Recent Progress in 3D Printing of Smart Structures: Classification, Challenges, and Trends

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1. Introduction

Smart structures are composed of sensitive materials that dynamically respond to external stimuli and thus realize specific desired functions,[1] and have attracted considerable research attention in recent years. The most common smart structures enable controllable deformations under external stimuli (e.g., heat,[2,3] electromagnetism,[4,5] light,[6,7] pH,[8] ion concentration,[9] sound,[10] and mechanical force[11,12]). In addition, self-healing and sensing capabilities are now being explored. Techniques and tools that rely on the capabilities provided by smart structures have been applied to soft robotics,[13–16] aerospace engineering,[17] electronics,[18–20] and biomedicine.[21–23]

Material development, structural design, and manufacturing are three key aspects of promoting the availability and use of smart structures. Moreover, such research towards smart structures can directly be used to improve those three aspects. This is particularly compelling because the generally recognized bottleneck to smart-structure promulgation is the manufacturing methods from continuous research and experimentation; smart structures have become increasingly complex, outpacing traditional manufacturing methods. The molding and milling methods are unsuitable. The construction of internal vascular networks is a remarkable example of this problem. Such networks are built by embedding hollow fiber tubes into a 3D solid structure. In addition, the traditional manufacturing method has difficulties in the integrated processing of heterogeneous materials, hence, the complete structure needs to be divided into multiple components and processed separately. Therefore, the construction of such multicomponent smart structures usually requires manual assembly, which limits their complexity and miniaturization of structures.

In recent years, 3D printing (additive manufacturing) has emerged as a versatile manufacturing method and an alternative to traditional manufacturing methods. During the 3D printing process, objects are virtually sliced into multiple layers, blocks, or blobs using computer-assisted design software. Thereafter, the printing material is stacked layer by layer or piece by piece using a printer to obtain the target design. As shown in Figure 1, the 3D printing process can be figuratively regarded as the reverse process of slicing potatoes in terms of its surface-forming method. Layer-by-layer 3D printing is accomplished using digital light processing (DLP), projection microstereolithography (μSL), magnetic field-assisted projection stereolithography (M-PSL), and microscale continuous optical printing (μCOP). If the process is instead regarded as the reverse of shredding a potato, an extrusion-based printing process is described (e.g., direct ink writing (DIW) and fused deposition modeling (FDM)). If the reverse process of potato dicing is considered, an inkjet-based printing process is described (e.g., PolyJet and aerosol jet-based printing (AJP)). More details,
classifications, and comparisons of various 3D printing methods can be found in our previous review of 3D printing technology.\(^2\)

The introduction of 3D printing technology into the fabrication of smart structures has led to remarkable achievements. First, it provides an economical and effective method to fabricate complex smart structures, such as the 3D interpenetrating internal vascular networks mentioned earlier,\(^3\) vascular scaffolds,\(^4\) and curvature-controllable and deformable 3D lattice structures.\(^5\) In addition, the multimaterial compatibility of 3D printing technology allows the integrated manufacturing of functionally gradient, anisotropic, multilayer composite, and multicomponent structures. Furthermore, high-resolution 3D printing technologies, such as AJP,\(^6\) provide an effective method for fabricating micro–nanoscale microstructures.

This Review discusses the latest research progress in 3D printing for smart structures and comprehensively summarizes and analyzes the printable materials used in the manufacturing process of various functional smart structures. It differs from extant reviews that have examined materials and manufacturing methods for certain smart structure types. It pays more attention to diverse 3D printing methods for a variety of smart structures from a manufacturing perspective. Various types of smart structures are catalogued in detail based on materials, manufacturing methods, and applications. As shown in Figure 2, this Review focuses on functional polymers and composite materials used in smart structures, suitable 3D printing methods for smart structures, and their applications. Correspondingly, it answers three main questions: “What materials can be used?” “How can they be manufactured?” and “Where can they be applied?” Furthermore, current limitations and future trends are discussed and recommended, respectively. Thus, we provide a new knowledge base that offers a more comprehensive understanding of smart structures alongside inspiration for further developments.

2. Temperature-Responsive Smart Structures

A temperature-responsive smart structure responds to changes in temperature to achieve predetermined functions. Such structures
are usually built using multilayer structures with different degrees of thermal sensitivity at each layer,\textsuperscript{[14,27–32]} such as with old-fashioned thermostats. Different heat-sensitive layers can be realized with either different materials or the same material having different thicknesses. Temperature-responsive smart structures can also be realized by embedding shape memory (SM) materials into softer base materials.\textsuperscript{[13–38]} Uneven or shaped heating patterns can also be used to achieve variable changes.\textsuperscript{[39,40]} The application of prestresses during or after manufacturing has also been used to create temperature-responsive results.\textsuperscript{[41–46]} These smart structures perform important biomimetic tasks\textsuperscript{[28,47]} and support soft robotics.\textsuperscript{[15,34,48]} Intelligent components,\textsuperscript{[26,49]} and nonbinary actuators.\textsuperscript{[36]} To satisfy the requirements of rapid and high-quality manufacturing of multimaterial complex structures, a series of 3D printing methods has been introduced.

DIW is commonly used for manufacturing temperature-responsive smart structures. Poly-N-isopropylacrylamide (PNIPAAm) is the most typical temperature-responsive polymer used in the DIW method. Bakarich-et al.\textsuperscript{[49]} developed a new type of alginate/PNIPAAm ICE gel ink and fabricated a smart valve with a controllable flow rate by printing the temperature-responsive gel ink with other static materials. This valve can reversibly vary the switch state using temperature changes. However, the conventional PNIPAAm hydrogel exhibits a relatively low response speed. To solve this problem, Ko et al.\textsuperscript{[28]} developed a novel type of hydrogel ink consisting of monomer acrylamide, NIPAAm, and sugar (glucose + sucrose). Then, they designed rapid temperature-responsive biomimetic architectures with sugar-induced macropores (Figure 3A). The reactive DIW printing of a liquid-crystal elastomer was proposed to fabricate various preprogrammed SM structures.\textsuperscript{[42,50–52]}

With the wide application of 3D-printed smart structures, the requirements for material properties have become increasingly complex. Inks containing urethane diacrylate and linear semicrystalline polymers have been developed for 3D printing an SM and self-healing polymer network elastomer that can be stretched up to 600%.\textsuperscript{[13]} Zhang et al.\textsuperscript{[34]} embedded an SM polymer (SMP) layer into a fully printed soft actuator body that enabled a stiffness-tunable function without compromising

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**Figure 3.** Temperature-responsive smart structure fabricated by DIW method: A) DIW printing of bellflower-inspired temperature-responsive smart structure. Reproduced with permission.\textsuperscript{[18]} Copyright 2020, Springer Nature; B) DIW printing of robotic gripper. Reproduced with permission.\textsuperscript{[34]} Copyright 2019, Wiley; C) DIW printing of simple linear contractile elements and multimodular 3D structure. Reproduced with permission.\textsuperscript{[33]} Copyright 2018, Wiley; and D) DIW printing of multimaterial lattice structure. Reproduced with permission.\textsuperscript{[27]} Copyright 2019, National Academy of Sciences.
flexibility or adaptivity. They fabricated a robotic gripper with three actuators that could grasp various objects having arbitrary shapes and different weights (i.e., 10 g to 1.5 kg) (Figure 3B). To achieve more complex functions, simple deformations (e.g., stretching and twisting) are no longer sufficient. Therefore, a modular idea was recently proposed. Multiple functional parts having simple linear contractile ability were assembled into multimodular structures to realize complex motions (Figure 3C). William et al. proposed a versatile platform based on a multimaterial lattice 3D-printing method to fabricate complex shape-morphing architectures (Figure 3D). Notably, the combination of temperature response with other functions (e.g., self-healing and electrical conductivity) has become increasingly popular.

The FDM method has also been widely used to fabricate temperature-responsive smart structures. Polylactic acid (PLA), a common SM material, is widely used because of its superior temperature sensitivity, mechanical properties, and recyclability. An underlying mechanism of stored internal strain can be applied during printing. This strain is then used to induce the controllable pattern transformation of the heat-sensitive polymer (Figure 4A). Zhang et al. fabricated temperature-responsive 3D lightweight self-folding sheets and leveraged the effect of internal strain in printed structures to control deformations (Figure 4B). In addition to the thermal self-folding behavior of thin sheet structures, thermally activated origami-inspired self-deploying thick plate structures having large-area change ratios have also been developed. Sun et al. found that the glass transition temperature ($T_g$) of PLA could be influenced by its plasticizer content and demonstrated that plasticized functionally graded layers could be used to fabricate localized SM structures (Figure 4C). The internal strain theory of PLA has also been applied to auxetic metamaterials to continuously tailor their in-plane moduli and Poisson’s ratios (Figure 4D). In addition to the aforementioned materials, Yang et al. fabricated an SM flower and gripper using the MM-4520 thermoplastic polyurethane elastomer. They also studied its optimal FDM printing parameters. Chen et al. presented a novel multimetal electrochemical FDM method to fabricate a copper–nickel bimetallic strip.

Inkjet printing is commonly used because of its multimaterial compatibility. A hydrogel ink with excellent printability and shape stability was prepared by the in situ polymerization of acrylamide in an agarose matrix containing laponite. A whale-like...
hydrogel and octopus-like gel that can transform their shapes with temperature changes were printed using this ink (Figure 5A).[46] Liu et al.[39] developed a novel and simple approach for the self-folding of thin sheets using unfocused light. They patterned black ink that was used in the form of hinges on a temperature-responsive polymer. The hinges absorb more light and attain higher temperatures. Therefore, the underlying polymer reaches the glass transition temperature earlier and deforms to achieve the self-folding ability (Figure 5B).

Moreover, self-folding structures can be fabricated by the programmed strain mismatch between two layers. The emergence of the PolyJet printing method, which applies materials having multiple colors or other variable properties to a single process, has considerably improved manufacturing efficiency. Remote actuation with uniform light illumination was realized by color-dependent selective light absorption in multicolor SMP composites.[40] Wu et al.[14] printed layered composite structures using multiple SMP fibers having different \( T_g \) values. They developed a theoretical model to better understand the deformation behavior of SMP composites and designed an insect-like structure and a smart hook guided by the model (Figure 5C).

Teoh et al.[47] and Ding et al.[58] combined VeroWhitePlus and TangoBlackPlus with different ratios to obtain materials having different \( T_g \) values. They fabricated an artificial flower capable of concurrent sequential shape changes under dry conditions. An untethered swimming soft robot driven by preprogrammed bistable SMP muscles was also developed (Figure 5D).[36]

Vat photopolymerization is a 3D-printing process that uses specific light (e.g., ultraviolet (UV)) to selectively cure liquid photopolymers in a vat using a layer-by-layer light-activated approach to form the target structure. The first vat photopolymerization method was stereolithography (SLA), developed in the early
The SLA printing of methacrylated semicrystalline molten macromonomers enabled the rapid fabrication of complex SMP structures, and printing using conductive ink on the SMP structure imparted the SM function to electrical circuits (Figure 6A). A novel ink containing renewable soybean oil epoxidized acrylate was recently developed, exhibiting broad potential in the field of biomedicine because of its high biocompatibility and human body temperature (37°C) response characteristics. Mishra et al. used a multimaterial SLA technology to create a soft hydrogel actuator with an autonomic perspiration function, which exhibited a 600% increase in cooling speed compared with nonsweating devices. Based on SLA, DLP further improved the build speed of vat photopolymerization printing process. Reversible shape-changing components having adjustable stiffnesses were fabricated by printing Tangoblack SMP material on hydrogels (Figure 6B). Wang et al. used photopolymerization 3D printing to create a horse-shoe-shaped temperature-responsive lattice structure with a controllable shape-changing ability. Kuang et al. developed a single-vat grayscale DLP (g-DLP) 3D printing method using different grayscale light intensities to cure the material and obtained a tunable Tg ranging from 14 to 68°C. Thereafter, they fabricated various sequential SMP components (Figure 6C). To satisfy the printing resolution requirements, the PμSL method was developed. Han et al. then used PμSL to fabricate PNIPAAm microstructures that exhibited sequential temperature-responsive deformations. High-resolution multimaterial SM structures, such as grippers and self-blooming flowers, were fabricated using PμSL (Figure 6D).

From Table 1, it can be seen that DIW and FDM are the widely used methods. There is a growing interest in fabricating temperature-responsive smart structures via inkjet printing and DLP methods. The application of PμSL enables the manufacture of temperature-responsive smart structures having high-resolution microstructures. Temperature-responsive smart materials mainly include hydrogels and SMPs, of which PNIPAAm is the most widely used smart gel. Limited response speed, actuating force, structure stability, and mechanical properties are the
3. Electromagnetic-Responsive Smart Structures

Electroresponsive smart structures are usually driven by electroactive polymers, such as dielectric elastomer actuators (DEAs) and electroactive hydrogels (EAHs), because of their considerable strain capacity and energy density. Electroresponsive smart structures usually combine electrostrictive materials and non-electroactive materials by layering or winding to achieve basic actions, such as bending and twisting under electrical stimulation. Complex actions can be modeled with the combination of multiple basic actions. Various 3D printing methods have been used to fabricate electroresponsive smart structures, and DIW is the most common method. Kamamichi et al. used this method to achieve the automatic fabrication of a Bucky-gel actuator, which is a low-voltage-driven soft electroactive actuator. They fabricated a reverse connecting film to demonstrate the feasibility of printing complex smart structures. Mestre et al. used the DIW method to print muscle-cell-containing bioink and created a highly sensitive electroresponsive bioactuator. Cai et al. combined FDM and DIW to realize the full printing of a soft robotic face whose pupils and lips could be actuated electrically to obtain a smiling expression. Wei et al. developed a novel type of ink having excellent printability and high electrical conductivity by combining silver-coated carbon nanofibers with a thermoplastic polymer. The developed ink achieved self-supporting printing at room temperature. They also fabricated an electroactive smart gripper to demonstrate the electrical SM property of the developed ink (Figure 7A). A low-cost and facile method using electroless deposition–assisted DIW to directly integrate high-resolution 2D and 3D copper microcircuits on insulating substrates was also proposed. Ambulo et al. prepared a new ink by dispersing droplets of eutectic gallium indium alloy into a liquid-crystal elastomer matrix. They used the DIW method to create a soft actuator having both electrothermal and photothermal response capabilities (Figure 7B). By patterning high-fidelity interpenetrating vertical electrodes, 3D DEA devices using in-plane contractile actuation were created (Figure 7C).
of high-resolution electroresponsive smart structures, including grippers, transporters, and walkable human-like structures, was achieved (Figure 7D). Takuya et al. created a caterpillar-like soft robot using the PolyJet printing method, and achieved the omnidirectional movement of the robot through embedding coiled electrical SM alloys.

3.2. Magnetoresponsive Smart Structures

Magnetic-response smart structures are widely applied because of their fast response and noncontact control characteristics. The magnetoresponsive function is usually achieved by doping ferromagnetic particles into a polymer matrix, such as polydimethylsiloxane (PDMS), UV resin, and hydrogel. To achieve complex deformations in a magnetic field, each part of the magnetoresponsive smart structure can be magnetized in different directions, and the concentration of magnetic particles can be adjusted to change the magnetic force. One of the most commonly used methods for printing magnetoresponsive smart structures is the DIW technique. Zhu et al. doped soft magnetic iron particles having an average particle size of 125 nm into PDMS to obtain a PDMS–iron composite ink and fabricated a fast-response 3D butterfly and a soft gripper using the DIW method (Figure 8A). Roh et al. used PDMS as the matrix and 4 μm carbonyl iron particles as filling particles to obtain a type of magnetic ink. Inspired by creatures living on a water
surface, a magnetically controlled soft gripper and a floating water-droplet dispenser were fabricated (Figure 8B). Chen et al. \[85\] mixed hydrogel, a carbomer as a rheology modifier, and Fe$_3$O$_4$ particles to obtain a magnetic ink with excellent printability. Thereafter, they used the DIW method to print a soft octopus robot that could move with changes in the magnetic field. Lee et al. \[86\] used iron oxide as doped particles to create a magnetically driven two-way size-controllable microroller (Figure 8C).

In addition to soft magnetic materials, hard magnetic materials are widely used in magnetoresponsive smart structures. To improve the manufacturing efficiency of magnetoresponsive smart structures and the printing ability of complex structures, a method for applying a magnetic field during printing to redirect magnetic particles has been proposed. Kim et al. \[5\] embedded hard magnetic NdFeB particles into a silicone rubber matrix to obtain a composite ink. A 2.7 T pulsed magnetic field was applied during the DIW printing process to allow the extruded ink filament to attain a specific magnetic polar direction. Rapid and complex deformations of 2D and 3D structures in a specific magnetic field were thus achieved. They also fabricated a reconfigurable soft electronic device and hexapod robot for a drug-delivery application. Using the same principle, Xu et al. \[77\] printed various magnetoresponsive smart structures, including self-assembly structures, a speed-tunable magnetic swimmer, an untethered multiarm microgripper, and a multilegged paddle-crawling robot. Based on their method, a submillimeter-scale soft wire-like robot with hydrogel skin was presented. The robot could pass through complex and narrow pipes similar to blood vessels under the action of a magnetic field. Therefore, it has considerable potential in the medical field. \[78\] Mea et al. \[74\] developed a novel DIW method that used a glass capillary microfluidic device to control the dispersion of eutectic gallium indium in PDMS to fabricate a magnetically actuated soft gripper. Using two hard magnetic smart inks, Ma et al. \[87\] proposed a magnetic multimaterial DIW technology for the manufacture of metamaterials with tunable Poisson’s ratio and a series of pop-up designs with multimodal deformation. Wu et al. \[88\] presented a voxel-encoding DIW printing method to program both the magnetic

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**Figure 8.** Various magnetoresponsive smart structures fabricated by 3D printing technology: A) DIW printing of a magnetic 3D butterfly and a soft gripper. Reproduced with permission. \[69\] Copyright 2018, American Chemical Society; B) DIW printing process and various specific deformations of magnetoresponsive lattice structures. Reproduced with permission. \[76\] Copyright 2019, Wiley; C) magnetically controlled motion of microrollers with controllable size. Reproduced with permission. \[86\] Copyright 2020, American Chemical Society; and D) schematic diagrams and grasping processes of two magnetic-controlled grippers. Reproduced with permission. \[75\] Copyright 2017, Wiley.
density and direction distributions during printing. This provided a feasible method to realize the controllable complex deformation of magnetoresponsive smart structures.

Vat photopolymerization is also widely used for printing magnetoresponsive smart structures. Kim et al.\(^{[93]}\) developed a type of magnetic ink that comprised superparamagnetic colloidal nanocrystal clusters and a photocurable resin monomer solution. They fabricated basic magnetic units having different magnetic axes using the DLP method and assembled them to achieve various types of motion forms, including snake-like movement and caterpillar-like crawling. Ji et al.\(^{[75]}\) prepared a magnetic ink by incorporating \(\text{Fe}_3\text{O}_4\) nanoparticles into photocuring resin. They used the DLP method to print the magnetic ink alongside the nonmagnetic resin to fabricate a magnetic-driven gripper with high sensitivity (Figure 8D). Credi et al.\(^{[90]}\) proposed two methods for manufacturing high-sensitivity magnetically responsive cantilever beams via 3D printing and verified their feasibility as magnetic sensors. Zhu et al.\(^{[70]}\) prepared a magnetic ink composed of poly(ethylene glycol) diacrylate-based hydrogels and \(\text{Fe}_3\text{O}_4\) nanoparticles. They used the microscale continuous optical printing method to print a magnetic-driven microfish whose swimming direction and speed were easily controllable. A 3D-printing method using sacrificial material (SMI) was applied to process a highly controllable and flexible starfish robot driven by five tensesguy structures. Giltinan et al.\(^{[93]}\) provided a biocompatible ink by doping iron platinum nanoparticles into a fish robot. They fabricated basic magnetic units having different magnetic axes using the DLP method and assembled them to achieve various motion forms, including snake-like movement and caterpillar-like crawling. Loebel et al.\(^{[97]}\) provided a feasible method to realize the controllable complex magnetic behavior of self-healing materials in the online printing and printing process. Meanwhile, strategies for electromagnetic control and miniaturization require further study, although the motion control of micro-robots in vascular-like pipes has been realized.\(^{[78]}\)

### 4. Self-Healing Smart Structures

Self-healing smart structures can repair material damage automatically or under the influence of external stimuli, such as light or heat. Thus, they afford a simple and low-cost approach for structural lifetime extension.\(^{[92]}\) Healing efficiency and number of successful consecutive healing cycles are the two main parameters characterizing the self-repairing performance of such structures. The modeling methods of self-repairing smart structures mainly include the use of self-repairing materials containing reversible bonds and the internal insertion of hollow vascular networks or capsules containing repairing compositions. Moreover, the introduction of 3D printing technology makes possible the manufacture of complicated self-healing structures, such as complex shapes of self-healing materials\(^{[11,93–98]}\) and complex internal vascular networks.\(^{[12,99–104]}\)

The 3D printing of self-healing materials with reversible bonds is a common method of manufacturing self-healing smart structures. Loebel et al.\(^{[97]}\) prepared a printable hyaluronic acid (HA) hydrogel ink with a rapid self-healing ability by eliminating the shear force. They used methacrylate as the supporting material and printed an HA hydrogel ink using the DIW method. Unfortunately, the photodegradability of methacrylate limits its application as a support material for 3D printing. Hu et al.\(^{[95]}\) developed a recyclable UV-resistant self-healing silicone oil-in-water emulsion glass material that provided a stable

| Category   | Printing method | Materials                                                                 | Application\(^{[9]}\)            |
|------------|-----------------|---------------------------------------------------------------------------|---------------------------------|
| Electricity| DIW             | Silicone elastomers                                                      | Soft robotic face                |
|            |                 | Bucky gel                                                                |                                 |
|            |                 | Ag catalyst ink                                                          |                                 |
|            |                 | Ag@CNF PLA nanocomposite                                                 | Multifunction and soft gripper   |
|            |                 | Carbon black particles and poly(ethylene glycol ethylene sulfide) oligomer|                                 |
| Magnetism  | DIW             | PDMS/Fe particles                                                        | Bionic robotics                 |
|            |                 | PDMS/hydrox iron particles                                               |                                 |
|            |                 | Hydrogel/ NdFeB particles                                                | Soft robotics                   |
|            |                 | AAm-carbomer hydrogel/\(\text{Fe}_3\text{O}_4\) particles               | Soft robotics                   |
|            |                 | PDMS/ NdFeB particles                                                    | Medical robotics                |
| DLP        |                 | PDMS/ferrofluid                                                          | Soft gripper                    |
|            |                 | Acrylic resin/\(\text{Fe}_3\text{O}_4\) NPs                             |                                 |
|            |                 | UV resin/NdFeB particles                                                 | Soft microbot                    |
|            |                 | Nanocomposite composed of reduced graphene oxide                         | Biomedicine                     |
|            |                 | nanosheets and nanocookies                                               |                                 |
|            |                 | Poly(ethylene glycol) diacrylate                                         | Bionic robotics                 |
|            |                 | (PEGDA)/chiral nematic                                                   |                                 |
| SLA        |                 | Bifunctional acrylic photosensitive resin                                |                                 |
| M-PSL      |                 | Spot E elastic/EMG 1200 dry magnetic NPs                                 | Bionic robotics                 |
| µCOP       |                 | PEGDA/ Pt NPs and \(\text{Fe}_3\text{O}_4\) NPs                         | Bionic robotics                 |

\(^{[9]}\) N: Not given.
supporting medium for the DIW method. Liu et al.\[94\] prepared a novel thermal self-healing κ-carrageenan/poly(acrylamide) (PAAm) double-network (DN) hydrogel with excellent printability, electrical conductivity, mechanical properties, and recyclability. They also printed several basic shapes using the DIW method and demonstrated the application of the DN hydrogel in flexible sensing (Figure 9A). Nadgorny et al.\[95\] developed a printable self-healing gel by cross-linking benzaldehyde-functionalized poly(2-hydroxyethyl methacrylate) with ethylenediamine to form dynamic imine bonds. They used the DIW method to print a complex butterfly pattern and a star-like structure having 98% healing efficiency. They also discovered the self-curling behavior of hydrogel films in a dry environment (Figure 9B).

Vat photopolymerization is also used to print self-healing materials. Li et al.\[11\] developed a photopolymer resin based on polyurethane acrylate containing disulfide bonds. They applied the DLP method to print high-precision complex structures, which can achieve multiple high-efficiency self-healing processes (Figure 9C). Yu et al.\[98\] prepared a molecularly designed photoelastomer ink with both thiol and disulfide groups and printed a complex structure using the PbμSL method that achieved 100% thermal self-healing. They also fabricated a self-healing soft actuator and architected electronics to demonstrate the applications of the proposed method (Figure 9D).

Inspired by biological vascular networks, research on self-healing vascular-network smart structures has become popular. The emergence of 3D printing provides a simple and effective approach to fabricating such structures. The 3D printing of various self-healing smart structures based on the reversible bond of the materials is illustrated in Figure 9. Figure 9A shows DIW printing of basic shapes and the self-healing function and tensile performance of printing structures. Reproduced with permission.\[94\] Copyright 2017, American Chemical Society. Figure 9B shows DIW printing of self-healing objects and an object with both self-healing and self-curling functions. Reproduced with permission.\[95\] Copyright 2017, Royal Society of Chemistry. Figure 9C shows DLP printing of high-precision complex self-healing structures. Reproduced with permission.\[11\] Copyright 2019, American Chemical Society. Figure 9D shows PbμSL printing of complex structures, a self-healing soft actuator, and self-healing architected electronics. Reproduced with permission.\[98\] Copyright 2019, Springer Nature.
approach for the construction of complex internal vascular networks. The method based on SML is the most common method. First proposed by Scott R. White in 2003,[105] the SML process can be divided into three steps: the manufacture of sacrificial molds via 3D printing; embedding the sacrificial molds into a matrix to form a complete part; and the removal of the sacrificial mold to obtain hollow vascular networks. White pioneered a microvasculature design based on interdigitated dual networks, which allowed the healing agent to flow into cracks automatically without external pressure. He studied the self-healing ability of a smart structure with a brittle coating. Catalyst in the brittle coating reacted with healing agent transported through microvascular network to achieve self-healing function. He also investigated the effect of catalyst concentration on healing efficiency and proposed a criterion for self-healing performance testing based on a four-point bending experiment (Figure 10A). Postiglione et al.[99] used water-soluble poly(vinyl alcohol) as the SML to obtain a microvascular network based on the SML method. The fabricated structure achieved an 82% healing efficiency using PDMS as the healing agent. They also investigated the effect of the vascular network structure and density on the healing performance (Figure 10B).

In contrast to the SML method, Luan et al.[12] proposed a method to directly construct a dual-channel vascular network using FDM method (Figure 10C). Li et al.[104] further optimized the design according to Murray’s law and directly printed 3D bionic vascular networks using the FDM method. They embedded the printed vascular network in cement, providing not only self-healing properties but also improved mechanical features (Figure 10D). Table 3 summarizes these 3D printing methods, materials, healing agents, and healing efficiencies (η) discussed.

As summarized in Table 3, the DIW is again the most commonly used method for the manufacture of self-healing smart structures. The healing efficiency of self-repairing smart structures based on reversible chemical bonds is practically 100%, whereas that of most self-repairing smart structures based on vascular networks is relatively low. There are several problems that require further study. First, the self-repair time of existing self-repairing smart structures is generally long, and many of the healing processes require external stimuli as triggers. It is particularly important to find suitable repair agents that can achieve autonomous and rapid healing for different materials. In addition, exploring self-healing capabilities under low-work-intensity conditions is crucial to the improvement of production efficiency. Furthermore, the structure of vascular networks considerably influences their self-healing performance. Therefore, it is extremely important to explore effective design methods for more scientific structures. In this regard, a bionic vascular network design method based on Murray’s law has been proposed.[104] Finally, the current research focuses on investigating the self-healing performance of mechanical properties. The restoration of other properties, such as electrical ones, requires further study.[99]

![Figure 10. 3D printing of various self-healing smart structures based on vascular networks: A) the schematic diagram of a self-healing smart structure based on interpenetrating dual microvascular networks. Reproduced with permission.[100] Copyright 2009, Wiley; B) FDM printing of sacrificial mold and self-healing of the smart structures. Reproduced with permission.[99] Copyright 2017, American Chemical Society; C) FDM printing of three complex dual-channel vascular networks. Reproduced with permission.[12] Copyright 2020, Elsevier; and D) FDM printing of the designed vascular networks based on Murray’s law and their self-healing performance. Reproduced with permission.[104] Copyright 2020, Elsevier.](image-url)
Table 3. Summary of self-healing smart structure.

| Category            | Printing method | Materials                                      | Healing agent                  | η [%] |
|---------------------|-----------------|------------------------------------------------|---------------------------------|-------|
| Reversible          | DIW             | Silicone oil-in-water emulsion glass material   | Material itself                | N[93] |
| chemical bond       |                 | κ-carrageenan/PAAm DN hydrogel                  |                                 | 91[94]|
|                     |                 | Benzaldehyde-functionalized PHEMA with imine cross-links | GH hydrogel                    | 98[95]|
|                     | DLP             | Polyurethane acrylate containing disulfide bonds|                                 | 100[96]|
|                     | FDM             | Molecularly designed photoelastomer ink with thiol and disulfide groups | PLA + Diels–Alder polymer | 77[97]|
| Vascular network    | DIW             | Paraffin-based organic ink                     | DCPD and Grubbs’ catalyst particles | 70[98]|
|                     | FDM             | PVA                                             | PDMS                           | 100[99]|
|                     |                 | PLA.                                            | 2120 Epoxy Cure agent and 2000 Epoxy Resin agent | 51[100]|
|                     |                 |                                                | Sodium silicate                | 62[101]|

* N: Not given.

5. Smart-Sensing Structures

A smart-sensing structure can perceive changes in external physical parameters, such as displacement,[18,106–109] pressure,[110] and humidity.[20] The sensing function is usually achieved by embedding electronic components or piezoelectric conductive materials in an insulating matrix. Such structures exhibit a wide range of application prospects, such as wearable electronic sensing equipment, intelligent tactile monitoring of soft robots, and real-time monitoring of critical components. Among these, research on wearable flexible sensing devices has become quite popular. Leigh et al.[111] proposed a simple and low-cost method for the 3D printing of smart-sensing structures. They printed piezoresistive sensors and embedded them into a glove, providing a wearable flexible sensor to monitor finger movement. They also printed capacitive sensors and embedded them on the sides of a container to monitor the presence of liquids. Zhou et al.[112] used the DIW method to print a 3D-printed origami structure based on a barium titanate matrix and a silver flake–based electrode, demonstrating its application prospects as a self-powered gait sensor. Muth et al.[109] used an embedded 3D-printing method to achieve high-fidelity arbitrary path printing of conductive carbon grease in an elastomer support matrix. They embedded their sensors into a glove to monitor finger movement. Zhou et al.[113] added nanosilica to a silicone elastomer to change the latter’s rheological properties and used the DIW method to print complex shapes having excellent extensibility. This further demonstrated the manufacturability of wearable flexible strain-sensing products (Figure 11 A). Wang et al.[20] proposed a novel inflight fiber printing (iFP) method to fabricate fiber arrays on a circuit. They developed a wearable breathing sensor based on the iFP method and proposed the concept of a 3D floating electronic architecture. Peng et al.[107] prepared a resin containing polytetrahydrofuran units having excellent tensile strength and fatigue resistance. Then, they coated a conductive ion hydrogel onto the prepared resin to create a durable and high-strength wearable strain sensor. The AJP method can achieve nanoscale ultrahigh-precision printing. Young et al.[19] fabricated all-printed wireless nanomembrane hybrid electronics (p-NHE) using the AJP method. Wearable flexible sensors based on p-NHE exhibited excellent skin adhesion and high measurement accuracy (Figure 11 B). Based on the same method, Zhang et al.[22] created high-resolution 3D nanomembranes on an EV-CLUE chip to fabricate a microfluidic biosensor for cancer detection. Alongside wearable flexible sensing devices, many studies on other high-sensitivity sensors have been performed especially for robotic tactile sensing systems. Bodhe et al.[113] mixed polyvinylidene fluoride with barium titanate nanoparticles to obtain an ink having a self-supporting ability with excellent printing performance. Using the solvent evaporation-assisted DIW method, they fabricated a highly sensitive piezoelectric sensor that can be used directly without postprocessing (Figure 11 C). Ntagios et al.[110] investigated the sensing properties of five materials. They used the FDM method to directly integrate soft capacitive touch sensors on the fingertips of a robot, providing a cost-effective approach to fabricate tactile sensing systems. Based on the FDM method, a multidirectional, anisotropic, high-sensitivity strain sensor capable of obtaining quantitative measurements was fabricated using carbon nanotube–reinforced PLA. Thereafter, it was embedded into a soft robot to demonstrate its wide application potential.[108] Zhang et al.[116] obtained piezoelectric composites having excellent properties by incorporating boron nitride nanotubes into a photocurable polymer solution. They found that using the PdSL method of constructing microstructures on thin films improved perceptual sensitivity. They also fabricated high-sensitivity conformal piezoelectric sensors for the smart tactile-strain monitoring of robotic
Boley et al. proposed a DIW method for gallium–indium alloys and fabricated a stretchable strain sensor using this method.

The failure of key system components may lead to the stagnation of the production line or major safety problems. Therefore, the real-time monitoring of key components is advantageous for understanding working conditions in advance and avoiding safety hazards. Yang et al. combined the FDM method with laser-scribing technology and generated a laser-induced thin graphene layer on a polyether ether ketone substrate to obtain a highly sensitive self-monitoring smart structure (Figure 11D). Luan et al. built a self-monitoring smart structure based on the dual-nozzle FDM method by embedding continuous carbon fiber bundles into a PLA matrix. The carbon fibers enhanced the mechanical properties of the structure, and the deformation and failure of the structure could be detected via real-time monitoring of changes in the carbon-fiber resistance.

Based on Table 4, DIW is again the most commonly used method for manufacturing smart-sensing structures. The application of 3D printing technology provides a simple and effective

![Figure 11. 3D printing of various smart-sensing structures: A) DIW printing of wearable flexible strain-sensing products to perceive finger movement. Reproduced with permission. Copyright 2019, American Chemical Society; B) AJP printing of all-printed wireless nanomembrane hybrid electronics. Reproduced with permission. Copyright 2020, Springer Nature; C) DIW printing of a high-sensitivity piezoelectric sensor. Reproduced with permission. Copyright 2017, American Chemical Society; and D) FDM printing of a smart gear with self-monitoring capability. Reproduced with permission. Copyright 2020, American Chemical Society.]

| Printing method | Materials | Application |
|-----------------|-----------|-------------|
| FDM | Carbmorph | Wearable flexible sensor and smart container |
| | PLA–CNT | Embeddable sensor |
| | Silicone rubber + silver graphite conductive PLA | Robot tactile sensing system |
| | PLA + carbon fiber | Self-monitoring structure |
| | Polyether ether ketone | Self-monitoring structure |
| | Polyvinylidene fluoride–based ink | Wearable flexible sensor |
| | Silicone rubbers | Piezoelectric sensor |
| | Silica nanoparticles | Microfluidic biosensor |
| | PI, Ag, and graphene ink | Wearable flexible sensor |
| DLP | PPTMGA-40 | Wearable flexible sensor |
| PmSL | Photocurable polymer doped with boron–nitride nanotubes | Robot tactile sensing system |
method for the fabrication of wearable flexible sensors and robot tactile sensing systems. Currently, most smart-sensing structures can only perform qualitative analysis and have difficulty achieving high-precision quantitative measurement. Therefore, further research is necessary.

6. Other Smart Structures

3D printing technology has been applied to other smart structures that can respond to stimuli, including pH level, light, acoustic waves, and ion concentrations. A reversible pH-responsive flow-control valve was fabricated via extrusion printing using recyclable poly(2-vinylpyridine) filaments. Garcia et al. introduced an acrylic-acid monofunctional monomer into polyethylene glycol methacrylate to obtain a hydrogel ink and used the SLA method to manufacture high-precision antibacterial smart structures that can repeatedly swell and shrink with changes in pH. Light-responsive smart structures also have high research value because they can be remotely controlled, and they have rapid-response characteristics. Presently, the light-response function is generally achieved through the use of azobenzene groups. Daniel et al. used the DIW method to print photosensitive poly(siloxane)-containing pendant azobenzene groups onto a polyimide film to obtain a remote-controlled light-responsive double-layer actuator. Roppolo et al. proposed that the photoresponse function of the material was endowed with photosensitive azobenzene into the dye without changing its printability. Yang et al. used the FDM method to produce an ink containing carbon black and polyurethane to construct a 3D-printed sunflower having excellent light response. Aghakhani et al. used the two-photon lithography method to create a bullet-shaped microrobot with a spherical bubble. The robot can be driven unidirectionally by resonating sound waves within an internal bubble and using a magnetic field to control its direction of motion. This invention has considerable significance in the medical field. Zheng et al. used the DIW method to embed poly(acrylic acid-co-acrylamide) and poly(acrylic acid-co-N-isopropyl acrylamide) into 3D structures. They utilized the mismatch of these materials in their response to ionic concentrations to produce a smart structure that could deform when immersed in a concentrated saline solution. To demonstrate the application of this type of smart structure, they fabricated a soft gripper having excellent mechanical properties and fast response speeds (Figure 12A).

Smart structures based on the swelling principle manufactured via 3D printing have also been proposed. Le Duigou et al. applied the FDM method to the manufacture of wood-fiber-reinforced biocomposites having high hygroscopic

![Figure 12](https://www.advancedsciencenews.com)
sensitivity but relatively poor mechanical properties. Yuan et al.\cite{122} combined two hydrogel layers with different swelling capabilities using the DIW method to create a soft hydrogel actuator with programmable deformation and camouflage capability (Figure 12B). Zhao et al.\cite{123} used the g-DLP method to produce various origami structures based on the swelling principle. To achieve more complex swelling deformations, a multiwavelength DLP method that can control the chemical composition of the printed material in real time during the printing process was proposed to achieve different swelling capabilities in multiple directions.\cite{124} Other studies on 3D-printed smart structures having selective transmission and color-changing functions were performed. Inspired by the eggshell membrane, a conductive carbon nanofiber membrane with selective permeability fabricated by the DIW method was proposed (Figure 12C).\cite{125} Gregory et al.\cite{126} used the DIW method to print poly(ε-caprolactone) polymer filaments containing spiropyran. A smart structure capable of mechanical and photochromic discoloration was then fabricated, and its potential application as a sensor capable of visually evaluating peak loads was demonstrated (Figure 12D). However, research on 3D-printing methods for most of these smart structures is still in its infancy.

7. Discussion

This work provides a comprehensive classification of smart structures and a detailed review of the functional materials and 3D-printing methods used for manufacturing various smart structures (Figure 13). Common 3D-printing methods and materials, the applications of various smart structures, and their main limitations are discussed. Future development trends are also proposed.

7.1. Limitations

3D printing is suitable for manufacturing various smart structures because of its multimaterial compatibility and capacity for personalized customization and integrated fabrication. Different types of smart structures require different 3D printing methods. In all of the reviewed smart structures, the DIW, DLP, and µSL printing methods were commonly adopted. The size requirements and high costs of AJP and µCOP methods and the magnetic response functional specificity of the M-PSL method limit the universal application of these methods. The relationship between 3D printing speed, resolution, and cost has always been a trade-off. Therefore, the key to choosing an appropriate 3D printing method is balancing that trade-off.

The 3D printing of smart structures faces certain problems. First, many smart materials having excellent properties are not printable because they have unsuitable rheological properties or poor light and thermal stability. Moreover, although many improved 3D-printing methods have been proposed, effective methods for many materials remain lacking. The defects and low interface bonding strength of 3D-printed structures significantly reduce their durability. The large-scale manufacturing capacity of 3D printing technology is also inadequate, limiting the size of printed smart structures. Furthermore, many smart structures remain in the demonstration stage with no specific application scenario. Accordingly, further research on 3D printing is necessary, as discussed in Section 8.

7.2. Conclusion

In summary, 3D printing technology allows the rapid and high-precision manufacture of complex smart structures. In turn, the increasing demand for the fabrication of smart structures promotes the advancement of 3D printing technology. Future research directions for 3D printing smart structures should focus on the following items.

In terms of materials, the search for new printable high-performance smart materials or the improvement of existing printable materials’ performance is crucial. Approaches to provide new properties to traditional matrix materials by doping them with functional factors or rheology modifiers are compelling. For example, carbon nanotubes may be mixed with PLAs to achieve excellent electrical conductivity;\cite{108} hydroxyl iron powder may be incorporated into the hydrogel to obtain superior magnetic response properties;\cite{76} and PLAs may be doped with

Figure 13. Summary of 3D-printing methods in various smart structures.
polymers containing dynamic Diels–Alder functionality to achieve superior interlayer bonding performance.\cite{11}

In terms of smart-structure function, the exploration of multifunctional structures is expected to be an inexorable trend. Currently, most smart structures can only perform single functions. The development of multifunctional smart structures will improve their performance and broaden their application scope. For example, damage inside a smart structure is accumulated through repetitive deformation, thus reducing durability. Accordingly, smart structures having both self-healing and stimulated deformation capabilities have been proposed.\cite{26,33,94} Structures that integrate stimulus deformation and conductive functions\cite{26} and those that integrate self-monitoring and self-repair functions\cite{12} have been presented. Moreover, smart structures that respond to multiple stimuli have been proposed, which will make it possible to apply the same smart structure to multiple scenarios.\cite{86}

From a manufacturing perspective, the development of new 3D-printing methods and equipment is expected to continue. Methods suitable for new high-performance smart materials will revolutionize smart-structure manufacturing. Notably, a 3D-printing method for in situ manufacturing that prints objects directly where they need to be used has been proposed. This could allow the immediate fabrication of smart structures at the installation position, thereby improving manufacturing efficiency and making possible in situ manufacturing in harsh environments (e.g., the moon). For developing new 3D printing equipment, the installation of quality monitoring devices will allow real-time monitoring and correction during printing and will reduce defects while improving printing accuracy. Equipment that can modify materials and allow real-time monitoring opens the door to new printing materials and more complex structures.

In terms of application, smart structures need to achieve real-world implementation. Therefore, after conceptualization, the driving force and displacement capacity of the smart structure should be increased, and the response period must be reduced. Second, the durability of smart structures requires more study. Currently, most temperature-responsive materials cannot withstand high temperatures, and some materials (e.g., hydrogels) commonly used in smart structures cannot function effectively under harsh conditions. Furthermore, theoretical models for high-precision quantitative analysis of the intelligent behavior of various smart structures must be formulated. Finally, considerable effort should be devoted to the miniaturization of smart structures, and further research on large-scale structures is necessary.

Keywords
3D printing, electromagnetic-response, self-healing materials, sensing materials, smart structures, stimulus response

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Conflict of Interest
The authors declare no conflict of interest.

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