

Developments in 4D-printing: a review on current smart materials, technologies, and applications

Zhizhou Zhang, Kahraman G. Demir and Grace X. Gu

Department of Mechanical Engineering, University of California, Berkeley, CA, USA

ABSTRACT

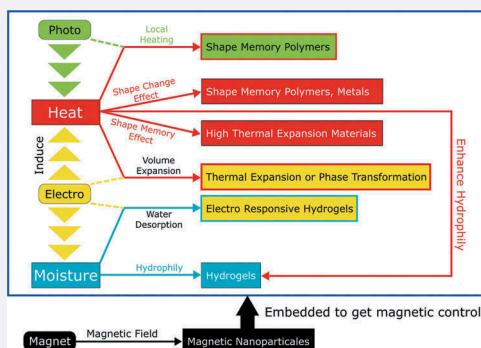
Recent advances in additive manufacturing (AM), commonly known as three-dimensional (3D)-printing, have allowed researchers to create complex shapes previously impossible using traditional fabrication methods. A research branch that originated from 3D-printing called four-dimensional (4D)-printing involves printing with smart materials that can respond to external stimuli. 4D-printing permits the creation of on-demand dynamically controllable shapes by integrating the dimension of time. Recent achievements in synthetic smart materials, novel printers, deformation mechanism, and mathematical modeling have greatly expanded the feasibility of 4D-printing. In this paper, progress in the 4D-printing field is reviewed with a focus on its practical applications. We discuss smart materials developed using 4D-printing with explanations of their morphing mechanisms. Additionally, case studies are presented on self-constructing structures, medical devices, and soft robotics. We conclude with challenges and future opportunities in the field of 4D-printing.

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1. Introduction

Additive Manufacturing (AM), commonly known as three-dimensional (3D)-printing, continues to evolve through nearly 40 years of development, allowing researchers to create complex shapes previously impossible using traditional fabrication methods [1–8].

CONTACT Grace X. Gu  ggu@berkeley.edu  Department of Mechanical Engineering, University of California, Berkeley, CA 94720, USA

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Using 3D-printing, researchers are able to create complex, bioinspired, multi-material designs [9–15], remotely actuated robots [16,17], designs generated from optimization algorithms and machine learning [18–24], drug delivery systems [25,26], and even microenvironments for bio-tissues [27,28]. Common 3D-printing technologies include material extrusion, vat photopolymerization process, powder bed fusion, material jetting, binder jetting, sheet lamination process, and directed energy deposition [3]. Among all these methods, fused deposition modeling (FDM) and stereolithography (SLA) are the most utilized. FDM is one of the material extrusion technologies, which feeds material filaments into a heated nozzle and then deposits the molten material on a printing bed layer-by-layer. SLA is a vat photopolymerization process, which scans the liquid UV-curable material with a laser. This technology can achieve better resolutions and higher printing speeds than FDM, however, it is much more expensive [3]. Despite the rapid advancement in AM technologies, 3D-printing is not yet ready to be used for large scale manufacturing due to its slow cycle time and the undesirable tradeoff between quality and scale [2]. However, 3D-printing is widely adopted by researchers for its high customizability and capability of printing complex geometries [2]. Moreover, recent developments in computer-aided design (CAD) software [29] and in novel materials [30] further expand the stage for 3D-printing.

Conventional 3D-printing technologies focus on fabricating static structures from a single material, which cannot meet all the demands of dynamic functions needed for applications such as soft grippers for surgery [31], self-folding packaging [32], and adaptive wind turbines [33]. According to research from Momeni et al., printing wind turbine blades that mimic plant leaf structures by shape memory polymers (SMP) can greatly improve the structural and mechanical properties of the blades [33]. The blade can deform when heated to adapt to the wind and retain its original shape when reheated in a calm environment [33]. The smart blade is fabricated in one print cycle with no electromechanical parts or post-assembly needed. This kind of functionality benefits from a fourth printing dimension: time, which will allow for transformation. This idea of a fourth dimension introduced a new research area branched from AM called four-dimensional(4D)-printing. 4D-printing, thus, generates metamaterial structures with different superimposed structural responses triggered by changes in their operational environment.

The idea of 4D-printing was first introduced by Tibbit [34], after which the definition has been refined by researchers as 3D-printed structures that display targeted shape or property transformation under external stimulus [1,30,35]. Currently, most research focuses on the shape changing ability of 4D-printed structures including bending, elongating, twisting, and corrugating. These behaviors can be further programmed to make lockers [36], lifters [37], microtubes [38], robots [39], and even toys [40]. To achieve directional shape change (contrary to trivial uniform swelling), the 4D-printed structure will consist of material anisotropy. This requires congruently printing a combination of multiple materials.

Research in 4D-printing is closely related to 3D-printing where its advancement can provide new possibilities for 4D-printing. The various research topics in 4D-printing fall into the following categories: development of equipment, deformation mechanisms, and mathematical modeling, as shown in Figure 1. Development of equipment encompasses synthesizing new materials characterized by different forms of responses [38,41–44] and

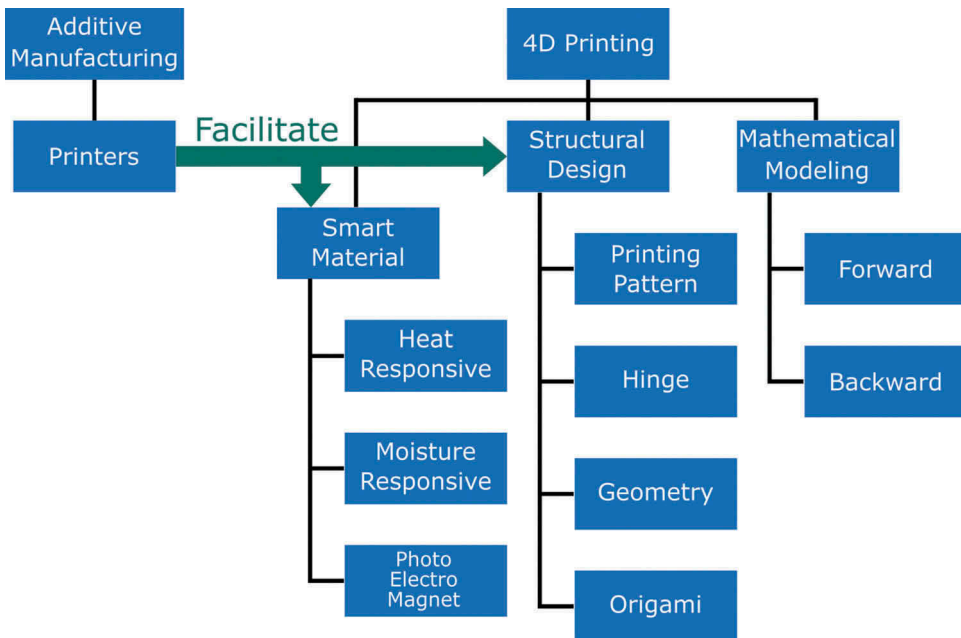


Figure 1. Research directions in 4D-printing encompassing development of equipment, deforming mechanisms, and mathematical modeling.

advancing printing technology [17,45], both of which are the foundations for 4D-printing. 4D-printers are essentially a specialized subset of 3D-printers. Currently, the applicable 4D-printing methods include direct inkjet cure [32,46–49], fused deposition modeling [37,50,51], stereolithography [52], laser-assisted bioprinting [53] and selective laser melting [54]. The choice of the printer should be carefully made based on different types of smart materials which will be introduced further in the next section. Research on deformation mechanisms focuses on the understanding of sequential folding [38,41], hinge design [32,49,55], geometry design [37,56], and pattern design [48,52]. Understanding these mechanisms is critical for 4D-printing functional structures. The mathematical modeling field includes backward prediction, which provides the printing profile given a target shape and function [57–60] and forward prediction which models the deformation process given the initial profile [43,47,48,59,60]. Many modeling processes can be understood by the comprehensive system developed by Momeni et al. who established three basic laws that most 4D-printed structures will follow [61].

In the following section, various types of smart materials in 4D-printing are summarized with explanations of their morphing mechanisms. Then, we present case studies in three major application fields: self-construction structures, medical devices, and soft robotics. Finally, we will discuss future opportunities and challenges in the field of 4D-printing.

2. Materials selection for 4D-printing

Recent developments in multi-material 3D-printing have allowed for more flexible and precise material placement [62], capabilities that are essential for 4D-printing. In this

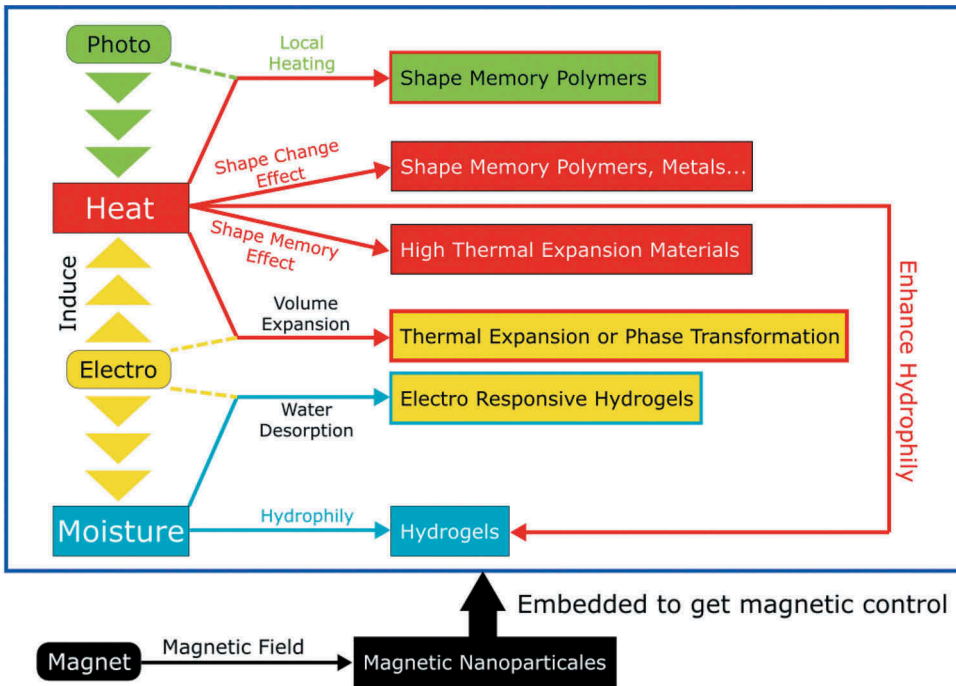


Figure 2. Classes of smart materials that can respond to different types of stimulus including heat, moisture, light, electricity, and magnetic fields.

section, we classify 4D-printing materials according to their environmental and/or temporal stimuli, as shown in Figure 2. Various stimuli will be discussed including temperature, moisture, electricity, light, current, and magnetic fields.

2.1 Thermo-responsive

The deformation of thermo-responsive materials is mainly driven by one of two mechanisms: the shape memory effect (SME) [63] or the shape change effect (SCE) [64]. Materials based on the SME are called shape memory materials (SMM) which can be further divided into shape memory alloys (SMA), shape memory polymers (SMP), shape memory hybrids (SMH), shape memory ceramics (SMC) and shape memory gels (SMG) [33]. SMPs are the most favored by researchers for their ease of printability. SMPs usually have glass transition temperatures (T_g) that are higher than their operating temperatures. They are programmed under specific heat and mechanical treatments above their glass transition temperatures and then cooled to be fixed at a temporary shape free of external loading. The specimen then returns to its original permanent shape after its temperature is raised above its T_g [63]. To utilize their special properties, various SMP materials are modified by researchers to be printable. An SMP ball is fabricated by Ying et al. through SLA where the liquid resins are polymerized under UV illumination to fix the permanent shape. The ball is able to span into a flat plane and retract back with high durability [65]. Ge et al.

printed an SMP flower that could bloom upon heating [66]. This technology is also used to fabricate smart grippers that require no assembly or electromechanical parts. Recent findings from Bodaghi et al. revealed the possibility of preprogramming an SMP structure through a good utilization of the heating process in FDM printers [67,68].

SMPs, which obey the SME mechanism, usually have two or three discrete states where no intermediate stable shape can be maintained. By contrast, the SCE is proportional to the stimulus applied, i.e., it is continuously variable between its extreme states [64]. For thermo-responsive materials, the SCE usually occurs in bilayer structures that have a sufficient difference in their coefficients of thermal expansion (CTE). Since the interface area between the layers must remain the same, a stress field is incurred and the structure bends. Hu et al. demonstrated a graphene-based bimorph structure that can expand to a flat sheet when heated and can roll back into a cylinder when cooled. To achieve such a drastic deformation by SCE, either a sharp temperature change or combination of unique materials is needed. As shown by Hu et al., there is a difference of two orders of magnitude between the CTE of the different layers in the film [69].

2.2 Moisture-responsive

Water- or moisture-responsive materials are of high interest owing to their ubiquitous stimulus and the broad range of applications. Hydrogels are extraordinary moisture-responsive materials because their hydrophilicity allows them to expand up to 200% of their original volume. Additionally, hydrogels as a class of polymer materials exhibit high printability. The advantage of using hydrogels lies in their biocompatibility and ease of printing with direct ink writing [70]. However, their slow reverse response means researchers must wait hours until hydrogels are dried and shrunk. To program the behavior of hydrogels, one must endow anisotropy to the swelling. Gladman et al. combined hydrogel ink with cellulose fibrils which can be aligned through shear forces induced by the contact between the ink and the print bed [60]. This alignment makes the transverse swelling strain four times that of the longitudinal strain; this allows for the programming of the 4D-printed structure. Mao et al. printed a structure where the hydrogels are confined in one direction by stiff materials so that the swelling is anisotropically directed [71]. Quick responses are reported by Zhang et al. who designed thin hydrophobic films fabricated from cellulose stearoyl esters (CSEs) that can react faster and more precisely [44].

Hydrogels are normally immersed in an aqueous environment causing them to absorb water until their moisture saturation point, which limits the intermediary controllability of hydrogels. However, the swelling of hydrogels can be controlled through the temperature of the aqueous environment. Breger et al. fabricated microgripper joints from the pNIPAM-AAc soft-hydrogels that are gradient crosslinked [31]. By heating or cooling the water where the gripper is immersed, it is possible to achieve reversible actuation by adjusting the saturation point. Special hinge designs are also applied to

avoid over-swelling. Tibbits et al. demonstrated a self-folding bilayer structure fabricated from PolyJet printers [40]. Rigid plates are printed in the joints so that the folding stopped at a preprogrammed angle. When this angle is reached, the plate tips touch each other to provide a resistance to excessive bending.

2.3 Photo-responsive

Unlike heat and moisture, light is an indirect stimulus; an exposed area of a photo-responsive material absorbs light as heat. Liu et al. showed a sequentially controlled self-folding structure [46]. Light power is absorbed by joints as heat where the rate is determined by the colors of the joint and the light source. Kuksenok et al. used light as a trigger for deformation in a very different way. A certain amount of photo-responsive chromophores are infiltrated into some locations of a polymer gel block so that these parts only swell under exposure to light [43]. Moreover, the versatility of light as a stimulus is shown in patterning the print. By projecting weak UV light on liquid resin, a gradient crosslink in depth can be reached where the anisotropy helps to bend the 4D-printed structure [72].

2.4 Electro-responsive

Like light, current can be used as an indirect stimulus in 4D-printing. Miriyev et al. demonstrated a printed soft artificial muscle made from a mixture of silicone elastomer and ethanol. When a current is applied, heat is generated through resistive heating causing the ethanol to evaporate. This phase shift from liquid to gas greatly increases the ethanol's volume and thus expands the whole matrix [42]. A current is also applied to polypyrrole (PPy) films to control the water absorption or desorption. Okuzaki et al. applied PPy films to an origami micro robot whose feet had special geometries so that it met less resistance when moving forward. When placed in a humid environment, a voltage drives the head forward due to absorption of moisture, and the tail follows up when desorption was caused by voltage absence [41].

2.5 Magneto-responsive

4D-printed structures that respond to magnetic fields are termed magneto-responsive materials. Breger et al. incorporated magnetic nanoparticles into a microgripper printed from hydrogels and achieved remote control by applying magnetic fields [31]. The embedment is carried out in pre-processing where ferric oxide powders are mixed with the material solution. This technology has potential, also, in polymer printing and metal printing. One drawback is the restriction of the size of the print, which must be sufficiently lightweight to be affected by the magnetic field.

In the next few sections, we discuss three case studies on current applications of 4D-printed materials.

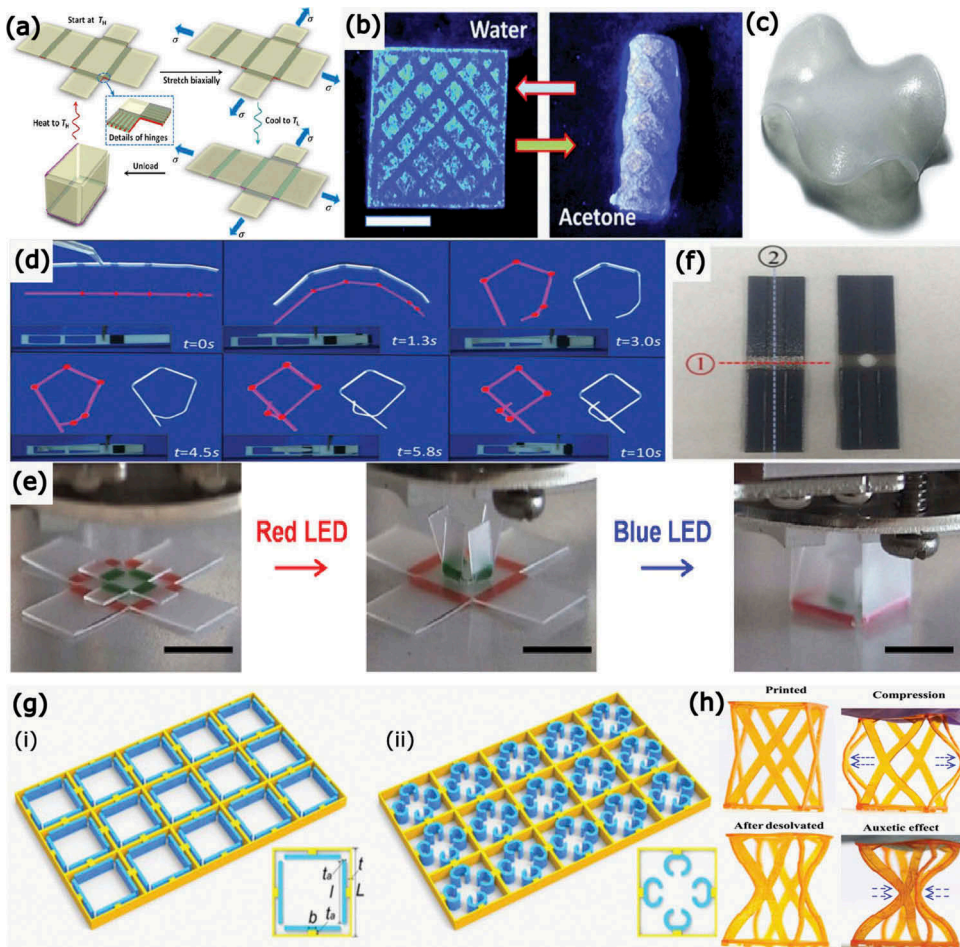


Figure 3. 4D-printed self-construction structures. (a) A self-folding cubic container printed from SMPs, adapted from [32]. (b) A self-rolling tube container printed from smart polymers and actuated by acetone, adapted from [73]. (c) A complex curved surface deformed from a flat state that was printed using moisture-responsive polymers (hydrogels) with a gradient distribution, adapted from [40]. (d) A sequentially folded stipe that can lock itself. The joints are printed with different combinations of SMPs, adapted from [36]. (e) Self-nested boxes whose joints are printed with SMPs in different colors, adapted from [46]. (f) Cross-folding test specimens with and without a stress release hole, adapted from [76]. (g) Conceptual design of a material with tunable lattice structure that permits bandgap control, adapted from [77]. (h) Multifunctional material with tunable Poisson's ratio, adapted from [72].

3. Case study 1: self-construction structures

As depicted by Campbell et al. in their article 'The Programmable World,' self-construction structures have evolved from a conceptual stage to an experimental stage [29]. 4D-printing is one major approach that can combine fabrication of shape-changing prints and self-assembly with carefully designed localized swelling or shrinking

regions [29]. In this section, we discuss some representative self-construction applications that are built by 4D-printing and the methods used in recent years.

3.1 Containers

The most basic 4D-printed self-construction structure can be a self-folding cube as shown in [Figure 3\(a\)](#). This design proposed by Ge et al. was first printed as a flat sheet with five patterned hinges. The hinges consist of two layers, where one of them contains polymer fibers that exhibit shape memory effect. The SMP fibers can sustain plastic deformation at low temperatures while the other layer remains elastic. This allows for a stress mismatch between the layers when the structure is heated and cooled under stretch and thus folds into a cubic container when released [32]. As a result of the shape memory effect, it self-unfolds when reheated. This technology has the potential to reinvent packaging once it reaches a larger scale such as self-folding furniture or packages.

Besides cubic containers, self-rolled cylindrical tubes can also be designed through 4D-printing. Su et al. printed a square flat sheet where a matrix of smart polymer grids is uniformly distributed [73]. This allows each point on the surface to bend uniformly in a prescribed direction instead of the local edge folding in the cubic design as shown in [Figure 3\(b\)](#). Thus, the sheet is able to self-roll into a tube. Similar ideas are also used where strips are obliquely rolled into a tube [38]. Containers are versatile in medical applications which will be discussed in more detail in [section 4](#).

3.2 Surface curving

Smart materials can also be printed in a gradient distribution to allow researchers to control the surface curvature of a print. Tibbit et al. printed a grid sheet with moisture-responsive polymers concentrated on specific areas to generate a sinusoidal surface after the print was immersed in water [40]. With more precise gradient control of polymer placement, a multi-phase deformation with more complex surface characteristics is achieved as shown in [Figure 3\(c\)](#). This technology can greatly save space by fabricating structures as flat shapes. It is also capable of realizing complicated surface curvatures that are difficult to manufacture through traditional methods. Suong 4D-printed straight beams which were programmed to bend to a certain curvature through a shape change effect [74]. This technique allowed them to easily manufacture springs with plane molds. Moreover, SMPs are also applicable to curved surface structures. Momeni et al. fabricated a composite surface structure with one of its layers printed using SMP. Additionally, the authors programmed the layer so that the surface could deform to a target curved shape that best concentrated solar energy [75].

3.3 Lockers and safe boxes

In the applications mentioned above, all the deformations are global, i.e., the smart materials are all triggered at the same time in a uniform stimulus environment. This simultaneous response limits the range of motion of a structure as there must be no physical interference between each moving segment. To address this, Mao et al. printed a polymer stripe ([Figure 3\(d\)](#)) combining two types of shape memory polymers at

different concentrations for each joint so that the joints had a series of incrementing glass transition temperatures. This allowed the shape memory effect to be triggered sequentially during heating which leads to the creation of self-interlocking shape memory structures [36].

Similar sequential folding is also achieved by Liu et al. who printed the joints in different colors to differentiate their light absorption rates as shown in Figure 3(e) [46]. The researchers printed a nested box structure where a smaller polymer sheet was folded into a box under red light exposure and a larger box was constructed under blue light to encompass the smaller one. Besides sequential folding, triple shape memory polymers can be a potential alternative to avoid self-interference through multi-stage folding. This possibility is shown by Bodaghi et al. who demonstrated three shape stages through hot FDM extrusion programming, cold loading programming, and a final heating-cooling process [67].

In addition to sequential folding and multi-stage folding, cross-folding is needed for complex designs where the folding edges can pass through each other. Teoh et al. proved the effectiveness of printing a stress-releasing hole at the intersection of two folding edges where cracks were greatly reduced, as shown in Figure 3(f) [76]. Also, thicker sheets survived longer in cyclic cross-folding in their experiments. Their results show a necessity of adding materials, or extra features, to cross-folding structures to avoid stress concentration.

3.4 Multifunctional materials

As mentioned in the introduction, 4D-printing also aims to control the properties of the composite material. In recent years, researchers have attempted to achieve a property change from structure deformation. Zhang et al. proposed a conceptual lattice structure printed from thermo-responsive polymers as seen in Figure 3(g) [77]. There are four polymer beams in each lattice covering the four edges. The beams are thermo-responsive and can roll into a circular shape upon heating. The simulation results indicate a band gap change caused by the lattice structure morphing [77]. Moreover, Wu et al. printed a structure that exhibited a negative Poisson's ratio (the cross-sectional area shrinks when compressed in the longitudinal direction) after it absorbed water as shown in Figure 3(h) [72]. The polymers used were cured by grayscale patterned UV light, which allowed an anisotropic structural deformation under stimulus [72]. The supports are fabricated in a straight vertical orientation where a positive Poisson's ratio pertained. Once immersed in acetone solution, the support would bend inward due to a higher absorption rate at the inner wall.

4. Case study 2: medical devices

3D-printing has made a significant impact on medical fields [78], especially where personalized medical treatments are important, such as dentistry [79], implants [80,81] and prosthetics [82–84]. 3D-printing has even opened the doors to the customization of pharmaceutical tablets by combining multiple drugs in defined quantities on single tablets [85]. These developments can be further enhanced with 4D-printed devices by utilizing their shape changing properties, which will be explored in this section.

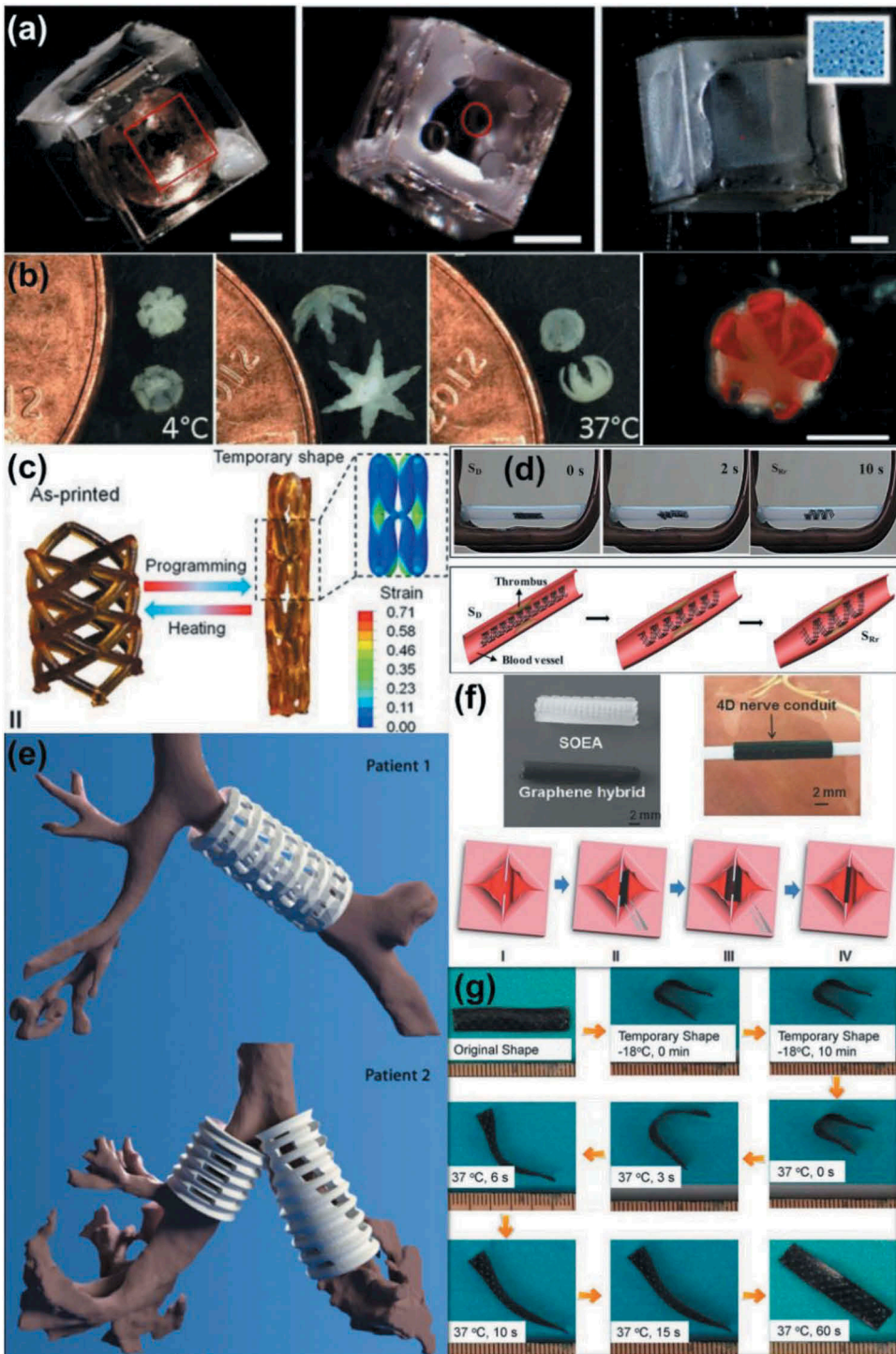


Figure 4. 4D-printed medical devices. (a) Photolithographically fabricated polymeric containers, intended for drug delivery, with different patterned porous faces outlined in red. Scale bar is 250 μm long, adapted from [86]. (b) Thermo-responsive shape change demonstration of a theragripper. Right most image shows a theragripper gripping a clump of cells, adapted from [87]. (c) 4D-printed stent, adapted from [66]. (d) A 4D-printed magnetic stent, adapted from [89]. (e) 4D-printed air-way splints that have been surgically implanted, adapted from [90]. (f) A 4D-printed nerve guidance conduit, adapted from [52]. (g) Shape memory demonstration of 4D-printed scaffold, adapted from [91].

4.1 Targeted drug delivery

Targeted drug delivery is a method of delivering medication to specific intended locations in the body. Owing to their many possible stimuli, 4D-printed devices can contain pharmaceutical drugs and release them when the environment of the targeted location provides the correct stimulus. 4D-printed containers (Figure 3(a), (b) and (e)) are a canonical example used for drug containment and release.

Azam et al. demonstrated the fabrication of containers using traditional photolithography. The containers are made from SU-8 photoresist panels and biodegradable polycaprolactone (PCL) hinges that are thermo-responsive. The SU-8 panels are made porous with different arrangements and sizes of holes, as can be seen in Figure 4(a), to study the release of the contents of the container [86]. Malachowski et al. fabricated thermo-responsive multi-fingered grippers, which they have referred to as 'theragrippers', that are intended to be used for the controlled release of drugs in the gastrointestinal tract. In the work, the theragrippers are fabricated using traditional photolithography and consist of rigid poly(propylene fumarate) segments and stimuli-responsive poly(N-isopropylacrylamide-co-acrylic acid) hinges. The theragrippers actuate above 32°C allowing them to grip onto tissue at body temperature to sustainably release drugs. Figure 4(b) shows a theragripper gripping a clump of cells [87].

4.2 Stents

The fabrication of a stent requires numerous steps due to its complex and patient-specific geometry [88]. 4D-printing allows for quick fabrication of customized stents. Additionally, the shape memory property of the stent allows for the minimization of surgical invasion of the implantation by first printing the stent at the desired final diameter and then programming the stent to a smaller diameter for ease of implantation and correct placement. After the implantation, the stent will return to its original diameter at body temperature.

Bodaghi et al. 4D-printed a metamaterial lattice consisting of flexible beams with integrated SMP fibers that were arranged into a tubular shape to demonstrate one of its potential applications as a stent. They also developed material and structural thermo-mechanical models which were used to perform finite element simulations to predict the deformation of the lattice structure when thermally actuated [56]. Furthermore, the authors developed a finite element formulation describing the programming of materials in the 4D-printing process. These computational tools are critical to the design and fabrication of 4D-printed devices [68].

Ge et al. 4D-printed a stent using high-resolution projection micro-stereolithography to prove the efficiency of fabricating stents using 4D-printing and to demonstrate the applicability of shape memory polymers to the fabrication of stents, as shown in Figure 4 (c) [66]. Wei et al. have demonstrated direct-write printing of ultraviolet cross-linking PLA-based inks as a viable fabrication technique with which they have 4D-printed stents (Figure 4(d)) with magneto-responsive materials allowing them to be magnetically remotely guided to their destination [89].

4.3 Splints

Geometry is a critical aspect for biomedical devices such as splints where the fit of the device on the subject is essential for the proper function of the device. However, in cases where the growth of the subject is significant, traditionally fabricated biomedical devices have difficulties in accommodating for the growth over time. 4D-printing these devices could allow for the accommodation of shape change due to growth.

Morrison et al. have performed three successful medical treatments involving the surgical implantation of 4D-printed external airway splints in multiple infant patients with tracheobronchomalacia (TBM). TBM is a life-threatening condition in which the airway of the patient excessively collapses during respiration. The splint is designed to accommodate the growth of the airway, consequently preventing unwanted compression. [Figure 4\(e\)](#) shows 3D models of the airways of two infant patients with custom splints around the affected regions. Morrison's surveys have shown that the infants' airways had continued to grow healthily and that their TBM complications had been resolved [90].

Nerve guidance conduits (NGCs) are tubular devices that facilitate nerve regeneration and must meet physical, chemical and biological requirements that allow for tissue formation. Using a naturally derived, photo-crosslinking monomer (soybean oil epoxidized acrylate, SOEA), Miao et al. 4D-printed (using stereolithography) an NGC that served multiple desired functions for nerve tissue regeneration ([Figure 4\(f\)](#)). The shape memory property of the NGC allows for easy readjustments (through thermal stimulation) to be made during implantation. Additionally, the shape memory property provides axial tension necessary for guiding regrowth. Graphene was used to further enhance other critical properties of the NGC [52]. Miao et al. also 4D-printed a biomedical scaffold ([Figure 4\(g\)](#)) with SOEA. The scaffold possesses excellent shape changing properties and has excellent attachment and proliferation of human mesenchymal stem cells when compared to the traditional polyethylene glycol diacrylate scaffolds [91].

5. Case study 3: soft robotics

Traditional robotics, primarily due to being made of rigid materials, have limitations in performing organic and compliant operations such as the grip of a human hand or the intricate motion of an octopus tentacle. Consequently, the field of soft robotics emerged in which certain soft materials, mainly special types of elastomers, are utilized as the interaction interface between robots and their environment. These soft materials allow for a gentle interaction with fragile objects and, when compared to traditional robotics, allow for a better tolerance towards damaging forces [92].

5.1 Shape-changing actuators

Miriyev et al. have developed a composite material for use as a McKibben type [93] soft actuator (SA). The SA is made up of a porous silicon elastomer matrix with the porous voids filled with ethanol. Owing to the ease of preparing and handling the composite

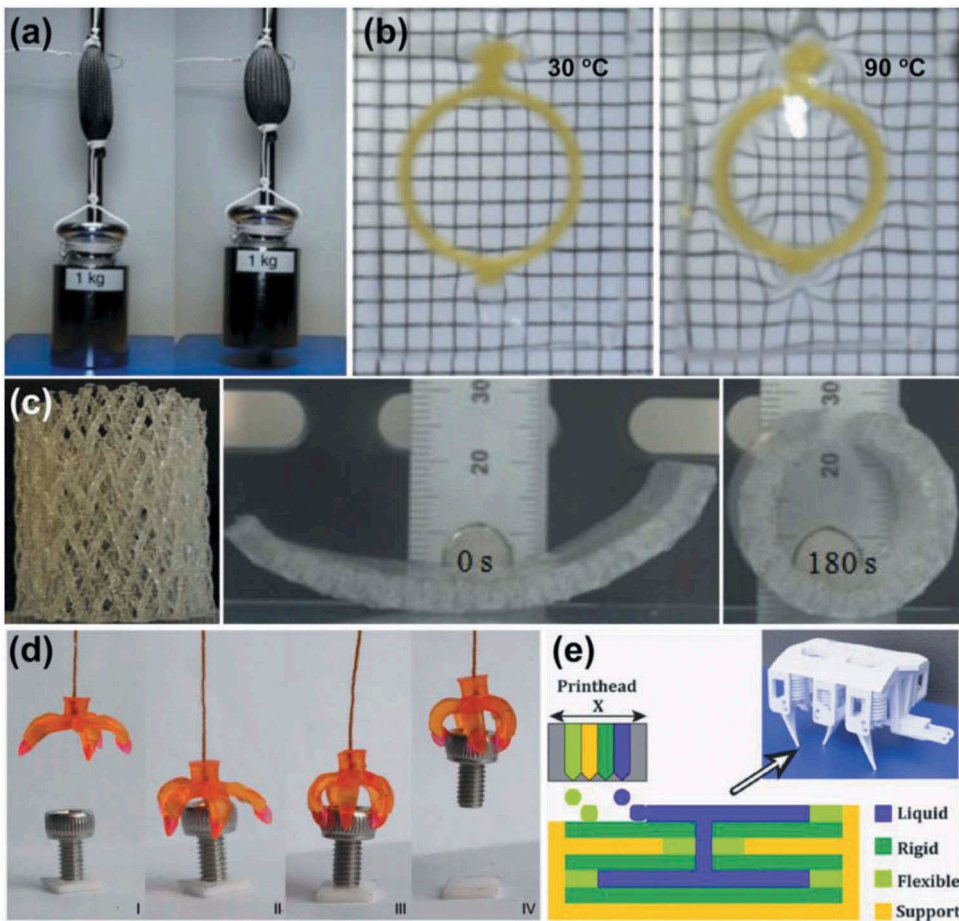


Figure 5. 4D-printed robotic devices. (a) Soft McKibben type actuator made from porous silicon elastomer with the pores filled with ethanol, adapted from [42]. (b) Demonstration of 4D printed thermo-responsive liquid crystal elastomer used for adaptive optics, adapted from [95] (c) 4D-printed polylactic acid braided tube preform demonstrating thermo-responsive shape memory behavior as a gripper, adapted from [99] (d) 4D-printed SMP gripper, adapted from [66]. (e) 3D-printed hydraulic robot using liquid support for bellow actuators, adapted from [17].

material, it can be 3D-printed with conventional techniques. It is actuated through the application of heat through resistive heating which evaporates the trapped ethanol causing the elastomer matrix to expand. The material is capable of strains up to 900% and stresses of up to 1.3 MPa at a low density of 0.84 g cm^{-3} . Miriyev et al. claimed that the material can lift a maximum of 1700 times its own weight. A cyclic loading of 60 N on 6 g of the composite material was performed in which no signs of degradation were visible. The SA is low cost, simple to fabricate and environmentally friendly. Figure 5(a) shows the SA lifting a 1 kg weight in a skeletal bicep configuration [42].

The dielectric elastomer (DE) is a type of smart material that can produce large strains through the application of a voltage across it. Its structure consists of an elastomeric dielectric sandwiched between two flexible electrodes. DEs have been vigorously studied for various applications, mainly as actuators. Traditional fabrication methods

involve multiple elements of manual labor resulting in inconsistencies in the DE device. McCoul et al. have devised a method of 3D-printing DE silicon materials using drop-on-demand (DOD) inkjet printing, paving the way for the fabrication of complex 3D structures of DEs. Their printed DEs outperform traditionally fabricated DEs by achieving larger area strains under the same applied nominal electric field [94].

SMPs have also been used as shape-changing actuators in simple grippers as a demonstration of their capabilities as seen in [Figure 5\(c\)](#) and [\(d\)](#). However, SMPs have slow response times, taking up to several minutes to fully actuate. This makes them an unsuitable alternative in conventional robotic applications where quick and precise motions are prioritized. Nevertheless, SMPs can be actuated through many different stimuli which may make them favorable to be used as actuators in highly specific situations. A realistic application of SMP actuators can be derived from the utilization of 4D-printing to create complex structures that can deform in unorthodox ways. A great example of this can be seen in the work of López-Valdeolivas et al. where 4D-printed thermo-responsive crosslinked liquid crystal polymers were used to demonstrate a potential application in adaptive optics, as seen in [Figure 5\(b\)](#) [95].

5.2 Hydraulic and pneumatic actuators

One major challenge with 3D-printing is supporting overhanging structures, especially geometries that are internal or difficult to access. Traditionally, support structures are built simultaneously with the end product. The support structure is either: (1) printed in a manner where the interface between the support material and the end product is made fragile to aid in removal of the support structure or (2) the support material is dissolved in a solution and the end product is obtained. However, both techniques are unable to assist in the removal of internal support structures without tedious design alterations. This has hindered the ability to 3D-print complex structures with functional internal geometries. MacCurdy et al. have demonstrated a technique for inkjet-based 3D-printing for the fabrication of internal structures using non-curing liquid as the supporting medium. Using this technique on a Stratasys Objet260, they 3D-printed a hydraulic robot ([Figure 5\(e\)](#)) with linear bellow actuators that required no assembly or modification after being printed [17].

Pneumatic artificial muscles (PAMs) are pneumatic actuators made of flexible and inflatable materials. PAMs are able to exert relatively large forces and are low in weight [93]. Combining these characteristics with the versatility of 3D-printing will allow for complex pneumatically actuating mechanisms. Yap et al. 3D-printed soft pneumatic actuators using fused deposition modeling (FDM) technology and have tested their performance characteristics. The actuators can produce large forces and cyclic fatigue experiments show that the actuators are highly durable. Furthermore, the trajectory of the actuation (i.e. bending) is consistent and reliable [96].

6. Challenges and future opportunities

4D-printing, although a novel technology, has the potential to solve many real-world problems. However, many challenges in this field are yet to be overcome. One major challenge is the limitations of current 3D-printers to address fundamental 3D-printing

issues such as avoiding support structures, especially for inaccessible internal structures (Section 5.2), simultaneously printing different material groups (e.g. polymers and metals), the lack of low-cost printable materials, and slow print times. Printing technologies need to be fundamentally improved or new printing technologies must be developed. 5-axis 3D-printing is currently of high interest to address some of these 3D-printing challenges. Another major challenge is the restriction imposed on the mechanical properties of 4D-printed structures by the desired shape or property transformation. For example, certain ratios of polymers are necessary for the sequential folding of the locker structures described in Section 3.3. Advances have been made in smart printable materials with wide ranges of mechanical properties [97,98]; this research area is critical for advancing 4D-printing.

Other challenges include slow and inaccurate actuation, lack of control of intermediary states of deformation, and limited material availability. Future studies could investigate more efficient techniques for the application of stimuli, for example, improving the heat application process in thermo-responsive SMPs, or methodologies for controlling moisture absorption for hydrogels. These improvements could also allow for enhanced actuation accuracy. Furthermore, property changes occur (discussed in section 3.4) as a result of macroscale structural transformations which may be undesirable. Understanding the effects of the scale of structural patterns and the mechanics of the transformations will allow for more flexibility and applicability, which shows the promise of widespread use and future opportunities in the field of 4D-printing.

7. Conclusions

4D-printing has progressed in the past few years and holds promise to impact many fields. In this review, highlighting research on 4D-printing and its applications, we discuss its multiple use cases. Specifically, we examine case studies in three domains: self-construction structures, soft robotics and medical devices where innovative devices were 4D-printed to serve functions that would be impossible or extremely costly to fabricate with traditional manufacturing methods. 4D-printed devices are candidates for applications in unusual environments due to their vast customizability and absence of mechanical elements. 4D-printed devices have immense potential in the medical field, where patient-specific designs of medical devices are crucial. Surgical treatments involving 4D-printing have already been performed and have been successful demonstrating the extent to which 4D-printing has grown in its influence. Advancements in printable smart materials, mathematical models, and printing technologies will allow for 4D-printing to further enhance surgical treatments, targeted drug delivery, soft robotics, and other unthought-of fields in engineering.

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Disclosure statement

No potential conflict of interest was reported by the authors.

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