2019, Elsevier B.V. B_1,B_2) 3D printing of actuator consisting of passive layer, dielectric layer, and hydrogel electrodes; B_3) load lifting capacity of 3D-printed actuators under different values of voltages. Reproduced with permission.[376] Copyright 2018, Elsevier Ltd.

humidity-responsive polymer networks to print actuators by using a DIW printing, as illustrated in **Figure 10**A. The study revealed that the 3D-printed actuator exhibited excellent reversible and fast shape-morphing behavior, which would open new pathways for developing microfluidic and other biomedical devices. In another study, Yang et al.[385] proposed a controlled mechanism for programmable deformation of humidity-responsive polyethylene glycol diacrylate (PEGDA)-based hydrogel architectures manufactured through DLP, as illustrated in Figure 10B. Furthermore, the results revealed that a water-absorbing and swelling-powered manipulator exhibited excellent ability to grasp and release objects.

Cellulose fibers have also been applied to develop hygromorphic structures, due to their hydrophilic nature.[387] Furthermore, 3D-printed cellulose also offers an excellent ability to change its shape upon environmental stimuli such as moisture and water.[388–391] For instance, Gauss et al.[392] demonstrated the use of PLA/cellulose-based composites manufactured through the FDM technique to develop hydromorphic structure. The results that the 3D-printed structure exhibited excellent shape-changing behavior upon immersion in water are illustrated in Figure 10C. This technique can be used to print smart systems in the build environment.

3.6. pH-Responsive Polymers

pH-responsive polymers are polyelectrolytes, which contain basic or acidic groups in their crystalline structures. Such groups release or accept protons when pH changes externally. Sulfonic, pyridine, phosphate, carboxyl, and tertiary amine groups of these polymers ionize with the change in pH that changes the structural or functional properties such as degradability, solubility, chain conformation swelling, configuration, self-assembly, and surface activity.[393–395] pH-responsive systems consist of basic monomers, which act as anionic polymers under a basic environment and behave as cationic polymers under an acidic environment.[396] These polymers are usually derived from natural sources such as chitosan, gelatin, hyaluronic acid, dextran, and alginic acid.[397–399] However, some synthetic polymers like poly(L-glutamic acid), poly(aspartic acid), and poly(histidine) are also used for applications where the pH of an external environment changes.[400–402] These responsive polymers are highly suitable for personalized pharmaceutical applications like gene delivery, drug delivery, and glucose sensors, thanks to their biodegradable, biocompatible, and nontoxic nature.[403–405] pH differences have been found in human body; for instance, a basic environment is observed along the gastrointestinal tract, an acidic environment is found in the stomach, and hypoxic nature has appeared in the tumor tissue environment. That is why these polymers can be exploited for drug delivery and tissue engineering operations.[406–408]

3D printing of pH-responsive polymers has demonstrated its potential for developing personalized drug delivery systems, tissue scaffolds, wound dressings, mobile micromachines, and smart sensing devices, thanks to its multimaterial printing ability and geometrical flexibility.[409–414] For instance, Wu et al.[415] used ABS filaments and poly(4-vinylpyridine)-incorporated ABS filaments to develop a pH-sensing claw, which exhibited reversible geometric changes, as illustrated in **Figure 11**A. This multimaterial printed sensing claw allowed reliable pH measurements of complicated samples. Thus, such an approach is highly suitable for smart sensing devices. In another study Hakan et al.[414] microprinted pH-responsive microswimmers for controlled cargo delivery (Figure 11B), using a combination of blood-derivable biomaterials (e.g., serum albumin protein, blood plasma, and platelet lysate). The results indicated that microswimmers exhibited an excellent response to pH

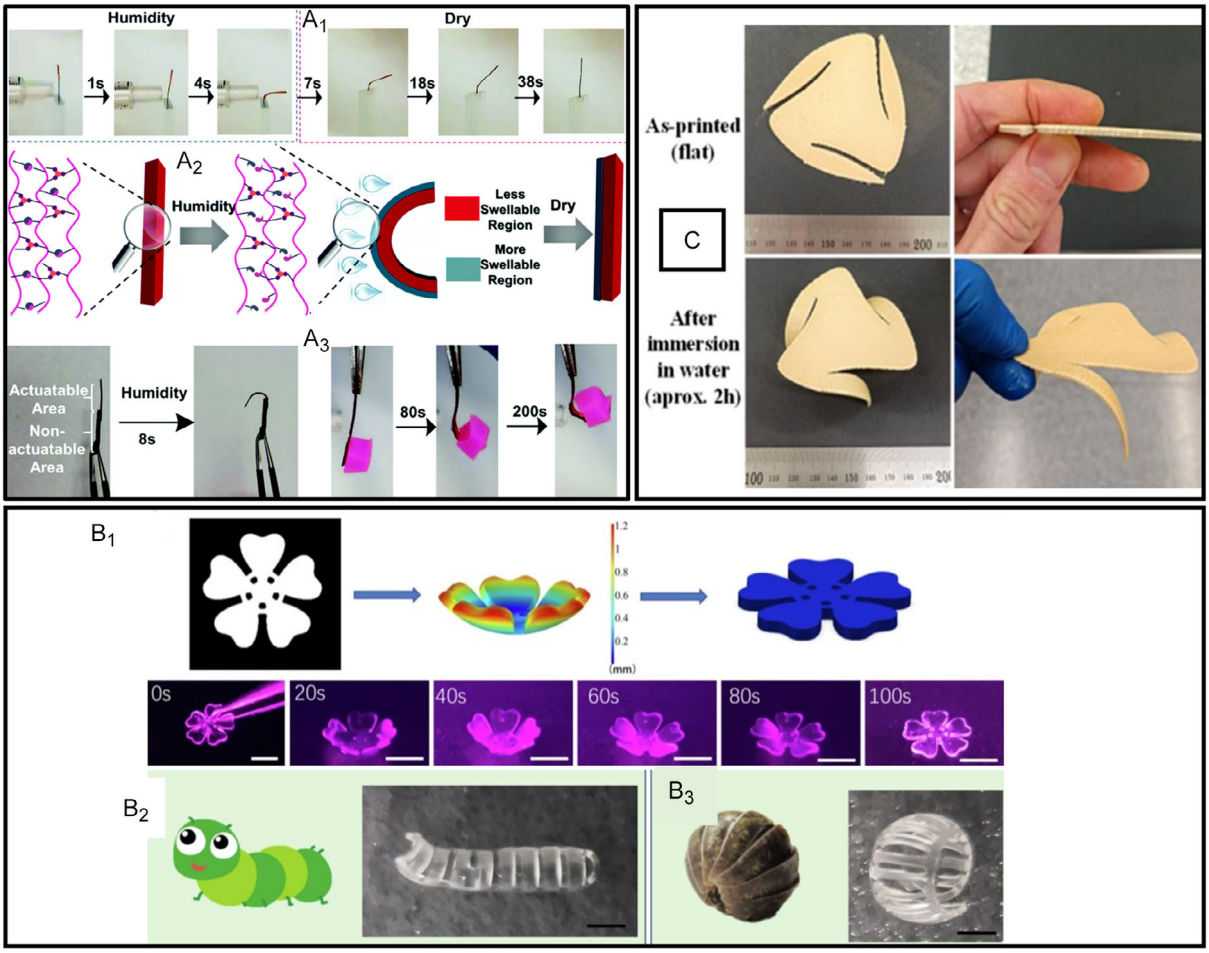

Figure 10. A) Humidity-driven double cross-linked polymer; A_1) shape-memory behavior of 3D-printed upon humidity as a stimulus; A_2) schematic illustration showing the development of cross-linked hydrogel; A_3) actuator showing deformation from the thinner region and lifted the loaded silicone. Reproduced under the terms of the CC-BY-NC license.[384] Copyright 2021, The Author(s), Published by the Royal Society of Chemistry. B_1) Folding and unfolding mechanism of floral structure; B_2) caterpillar bionic structure responsive to water stimulus; B_3) curling process of pill bug. Reproduced with permission.[385] Copyright 2022, Elsevier Ltd. C) Cellulose/PLA-based 3D-printed composites before and after immersion in water at 45 °C. Reproduced with permission.[392] Copyright 2023, Elsevier Ltd.

and can be tailored to develop personalized micromachines for special biomedical applications.

3.7. Multi-Responsive Polymers

Multi-responsive or dual-responsive polymers can respond to the combination of external stimuli, triggering them to a variety of material changes with high accuracy.[416] The combination of two or more responsive materials generates mismatch strain within the 3D-printed structures and can help to introduce multidimensional cues.[417–419] These polymers offer relatively higher controllability and create multifunctional structures and permit the fabrication of highly precise and intricate structures with more favorable mechanical characteristics.[420–423] Multiple types of polymers such as thermo-photo, thermo-magnetic, thermo-pH, and others have been used for tissue regeneration applications in real physiological conditions. AM of multi-SRPs has been gaining significant attraction in recent years.[424–427] For instance, Cremonini et al.[425] developed a novel oligomeric ink containing azobenzene photo-responsive moiety to fabricate a multi-responsive LCE-based actuator using DIW printing, as illustrated in **Figure 12**A. The study demonstrated that 3D-printed structure exhibited specific responses (bending and swelling) depending on light, heat, and water stimuli.

In another study, Hu et al.[426] developed a multifunctional magneto- and thermo-responsive soft millirobot by using the photopolymerization technique, as illustrated in Figure 12B. PNIPAm/NdFeB/laponite nanoclay-based smart hydrogel exhibited simultaneous magnetic actuation and temperature sensation. Similarly, Ren et al.[427] proposed a novel approach to develop PU/polyvinyl chloride (PVC)-based multi-responsive bilayer structures, which responded to both water and temperature using a DIW printing technique, as illustrated in Figure 12C. The study demonstrated that the bilayer structure exhibited excellent shape-shifting behavior.

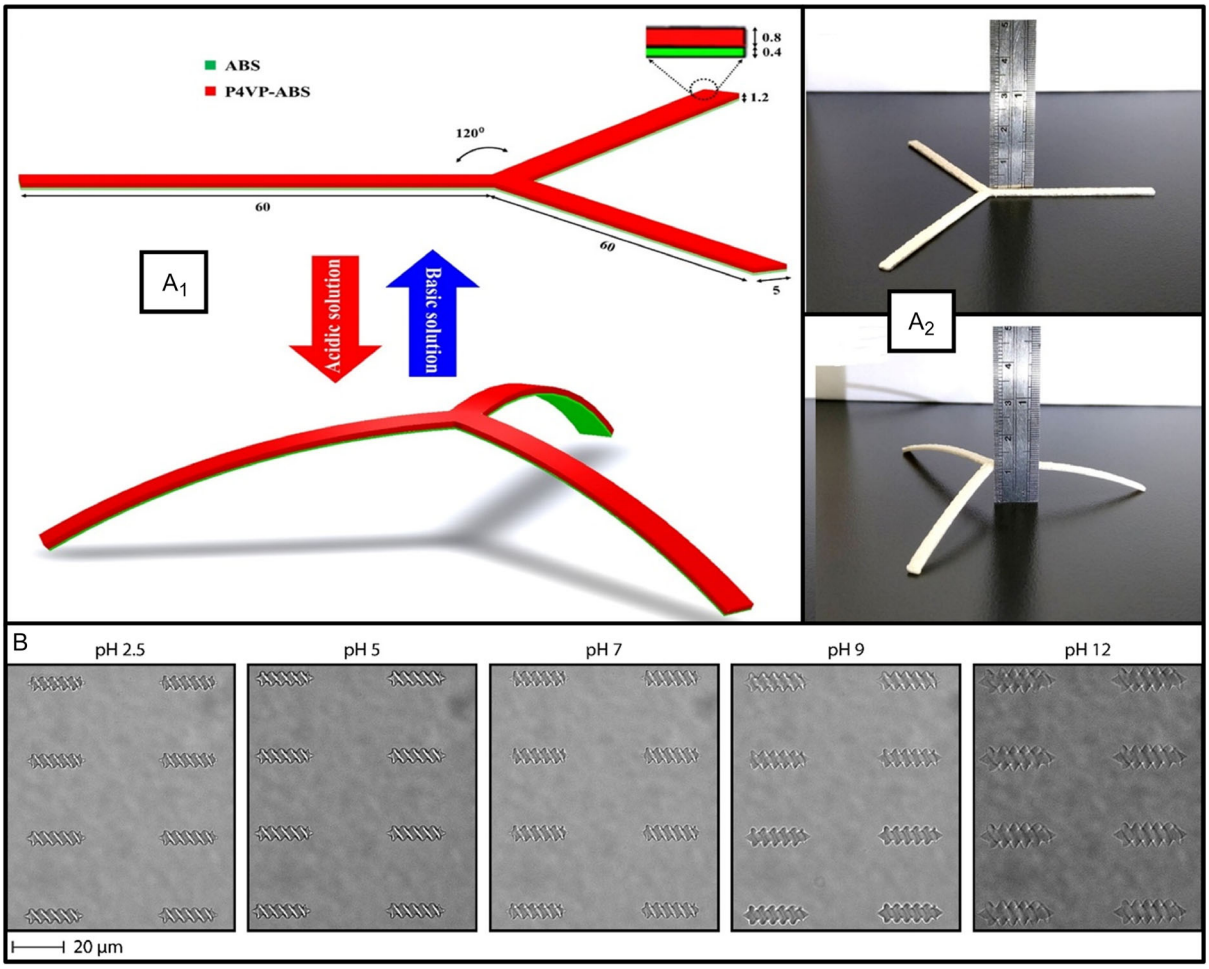

Figure 11. A_1) 3D-printed pH-responsive claw, which consist of three cuboids; A_2) photographs showing the shape-morphing behavior of claw, when exposed to pH stimulus. Reproduced with permission.[415] Copyright 2022, Elsevier B.V. B) Shape-memory behavior of pH-responsive albumin microswimmers. Reproduced under the terms of the CC-BY license.[414] Copyright 2021, The Authors, some rights reserved; exclusive licensee American Association for the Advancement of Science.

3.8. Self-Healing Polymers

Self-healing polymers are also considered SRPs, which exhibit the extraordinary intrinsic ability to heal damaging areas.[428] The self-healing feature enables macrostructural restoration and functional recovery of polymers.[429] Consequently, it enhances the safety and service life of materials without any human intervention.[430] Self-healing technology has been growing continuously and develops new polymers and their composites, and smart materials with self-healing features.[431] Additionally, it is difficult for stimuli-responsive materials to withstand fractures or scratches during their service time.[432] Furthermore, responsive materials are more prone to fatigue and form cracks during the shape-morphing process.[433] The amalgamation of 4D printing with self-healing materials brings two objectives under one umbrella, SRPs with self-healing advantage.[434] Recently, 3D-printed dynamic structures with the capability to self-heal cracks or fractured regions have been developed by researchers. For instance, Ma et al.[435] modified epoxy resin with PU for improving its toughness and reducing glass transition temperature. In the second step, the Fe_3O_4@graphene oxide (GO) module is mixed with epoxy-modified PU. Finally, the self-healing microcapsule module containing isophorone diisocyanate, as a healing agent, was incorporated into the above mixture to develop multifunctional SRPs. The results indicated that 3D-printed composites exhibited excellent self-healing ability (91.2%), as illustrated in **Figure 13**A. Additionally, these composites showed excellent shape-memory performance under the magnetic stimulus. Thus, these SRP composites can be used to develop bionic grippers.[340]

In another study, Yu and co-workers[436] developed self-healable elastomers based on sulfide bonds and fabricated different self-healable single and multimaterial structures including soft grippers (Figure $13B_1$) and architected electronics (Figure $13B_2$) using μSLA printing. Thus, self-healing polymers can enhance the lifetime and reliability of materials. Furthermore, these polymers can be recycled thanks to their self-healing capability, which would reduce material consumption.

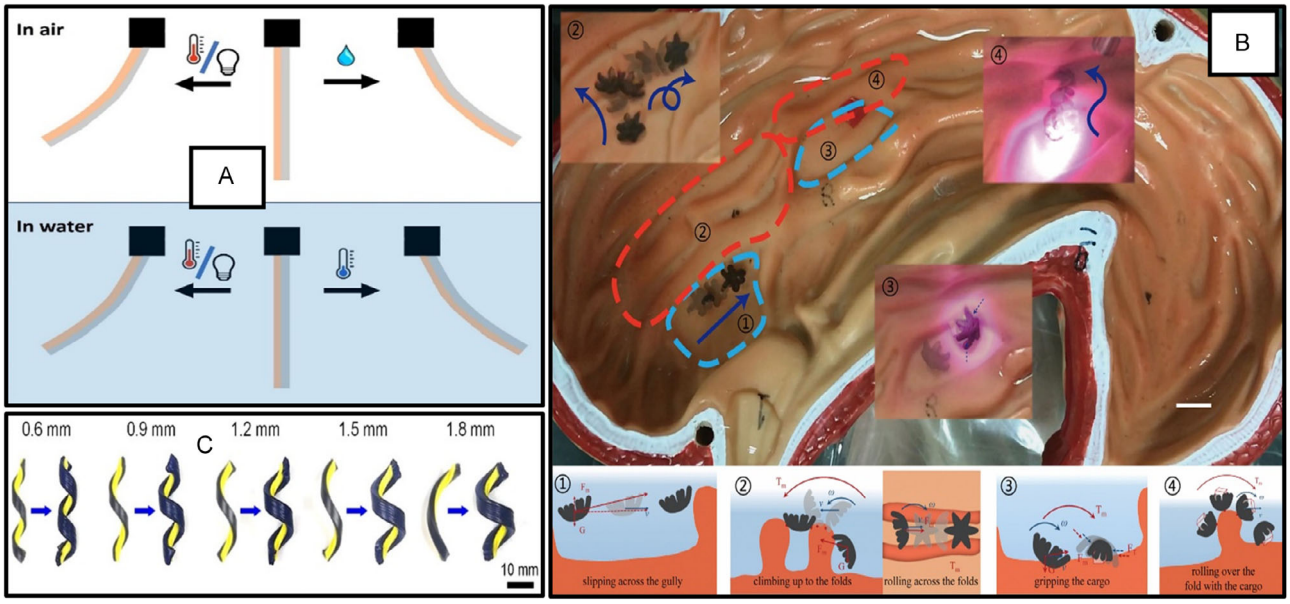

Figure 12. A) Multi-responsive actuator developed by DIW printing. Reproduced with permission.[425] Copyright 2023, Wiley-VCH GmbH. B) Multi-functional leptasteria-like smart gripper completes active delivery in stomach model. Reproduced with permission.[426] Copyright 2021, Elsevier Ltd. C) Spiral deformations of PU/PVC-based bilayer structures obtained by varying the thickness of PU layer. Reproduced under the terms of the CC-BY license.[427] Copyright 2021, The Authors, published by Frontiers.

4. SRP-Based Metamaterials

Metamaterials can be used to fabricate dynamically responsive structures with extraordinary properties like negative variable stiffness and negative Poisson's ratio.[151,437–439] These structures have drawn growing research interests, thanks to their distinct functions and potential applications in deployable structures and reversible energy absorption devices.[440–443] Mechanical properties of these materials emerge from the 3D spatial orientation of the microlattice structures.[444] SRP-based metamaterials with large deformation, reconfigurable, programmable, and tunable properties have been developed by different researchers.[445–447] For instance, Yang et al.[448] presented functionally deployable, geometrically reconfigurable, and mechanically tailorable lightweight SMP-based metamaterials, developed through μSLA printing. These programmable structures exhibited excellent shape-memory behavior within a temperature range 30–90 °C, as illustrated in **Figure 14**A. These metamaterials can be used in a variety of applications ranging from morphing aerospace structures to tunable shock absorbers.

In another study, Ding et al.[449] fabricated SMP/elastomer lattice multimaterial structures, as presented in Figure 14B. The results revealed that these programmable structures exhibited excellent deformation and shape-memory behavior, which makes them highly suitable for aerospace and biomedical applications. Similarly, Zhang et al.[450] printed a 2D lattice structure via FDM printing, which triggered pattern transformation in a controllable way, as shown in Figure 14C_1. Additionally, the shape transformed into a flower-like shape under heat stimulation, when the polymer printed on the paper sheet (Figure 14C_2). In summary, shape morphing-based metamaterials with lightweight, high strength/stiffness, and energy-absorbing capabilities can develop intricate and dynamic structures for multifunctional applications in biomedical, aerospace, and marine engineering.

5. Applications of Additively Manufactured SRPs

SRPs exhibit shape deformation when triggered through an external stimulus like humidity, heat, light, pH, electricity, solvents, or magnetism. These polymers have been applied in the field of soft robotics, artificial muscles, tissue engineering, wound dressing, aerospace, and other applications,[451–459] as illustrated in **Figure 15**. Overall, **Table 3** presents a summary of different 3D-printed soft responsive structures developed by different researchers using SRPs for various engineering applications.

5.1. Biomedical Sector

3D printing of stimuli-responsive materials enables the formation of intricate and dynamic structures with high resolution, which makes it highly suitable for biomedical engineering including drug delivery, tissue engineering, and wound healing.[460–464] It also helps to construct cell-laden smart structures that closely mimic living tissues. Accidents are part and parcel of human life and may damage and fracture hard or soft tissues like bone defects or ligament rupture.[465–468] These fractures of complex structures can be solved by using the combination of SRPs and AM to fabricate dynamic and patient-specific scaffolds, which permit the regeneration of tissues.[469–471] Additionally, the flexibility and minimally invasive implantation of 4D-printed scaffolds make them highly suitable for bone

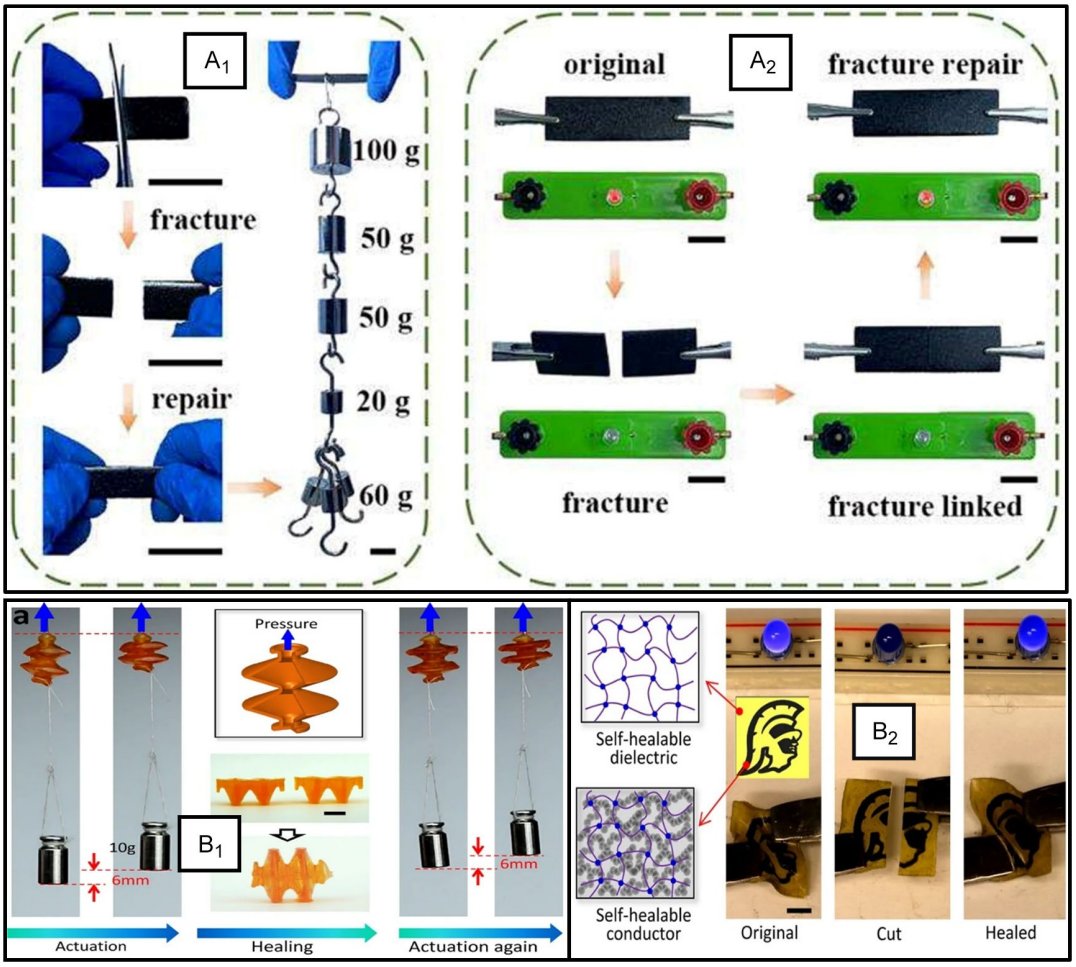

Figure 13. A) Self-healing performance of 3D-printed SRP composites; A_1) self-healing nature of smart polymer fractured upon lifting 280 g weight; A_2) self-healing ability of switch of LED lamp. Adapted with permission.[435] Copyright 2023, Elsevier B.V. B_1) self-healing process of 3D-printed actuator based on disulfide bond; B_2) self-healed electric structure. Adapted with permission.[436] Copyright 2019, Springer Nature.

treatments.[472] Thermo-responsive SRP composites are generally applied to develop bone scaffolds.[473] For instance, Senatov et al.[474] printed PLA/HAp-based thermo-responsive porous scaffolds using the FDM process for bone regeneration, as illustrated in **Figure 16**A. Such 3D-printed scaffolds exhibited excellent mechanical, shape recovery, and self-healing properties.

3D-printed SRPs can be used to develop pipe-like structures or blood vessels.[475] In this approach, a flat construct is printed, which folds upon receiving an external stimulus to generate a pipe-like vessel.[476] The diameter of the 4D-printed vascular vessel ranging from several micrometers to centimeters permits the homogeneous encapsulation of blood cells.[477] For instance, Constante et al.[478] combined the extrusion-based printing of alginate methacrylate and melt-electrowriting of PCL fibers to develop multimaterial scaffold capable of self-folding upon stimulation. The shape morphing of bilayer scaffold was controlled by regulating the calcium-ion concentration. The results indicated that 3D-printed scaffolds exhibited good cellular proliferation and viability.

Medical stents are tubular structures, which are applied to remove the constriction of vessels like bronchi, esophagus, blood vessels, and urethra.[371,479–484] However, traditional stents require replacement after approximately 6 months of implantation, due to a high rate of restenosis.[485] Additionally, stents can also become dysfunctional owing to cell regeneration and thrombosis disease.[486] SMP-based stents also reduce the risk of restenosis and thrombosis.[487] In recent years, SMP stents developed using unparalleled 3D printing technology have gained enormous attraction, thanks to their excellent biodegradability and biocompatibility.[488] This technology also permits the fast fabrication of personalized and intricate stents of a vast range of sizes with absolute accuracy.[489] For instance, Zhou et al.[490] fabricated a thermo-responsive shape-memory vascular stent using the combination of DIW printing and βCD-g-PCL, as illustrated in Figure 16B. The results indicated that the 3D-printed stents showed excellent tensile strength, sufficient elasticity, biocompatibility, and sustained drug release. Additionally, the self-expandable nature of the 3D-printed stent can help in reducing the surgical procedure.

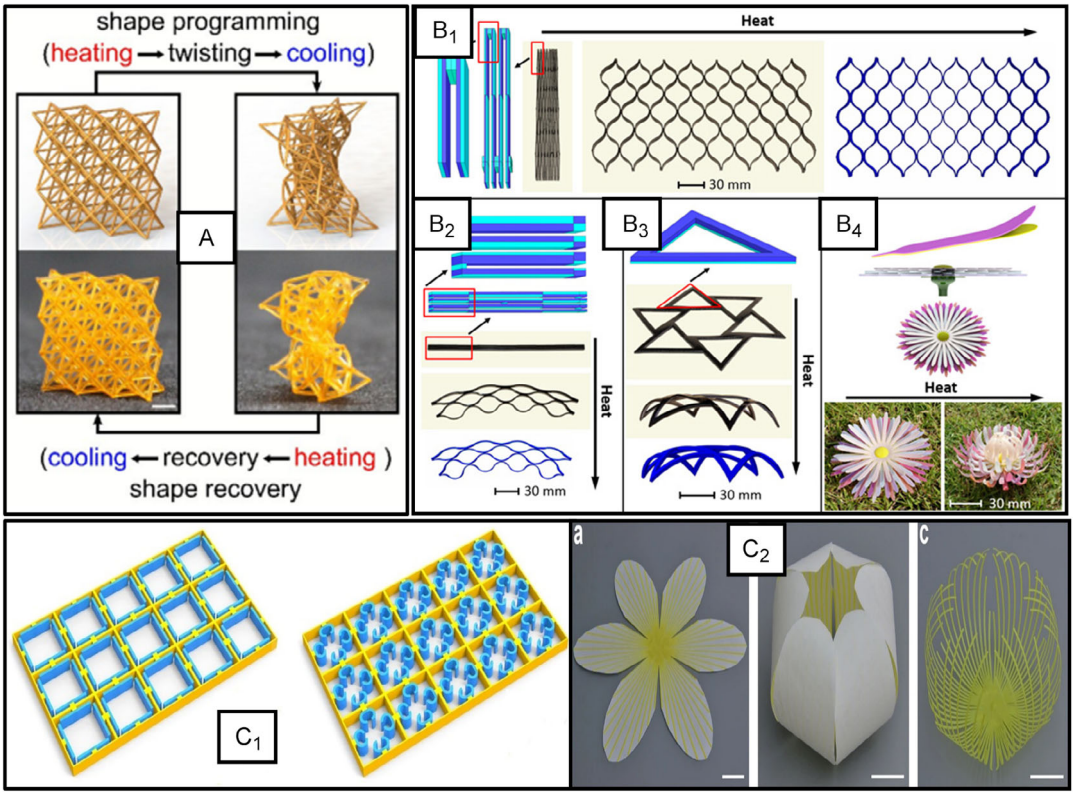

Figure 14. A) Shape-memory behavior of 3D-printed photocurable metamaterials under heat stimulus. Adapted with permission.[448] Copyright 2019, Royal Society of Chemistry. B) 4D-printed SMP/elastomer lattice structure; B_1) collapse configuration of printed lattice structure; B_2) bending and expansion of structure upon heating; B_3) flat star-shaped architecture that deploys into a dome; B_4) 3D-printed thermo-responsive flower with multiple petals. Reproduced under the terms of the CC-BY license.[449] Copyright 2022, The Authors, some rights reserved; exclusive licensee American Association for the Advancement of Science. C_1) Adaptive metamaterial, which transformed its configuration (curved beams) upon heat stimulation; C_2) a complex flower-shaped printed on the paper sheet. Reproduced under the terms of the CC-BY license.[450] Copyright 2016, The Authors, Published by Springer Nature.

Figure 15. Schematic illustration of additively manufactured SRPs applied for different applications.

Additively manufactured SRPs are also used in pharmaceutical applications to regulate drug delivery.[491] Mainly two approaches are used to deliver drugs using smart devices. One way is to incorporate the drug in the bioink and the drug can be released after the implantation of the 3D-printed scaffold.[492] However, the sustained release of drugs is not possible with this approach. This issue can be solved by designing intelligent polymer chains or deformable devices through 3D printing.[493–496] This approach helps a drug-loaded stimuli-responsive capsule release the medicine in a controlled manner under a specific stimulus at a specified body location.[497–499] Unlike traditional manufacturing methods, 3D printing is highly advantageous to develop personalized, made-on-demand, and unprecedented complex structures, which enhance the effectiveness, accessibility, and safety of drugs.[500–504] For instance, Melocchi et al.[505] proposed an expandable system for gastric retention by assessing the shape-morphing behavior of pharmaceutical-grade polyvinyl alcohol (PVA). The authors successfully manufactured different original configurations to facilitate gastric retentions, as illustrated in Figure 16C. Prototypes containing allopurinol were fabricated either by a FDM technique or by manual processing of hot melt extruded rods. The results indicated that submerged prototypes in 0.1 hydrochloric solution at physiological temperature recovered their original shapes.

Table 3. 3D-printed smart structures developed for different applications.

AM technology	Materials	Stimulus	Stimulus response	Applications	References
DIW	PDMS/iron particles	External magnetic field	Bending	Bionic robots	[347]
DIW	Natural rubber/carbonyl iron	Magnetic field	Bending and gripping	Soft robotic grippers	[345]
DIW	PDMS/NdFeB	Magnetic field	Bending and gripping	Medical robots	[675]
DIW	PDMS/ZrO_2	Heat	Shrinking	Aerospace components	[676]
DIW	PDMS/carbon/Fe	Magnetic field	Bending and rolling	Underwater soft robots	[677]
DIW	PNIPAm/Fe_3O_4	Magnetic field	Bending and stretching	Hyperthermia cancer therapy	[678]
DIW	PCL/PLA	Heat	Bending	Tracheal scaffolds	[679]
DIW	βCD-g-PCL	Heat	Shrinking	Vascular stent	[490]
DIW (gel extrusion)	PCL/PDA	NIR radiation	Bending and shrinking	Localized cancer therapy and wound healing	[328]
DIW	Gelatin/EC	Water	Swelling	Edible composites	[680]
DIW	s-PCL-MA	Heat	Shrinking	Deployable medical devices	[681]
DIW	PU/PVC	Heat and water	Swelling and bending	Soft gripper	[427]
DIW	PLMC/CNT	Electric	Folding	Liquid sensors	[372]
DIW	PLMC/PTMC/Fe_3O_4	Heat and magnetic field	Bending	Multifunctional devices like soft robots	[682]
DIW	TPU/PCL/Fe_3O_4	Heat and magnetic field	Bending and grasping	Flexible robotics	[683]
DIW	PLA/benzophenone/Fe_3O_4	Magnetic field	Shrinking	Cardiovascular implant	[333]
DIW	LCE	Heat	Rolling	Untethered self-propelling soft robots	[541]
DIW	Poly(MAA-co-OEGMA)	Humidity	Swelling	Catalysis devices and smart actuators	[384]
DIW	PCLDMA/SAMA	Heat and ionic	Self-rolling	Vascular stent	[684]
DIW	PU/LNTs	Light	Bending and shrinking	Soft robots	[327]
DIW	LCE	Light, heat, and water	Bending, folding, and shrinking	Light-driven swimming objects	[425]
DIW	PLA/PCL/PU	Heat	Shrinking	Soft robotics	[685]
DIW	Zein protein	Solvent	Rolling for tubular conduits	Nerve regeneration	[686]
Extrusion	Collagen/alginate	pH	Swelling	Cartilage tissue engineering	[687]
Extrusion	PU elastomer	Water and heat	Swelling and shrinking	Drug delivery device	[688]
Extrusion	Gelatin/SC/β-TCP	NIR	Shrinking	Treatment of bone cancer	[516]
Extrusion	PCL/PANI	Electric	Bending	Bone tissue regeneration	[689]
Extrusion	PPy/gold	Electric	Rolling and gripping	Soft microactuators	[690]
Extrusion	PCL/graphene	Electric	Bending	Bone scaffolds	[691]
Extrusion	PEGDA/iron particles	Magnetic field	Bending and folding	Soft robotics and magnetic actuators	[692]
FDM	TPU/PLA/PANI	Light and heat	Shrinking and bending	Smart actuators	[693]
FDM	PLA/cellulose	Water	Swelling and shrinking	Hygromorphic structures for build environment	[392]
FDM	PVA	Heat	Bending	Intravesical and gastro-retentive and drug delivery systems	[694]
FDM	PLA/PBS/GO	NIR light	Bending	Porous scaffolds	[479]
FDM	PLA	Heat	Bending	Smart textiles	[551]
FDM	PLA/Fe_3O_4	Magnetic field	Expansion and stretching	Patient-specific absorbable left atrial appendage occluder	[695]
FDM	Chitosan/PLA	Heat	Bending and shrinking	Patient customized scaffolds	[696]
FDM	PLA/Fe_3O_4	Heat and magnetic field	Shrinking and bending	Bone tissue engineering	[207]
FDM	PLA/Fe_3O_4	Magnetic field	Bending and shrinking	Tracheal scaffolds	[697]
FDM	PLA/TPU/Fe_3O_4	Magnetic field	Folding and gripping	Smart actuators	[348]

Table 3. Continued.

AM technology	Materials	Stimulus	Stimulus response	Applications	References
FDM	PLA/Hap	Heat	Self-healing and bending	Self-healing bone implants	[474]
FDM	ABS	Heat	Origami and shrinking	Self-assembly structures	[698]
FDM	PLA/iron particles	Magnetic field	Bending and gripping	Smart grippers	[699]
FDM	PEEK/Fe_3O_4	Magnetic field	Bending and folding	Space compliant electrical motors	[700]
FDM	PCL/PHB/CNFs/Fe_3O_4	Magnetic field	Bending	Smart actuators	[346]
FDM	PCL/PLA	Heat	Folding	Medical protective devices	[292]
FDM	PLA	Heat	Spiraling	Self-spiraling pattern driven actuator	[701]
FDM	PLA/iron	Magnetic field	Folding	Hyperthermia treatment	[702]
FDM	PLA/CNTs	Electric	Bending and folding	Embeddable sensors	[546]
FDM	PCL/TPU	Heat	Bending and grasping	Bionic grippers	[301]
FDM	PLA/TPU/CNT/carbon fibers	Electric	Shrinking	Adaptive energy absorption devices	[439]
FDM	PLA/PEU/MWCNTs	Electric	Bending and grasping	Soft robotics and actuators	[703]
FDM	PLA/CB	Light	Shrinking and origami	Smart electronics, origami structures, and robot precision control.	[326]
FDM	PLA/CNTs	Electric field	Folding	Electroactive deformable devices	[371]
FDM	PLA	Heat and mechanical	Shrinking and origami	Energy absorbing devices	[704]
FDM	PVA/glycerol	Heat and fluid	Swelling and unfolding	Drug delivery device for gastric retention	[505]
FDM	PCL/PBAT/CNFs	Heat	Bending and folding	Bionic soft gripper	[542]
FDM	SEBS/PEW/low density polyethylene (LDPE)	Heat	Folding and bending	Smart grippers	[705]
DLP	PDMS/ferrofluid	Magnetic field	Bending	Soft gripper	[706]
DLP	AA/PEGDA	pH	Swelling, shrinking and grasping	Controlled drug releasing device	[707]
DLP	PEGDMA	Heat	Folding	Microfluidic devices	[520]
DLP	PEGDMA	Water	Swelling	Bionic structures	[385]
DLP	GelMA/PEGDMA	Hydration	Swelling	Tissue scaffolds	[708]
DLP	PAA	Electric field	Folding and gripping	Soft robotic actuator	[709]
SLA	Acrylate-epoxy hybrid photopolymer	Heat	Shrinking	Deployable structures	[710]
SLA	Acrylic resin	Heat	Shrinking and folding	Biomedical devices	[711]
SLA	PEGDMA/CNF/chitosan	Heat	Folding	Tissue engineering	[712]
μSLA	PNIPAm	Heat	Bending	Soft gripper	[713]
μSLA	PEGDMA/AA	pH	Swelling and shrinking	Tissue scaffolds	[412]
TPP	Elastomer	Heat	Bending	Photonic devices	[714]
TPP	Elastomer/NPs (gold nanorods, GO, MWCNTs)	Light	Folding and bending	Biomedical sector	[715]
TPP	PEGDA	pH	Swelling	Biosensors	[716]
TPP	Blood-derived biomaterials	pH	Swelling	Microswimmers	[414]
IJP	PCL/CNTs	Electric field	Folding	Flexible electronic devices	[717]
Laser cladding	PLA	Heat	Bending and stretching	Flexible electronics and scaffolds	[442]
SLS	PA-12/γ-Fe_2O_3	Magnetic field	Bending and grasping	Smart grippers	[718]
SLS	TPAE	Heat	Bending and folding	Smart architectures like soft robots	[719]
Vat photopolymerization	PNIPAm/NdFeB/laponite nanoclay	Heat and magnetic field	Bending and folding	Soft millirobots	[426]

Chemotherapy, an anti-cancer agent, is effectively applied for the clinical treatment of cancer. However, this treatment also affects healthy tissues and organs.[506] Most strikingly, AM of SRPs can be used to print personalized drug-loaded systems with precise control for localized cancer treatment.[507–509] 3D printing is useful, especially for the treatment of local tumor residuals,

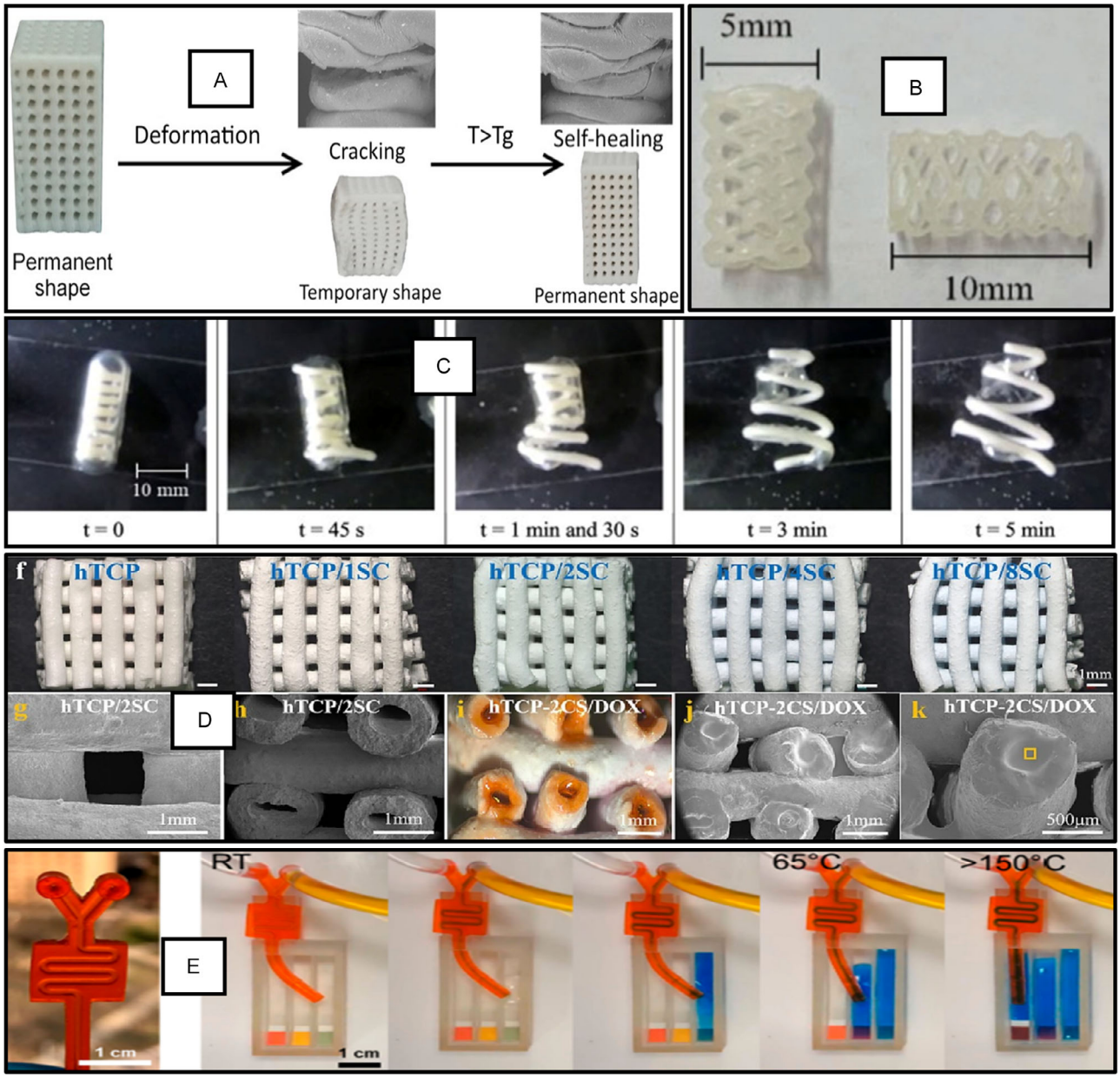

Figure 16. A) PLA/HAp-based scaffold, which exhibited excellent shape memory and self-healing behavior. Adapted with permission.[474] Copyright 2015, Elsevier Ltd. B) Photograph 3D-printed vascular stent. Adapted with permission.[490] Copyright 2021, Wiley-VCH GmbH. C) Shape recovery performance of a conical helix-shaped expandable drug delivery system. Adapted with permission.[505] Copyright 2019, Elsevier B.V. D) Optical and microscopic images of DOX-loaded gelatin/SC/TCP scaffolds. Reproduced with permission.[516] Copyright 2023, Elsevier B.V. E) 4D-printed microfluidic device. Reproduced with permission.[520] Copyright 2019, American Chemical Society.

and enhances the efficiency of drug administration and minimizes the distribution and side effects of drugs in other parts of the body.[510–514] For instance, Wang et al.[515] printed a PLA-based drug delivery device, which has shown excellent ability to carry anticancer drugs. Furthermore, this implantable device exhibited fine-tuned morphology, controllable micropore structure, and biocompatibility. In another study, Zhang et al.[516] developed doxorubicin (DOX)-loaded gelatin/$SrCuSi_4O_{10}$ (SC)/beta-tricalcium phosphate (β-TCP)-based core–shell scaffolds (Figure 16D) for the treatment of bone cancer. The results revealed that SC nanosheets endowed the scaffold with a photothermal effect as well as triggered the on-demand release of drugs under NIR stimulus. Overall, 3D/4D printing of stimuli-responsive materials permits the synthesis of precise, sophisticated, and controllable drug delivery systems.

In recent years, microfluidic devices are manufactured through the 3D printing of SRPs, and these devices can be applied for different biomedical applications such as cell

separation, cancer diagnosis, organ-on-a-chip, and tissue engineering.[517–519] For instance, Peng et al.[520] used a triple SMP composite to develop a shape-shifting and customized microfluidic device by using a DLP technique, as depicted in Figure 16E. The results revealed that the 3D-printed channel pointed toward the middle groove at a temperature of 65 °C and transformed into a straight shape when heated above 150 °C.

In summary, AM of SRPs has vastly benefited the biomedical industry and opened up new possibilities for creating customized implants, brain models, intestinal defect repair, bionic artificial muscles, and prosthetics. These smart implants can incorporate shape changes for a more precise fit in response to body temperature changes and can conform to irregular tissue defects, perfectly, hence reducing the risk of complications. Additionally, in situ damage to tissues can be eliminated by implanting smart scaffolds through a minimally invasive procedure. Surgical instruments manufactured through AM can be specifically designed to meet the bespoke needs of a particular surgery, thus reducing the risk of complications.

5.2. Soft Robotics

There has been growing interest in the 3D printing of SRPs, especially EAPs, thanks to their ability to develop complex responsive structures for smart grippers, actuators, and soft robots.[521–524] These smart structures use the inherent properties of soft materials to fabricate robots for performing complicated tasks with minimal actuation.[306,525–529] Table 3 also incorporates the recent fabrication of soft robots for different applications. The integration of 3D printing and SRPs can help to develop soft robotics of a variety of sizes for performing complex functions with minimal manipulation.[530–533] Recent studies showed the development of smart grippers, actuators, and robots for different applications.[534–540] For instance, Cecchini et al.[381] exploited the reshaping ability of humidity-responsive materials to develop a seed-like soft robot by using FDM printing, as illustrated in **Figure 17**A. In this approach, PCL-based filament were used to print a passive layer and electrospun cellulose nanocrystals (CNCs)/polyethylene oxide (PEO)-based was used as an active layer to develop bilayer hygroscopic structures. The results revealed that the 3D-printed robot mimicked the movement and performance of the natural seed. Furthermore, it showed the ability to lift weight 100 times more than its self-weight.

In another study, Zhai et al.[541] manufactured a LCE-based robot, which exhibited untethered self-propelled rolling upon heating above 160 °C. Additionally, the sustainable directional movement of the 3D-printed robot was provided by the curvature of the tubule, as illustrated in Figure 17B. Furthermore, Gu et al.[542] printed an intelligent bionic gripper by using PCL/poly(butylene adipate-*co*-terephthalate) (PBAT)/CNF-based thermo-responsive composites, as illustrated in Figure 17C. The results revealed that the 3D-printed structure exhibited a fast response with an excellent shape-memory effect.

5.3. Electronic Devices

3D printing of SRP composites has also gained tremendous attraction in the electronics field, thanks to their inherent advantages including controllable structural prototyping.[543]

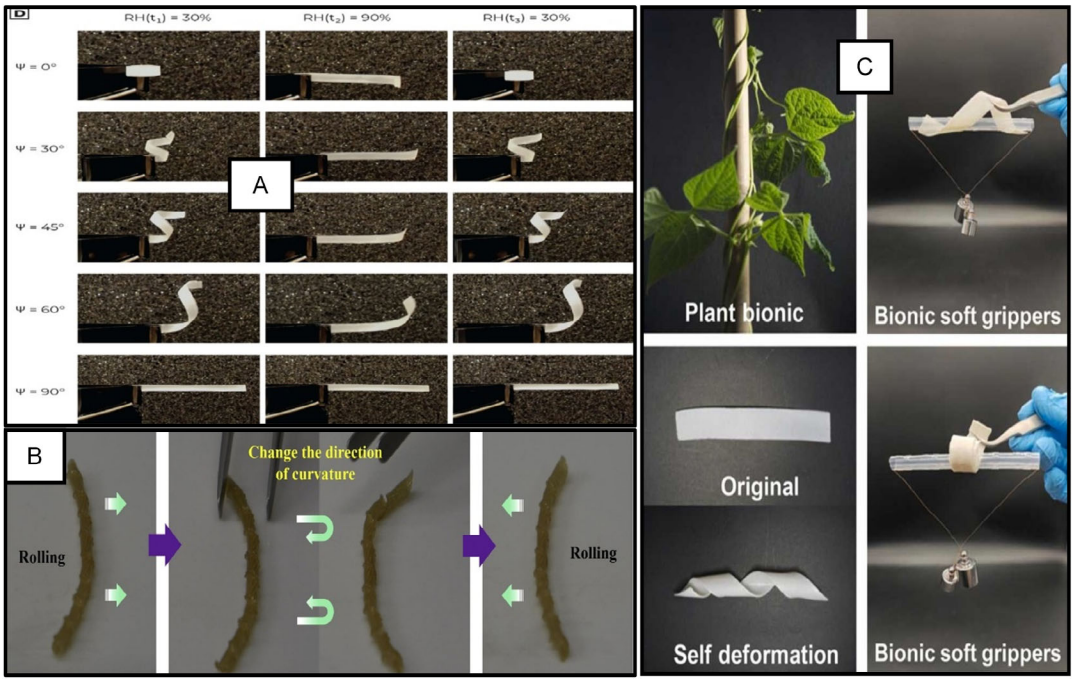

Figure 17. A) Reshaping behavior of humidity-responsive robots under different humidity. Reproduced under the terms of the CC-BY license.[381] Copyright 2023, The Authors, Advanced Science published by Wiley-VCH GmbH. B) Rolling direction of LCE-based robot fabricated by using DIW printing. Reproduced with permission.[541] Copyright 2021, Elsevier Inc. C) PCL/PBAT/CNF-based thermo-responsive bionic gripper. Adapted with permission.[542] Copyright 2023, Elsevier B.V.

The integration of conductive materials into SMPs results in the development of flexible electronic devices.[544,545] For instance, Mousavi et al.[546] used a CNT-reinforced PLA-based composite to fabricate a piezoelectric soft tactile sensor using the FDM printing technique. The authors used a sandwich design to assess anisotropic tactile sensing under large deformation, and PLA/CNT sensors were placed between TPU layers, as illustrated in **Figure 18**A. The results revealed excellent tensile and bending properties along with high sensitivity.

In another study, Bon et al.[547] combined poly(3-hydroxybutyrate-*co*-3-hydroxyvalerate) (PHBV) and regenerated silk to fabricate piezoelectric sensor using an extrusion-based printing, as illustrated in Figure 18B. The results indicated that the infill density and grid pattern can help to the piezoelectric properties of the 3D-printed force sensor.

5.4. Wearable Smart Textiles and Fashion Industry

The technological developments in AM combined with stimuli-responsive materials open new dimensions to use their distinct properties in exploring functional textiles, which can transform shape upon external stimulations.[377] These materials, especially SRPs, can be blended in cotton, polywood, polypropylene, and polyester fibers to fabricate more comfortable clothing, thanks to the change in the molecular structure of SRPs for assisting air ventilation and better comfort.[548,549] Recently, "Diaplex", an active sports clothing, was manufactured by Mitsubishi Heavy Industries, using SRPs. It helps to regulate the body temperature under different weather conditions.[550] Some authors also printed smart textiles by using 3D printing technology. For instance, Zhang et al.[551] evaluated the shape-memory performance of 3D-printed circular braided preforms and their silicone elastomer matrix composites. The results revealed that the incorporation of silicone elastomers greatly enhanced the shape memory (Figure 18C), shape recovery, and compressive strength of functional composites. These functional composites can be applied in the textile sector to fabricate hybrid smart textiles.

5.5. Construction Sector

AM of stimuli-responsive materials can overcome the existing challenges in the design and fabrication of self-adaptive architectures.[552] For this, self-shaping and self-assembly features of smart composites can help in automating the movements in response to climate conditions.[553] Similarly, 3D printing of stimuli-responsive materials can help to develop climate-responsive architectural facades.[554] Recently, Yi et al.[555] printed reversible thermo-responsive architectural facades by combining programmed SMA and SMP matrix, as illustrated in **Figure 19**A. The 3D-printed structure showed excellent thermomechanical properties and shape-morphing behavior upon temperature stimulation. Thus, self-assembly, self-healing, and self-sensitive features induced by ambient conditions promote the use of this novel technology in the building sector. Furthermore, 3D printing of SRP-based composites can help in developing self-adaptive, eco-friendly, and energy-saving buildings.[556] Water demand can be adjusted by using SRP-based pipes, which change their diameters, when subject to environmental stimulation. The self-healing nature of SRPs can help to heal cracks in the piping system.[557]

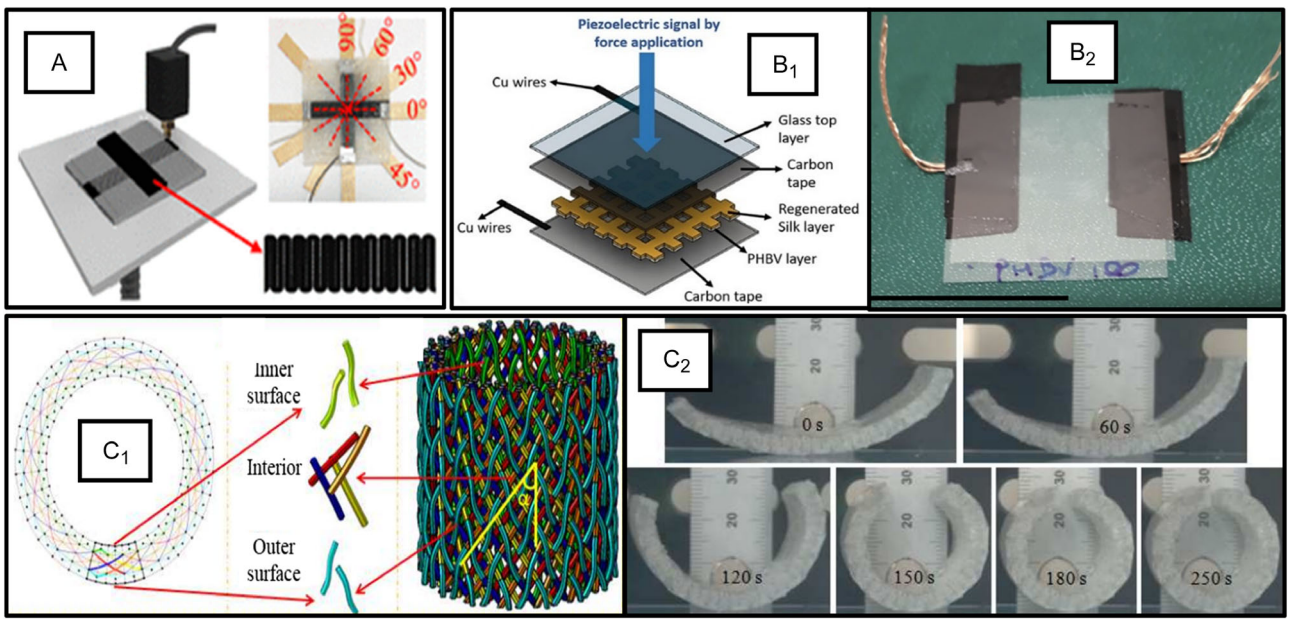

Figure 18. A) Printing of anisotropic, flexible, and constriction-resistive sensors. Adapted with permission.[546] Copyright 2020, American Chemical Society. B) 3D-printed sensors; B_1) schematic illustration of piezoelectric sensors; B_2) photograph piezoelectric device. Reproduced under the terms of the CC-BY license.[547] Copyright 2021, The Authors, Published by Elsevier. C_1) 3D circular braided tube model; C_2) shape-memory behavior of 4D-printed functional composites with shape recovery temperature of 70 °C. Adapted with permission.[551] Copyright 2018, Elsevier Ltd.

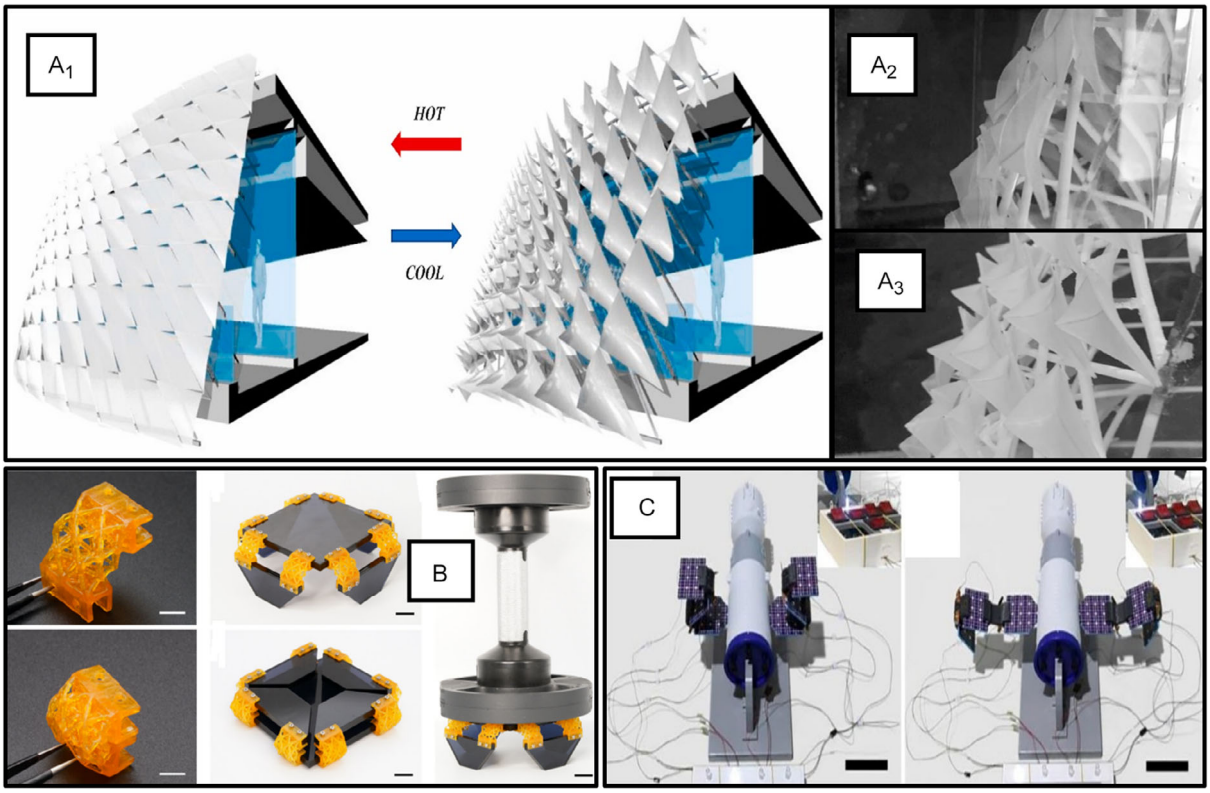

Figure 19. A) Building design concept; A_1) biomimetic self-shaping skin; 4D-printed building skin at A_2) high temperature and A_3) low temperature. Adapted with permission.[555] Copyright 2021, Elsevier Ltd. B) 3D-printed SMP-based hinges can be used to support load. Adapted with permission.[582] Copyright 2021, Wiley-VCH GmbH. C) 3D-printed self-deployable hinge connected to solar panel system. Adapted with permission.[583] Copyright 2022, Elsevier B.V.

5.6. Aerospace Sector

Stimuli-responsive materials are important in the aerospace industry, thanks to their self-folding, self-assembly, and adjustability in response to environmental conditions. Compared with SMAs, SMPs offer more deformation and less density.[558–560] SRP-based self-deployable structures are lightweight, self-expanding, and have excellent carrying capacity, which makes them highly suitable for aerospace applications.[561] 3D printing of SRPs is showing remarkable potential in aerospace applications.[337,562–565] Additionally, 3D printing of SMP composites are also used to fabricate hinges, solar panels, and self-deployable antennas,[562] as illustrated in Figure 19B,C. In summary, 3D printing of SRPs is a futuristic research arena, which is highly beneficial for many applications including biomedical, construction, textile, and aerospace fields. **Figure 20** provides landmark achievements of the 3D printing scientific community that use 3D printing techniques and SRPs to develop products for groundbreaking applications.

6. Current Challenges and Future Outlook

In recent years, the 3D printing technology has opened pathways for developing a variety of structures suitable for soft robotics, biomedical, smart actuators, sensors, 5G communication, and aerospace applications.[566–570] **Figure 21** shows the past, present, and the future of 3D-printed SRP composites. AM technology permits the formation of smart and complex structures by using the combination of 3D printing and SRPs which is accomplished by using extrusion-based printing, vat photopolymerization, and powder-based printing techniques. With the widespread applications of 3D-printed smart structures in the most challenging sectors,[571] there is a need to take concrete steps for addressing the challenges faced by this unparalleled manufacturing technology.

For technological challenges, AM technologies for printing SRPs are limited and are accessible only in research institutes around the world. FDM, DIW, SLA, and IJP are applied to print intricate structures.[572] However, filaments for FDM are limited, whereas SLA requires photocurable liquid resins, and DIW requires feedstock in a liquid form. Additionally, inks suitable for DIW must meet the requirements of tunable rheology and high swelling degree. Thus, only a limited number of SRPs can be printed by using these AM technologies.

3D printing of SRPs is still in its infancy in which printable materials are limited at laboratory scale. To date, no commercially available printer is specifically designed for the printing of stimuli-responsive materials. The contemporary printing precision and the performance of SRPs are not meeting the required standards. Therefore, this technology needs further

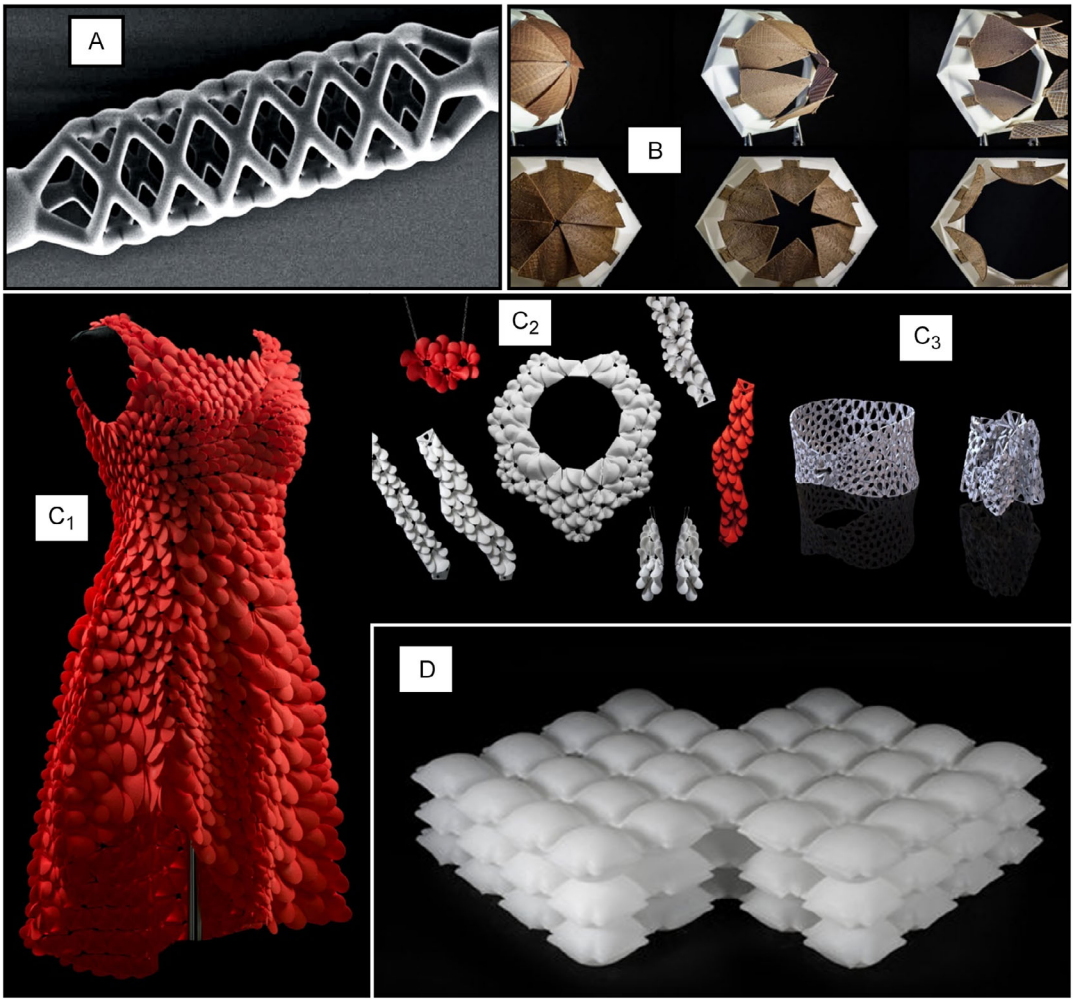

Figure 20. Some landmark complex structures developed by different research groups by using the combination of 3D printing and SRPs. A) Microstent of 0.05 mm wide and 0.5 mm long was developed by Bradley Nelson's research group at ETH, Zurich. Adapted with permission.[584] Copyright 2019, WILEY-VCH Verlag GmbH. B) Bioinspired structure responsive to environmental stimulation.[585] C_1) 3D-printed nylon petals dress for fashion industry;[586] C_2) 3D-printed foldable jewelry; C_3) foldable 3D-printed fabric belt.[586] D) BMW and MIT's Self-Assembly Laboratory successfully developed 3D-printed inflatable interiors.[587]

improvement for the development of highly responsive and precise structures. Additionally, insufficient driving force and slow response time are other issues associated with SRPs. Insufficient driving force arises due to the strong temperature dependence of the material modulus. Also, SRPs take several minutes to achieve the desirable response. In the future, this problem can be tackled through material modifications.

Numerous SRPs and their composites have shown great ability to change their function, shape, or color in response to external stimulation.[573] For biomedical applications, these materials must exhibit biocompatibility, noncytotoxicity, required mechanical strength, and appropriate dynamic response, which do not impair the tissues. However, these requirements are met by only a few dynamic materials. Furthermore, most SRPs are only responsive to a single stimulus.[574] Therefore, focused research is required to develop multi-responsive materials with precise and multidimensional responses. The biological environment is different, dynamic, and complex for each person. Thus, 3D-printed structures should be adaptable to the microenvironment of organisms. Furthermore, the development of novel SRP composites or advancements in the functionalization of the existing polymers can be used to fabricate multifunctional scaffolds and dynamic structures for biomedical and other engineering fields, respectively. Even today, 3D printing of SRPs is primarily focusing on the shape morphing ability, but other functionalities of smart structures in the future can help to develop multifunctional architectures.

Existing SRPs are limited to simple folding or bending responses only at the macroscale.[575–577] Therefore, there is a need to shift design control from the macro level to the micro level. Additionally, precise control of external stimulations, resolution, and response can help in controlling the deformation of the patient-specific structures; therefore, these materials can be a

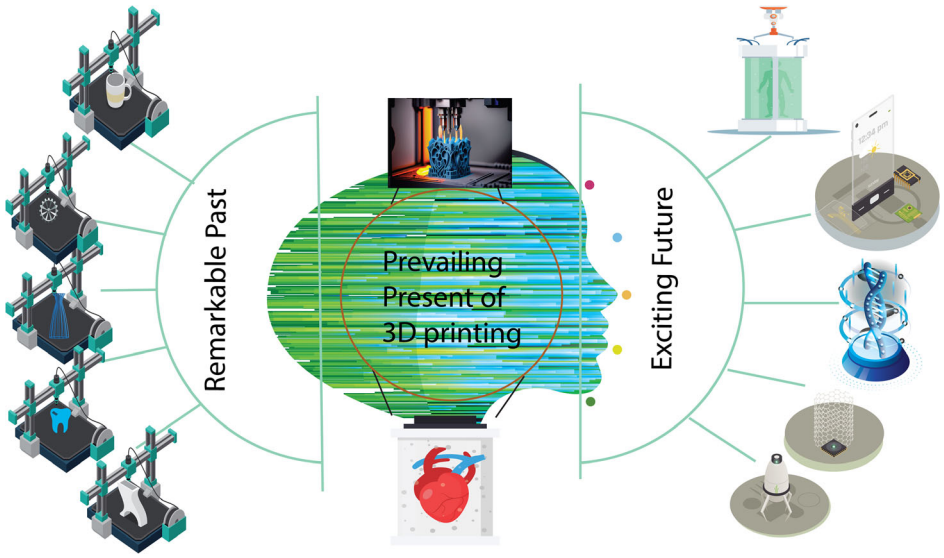

Figure 21. Schematic illustration of past, present, and future research directions of 3D-printed SRPs.

game-changer for the biomedical sector. Similarly, bilayer SMP composites are reported to be used for developing actuators and grippers, due to the asymmetric deformation of two layers.[578] However, the deformation is limited to simple bending and elongation. There is a need to develop novel bilayered SMP composites with complex deformation mechanisms, which will further expand their applications. **Figure 22** depicts the strengths, weaknesses, opportunities, and threats (SWOT) analysis of 3D-printed SRPs.

Figure 23 highlights the potential impact as well as future research areas of 4D printing technology in the biomedical sector. Nowadays, in vivo and in vitro, and 2D assays can be approached by using on-chip models and 3D assays, thanks to AM of SRPs.[579] In the future, the manufacturing of artificial biostructures loaded with therapeutic agents, cells, and growth factors, which offer the physiological characteristics of native tissues, may help in revolutionizing the tissue engineering and regenerative medicine field.

Computational approaches can prove vital to predict the shape-morphing behavior of SRPs.[580] These predictions are highly beneficial for linear rigid materials. With the contemporary data-driven approaches, it is highly challenging to verify the

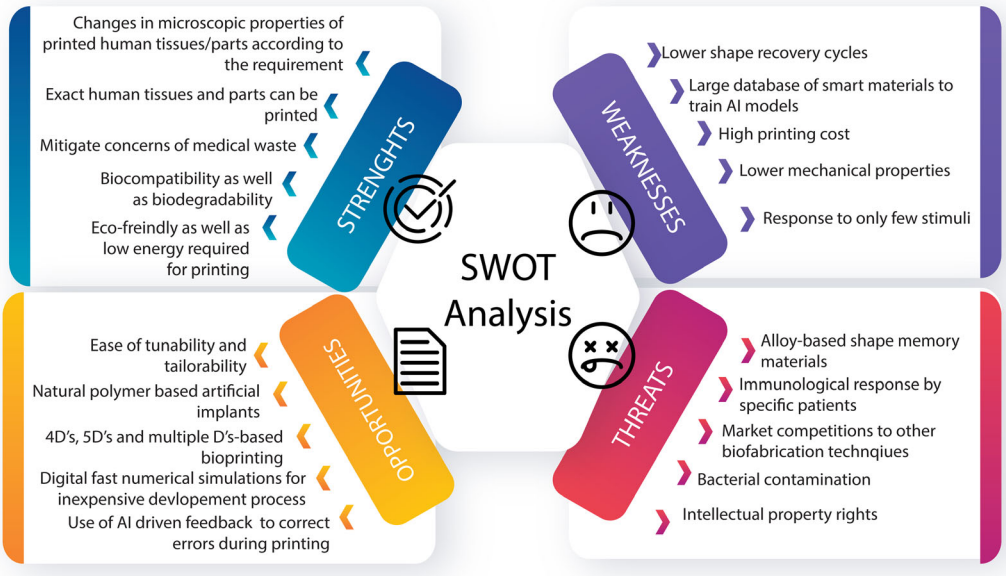

Figure 22. SWOT analysis of SRP-based structures developed through 3D printing.

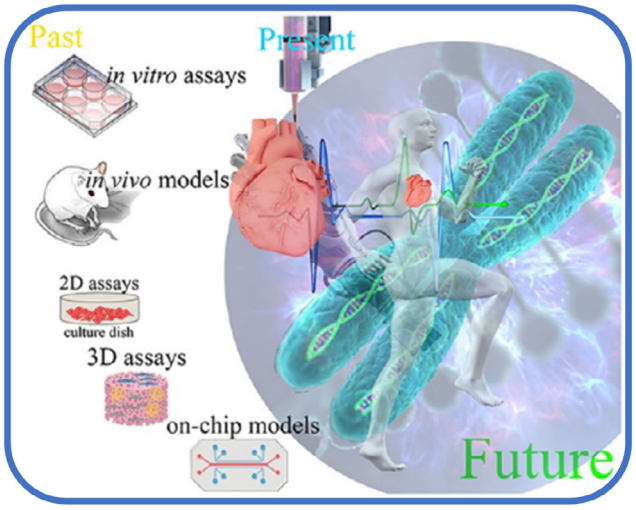

Figure 23. Schematic representation of impact and future research directions of 4D printing in biomedical sector. Reproduced with permission.[130] Copyright 2022, American Chemical Society.

behavior of soft SRPs, owing to their nonlinear locomotion characteristics. In the future, the accretion of experimental data and results of computational predictions can help in providing a significant database for artificial intelligence (AI) systems to develop novel SRP composites.

Despite all the challenges, the 3D printing of SRPs is a promising combination, which created new frontiers in a broad range of engineering fields, paving the way for fabricating customized, complex, dynamic, and precise products. These products were previously impossible to manufacture through traditional manufacturing methods and materials. However, the present research progress is the tip of the iceberg. The technology is evolving, improving, skyrocketing, and opening up new opportunities for researchers and scientists to expand this cutting-edge technology in the industries. In the future, people may be able to print multifunctional structures using the combination of personalized recommendations, smart polymers, and 3D printing technology, which will be translated into actual engineering applications.

Acknowledgements

A.T., Z.U.A., and M.Y.K. contributed equally to this work. M.H. acknowledges the support of the Royal Society through the International Exchange (grant no. IEC\NSFC\211316) with the National Natural Science Foundation of China (NSFC).

Conflict of Interest

The authors declare no conflict of interest.

Keywords

3D/4D printing, additive manufacturing, biomedical applications, smart polymers, stimuli-responsive polymers

Received: July 15, 2023
Revised: August 16, 2023
Published online:

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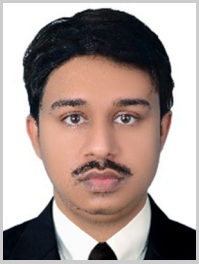

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