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A brief review of mechanical designs for additive manufactured soft materials

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Abstract

Additive manufacturing is an arising technology for soft materials and structures with improved complexity and functionality and has gradually become widespread in various fields, including soft robotics, flexible electronics and biomedical devices. Along with the development of material systems and fabrication techniques, mechanical design principles for additive manufactured soft materials have been greatly developed and evolved in recent years and some unique issues that are distinct from conventional manufacturing techniques have emerged. In this short review, we mainly focus on additive manufactured soft materials that are in significant need of mechanical models/simulations to provide design guidelines; therefore, topics such as soft robotics and electronics are not considered here. We first discuss the mechanical design methods for controlling shape distortions and interfacial strength, as they are directly related to the quality and reliability of additive manufactured soft materials. Design principles and manufacturing strategies for bioinspired composites, which represent a large part of current research on additive manufactured soft materials, are then summarized integrally with regards to three aspects. In addition, basic mechanical considerations for additive manufactured four-dimensional shape-changing structures are explained, together with a review of the recent theories and numerical approaches. Finally, suggestions and perspectives are given for future developments in soft material additive manufacturing.

Keywords: Additive manufacturing, soft materials and structures, mechanical design

INTRODUCTION

Recent advances in additive manufacturing [or three-dimensional (3D) printing] has greatly broadened the design space and application fields of soft materials[1,2]. Traditional soft structures and devices are generally fabricated by molding[3] or cutting[4], which are only suitable for simple geometries. Other common subtractive manufacturing approaches for stiff materials can easily cause damage when applied to ultrasoft material systems[5]. In additive manufacturing, soft materials are step-by-step piled up with relatively small contact or deflective force, thereby enabling the non-destructive construction of complex soft structures. Benefitting from the discrete manufacturing process, it is also possible to spatially control the distribution of heterogeneous materials without involving tedious steps. Such advantages make additive manufacturing a promising technique for the direct fabrication of soft robotics[6], flexible electronics[7] and biomedical devices[8]. For example, turtle-inspired robotics consisting of fluidic circuits and interconnects, soft actuators and outer castings were integrally manufactured by multi-material Polyjet printing, and could be actuated by an external pressure source[9]. 3D electronic metamaterials with programmable large-strain piezoresistive and piezoelectric effects were fabricated by combining additive manufacturing with selective metal deposition[10]. Active biomedical devices that can perceive deformation and process signals were additive manufactured using hydrogel inks and bacterial cells[11]. Other typical additive manufactured soft materials and structures include mechanical metamaterials[12], optical devices[13] and microfluidic devices[14].

Traditional mechanical analysis and characterization of soft materials are generally decoupled from the manufacturing process[15,16]. Before manufacturing, an optimal structure is designed considering the target functionality, structural reliability and manufacturability, which are established by carrying out theoretical analysis or numerical simulations. After the soft structure is manufactured, mechanical experiments and analysis are conducted to examine the structural stiffness, vibration behavior, interfacial strength and so on. In additive manufacturing, where the fabrication process is highly flexible and programmable, the steps of mechanical design have significantly changed[17,18]. Firstly, with the development of multi-material printing techniques, many additive manufactured soft structures can be considered as ready-to-use devices, instead of raw materials or components[19]. The as-printed soft structure has its own functionality, either multifield response or programmed deformation, and does not need to be further assembled with other accessories. Key issues arise in mechanical designs for these additive manufactured soft devices, including, but not limited to, the design of material distribution, microarchitectures, reaction kinetics in manufacturing and the pre-definition of deformation patterns. Secondly, additive manufacturing is a process strongly related to a chemical reaction or phase change, and thus the mechanics during fabrication become more important compared with traditional subtractive manufacturing[20]. Here, the most significant issues in mechanical design include how to control manufacturing-induced shape distortion and ensure high geometric accuracy and how to program and utilize the intrinsic anisotropy of additive manufactured soft materials [Figure 1][21,22].

Although there are already several review articles on additive manufactured soft materials in the literature, most of them are focused on novel material systems, advanced print strategies and the application of additive manufacturing in different fields[1,2,6,7,12,22-26]. As discussed above, the significance of mechanical design arises concerning the accuracy, and reliability of additive manufactured soft materials. In this article, we briefly review recent progress in mechanical theories and design methods for additive manufactured soft materials. The mechanical designs related to the manufacturing process are reviewed, where we mainly concentrate on how to control geometric accuracy ("distortion design" in Figure 1), how to program

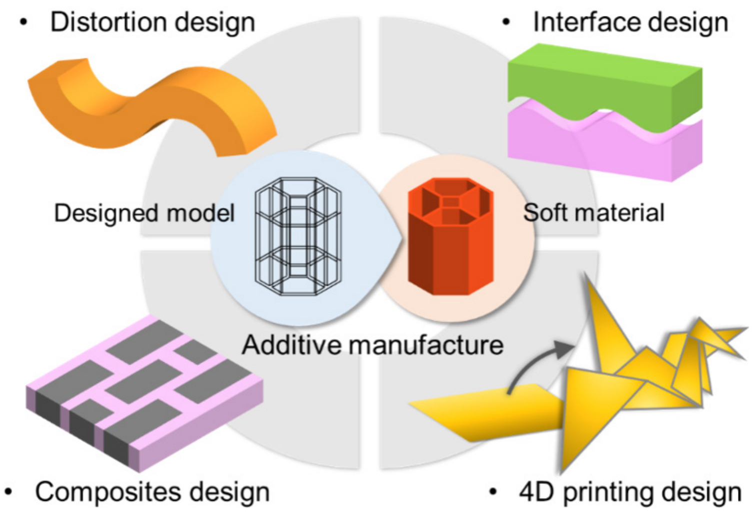

Figure 1. Schematic of mechanical design of additive manufactured soft materials based on the four most important aspects [distortion design: how to control the geometric accuracy of additive manufactured soft materials; interface design: how to ensure the interfacial strength and to program the anisotropy of additive manufactured soft materials; composites design: how to manufacture multi-material soft composites; four-dimensional (4D) printing design: how to manufacture 4D shape-changing soft structures].

material anisotropy and how to improve the interfacial strength ("interface design" in Figure 1) of printed structures. The mechanical design principles for multi-material soft structures that can be treated as structural or functional composites ("composites design" in Figure 1) are summarized. Mechanical design methods of 4D shape-changing soft structures ("4D printing design" in Figure 1) are also discussed. Finally, the perspectives and conclusions are given.

MECHANICAL DESIGNS FOR SHAPE DISTORTION

Light-based 3D printing technologies [e.g., stereolithoraphy (SLA)[27], selective laser sintering[28], digital light processing (DLP)[29,30] and two-photon polymerization[31]] and ink-based 3D printing technologies [e.g., inkjet printing[32], fused deposition modelling[33,34] and direct ink writing (DIW)[35,36]] allow for the rapid design and assembly of various soft materials into structures with tunable mechanical, electrical, optical and other functional properties at a wide spectrum of length scales (from ~100 nm to ~300 cm)[26,37,38]. So far, various 3D-printed prototypes have been conceptualized and validated. However, it is often not feasible to directly utilize 3D-printed prototypes to replace their conventionally-manufactured counterparts, especially for industrial products[39]. One of the issues that hinder the practical application of 3D printing lies in the inaccurate geometric shape in the 3D-printed parts due to shape distortion, which is induced by the development of complex internal stress during the layer-by-layer material adding process of printing. There are many process mechanism-dependent sources that can cause internal stress, with the two most common being volume shrinkage and thermal deformation. To efficiently reduce shape distortion and improve shape accuracy in 3D printing, the development of quantitative methods to predict the evolution of internal stress is urgently required.

In a light-based 3D printing process that solidifies materials through photopolymerization, the internal stress mainly builds with volume shrinking deformation in materials experiencing a transition from liquid to solid[40]. Previous studies have reported that during photopolymerization, some acrylate-based monomers exhibit a ratio of volume shrinkage exceeding 20%[41]. Such a large shrinking deformation can cause severe shape distortion in the printed parts, making them unacceptable for applications where a high-resolution geometric shape is required.

Shape distortion in the printing method of SLA has been studied since the 1990s. Starting from the pioneering work of Jacobs[42], many researchers have investigated the printing process of SLA[43,44]. Bugeda numerically analyzed the SLA processes using a self-developed finite element code and concluded that the dependence of the curl distortion with respect to the magnitude of the volumetric shrinkage is linear only for short parts and becomes non-linear for longer parts[45]. Wiedemann *et al*.[46] also performed a numerical analysis on the SLA printing process and revealed a common rule where half the layer thickness doubles the curl. By relating the material modulus and cure shrinkage to the combinations of the averaged energy dose and curing time through several empirical equations, Huang *et al*.[47] developed a dynamic finite element method to simulate the curl distortion of SLA-printed structures and found that short raster scanning produces less distortion than long raster scanning [Figure 2A]. The dynamic finite element method was later adopted by the same authors in another study in order to optimize the scanning parameters[48].

In addition to finite element method-based numerical approaches, simplified theoretical models have also been developed for studying shrinkage strains in a SLA-printed part. For example, Karalekas *et al*.[40] applied elastic lamination theory to determine the magnitude of the resulting shrinkage strains in a laser solidified acrylic-based photopolymer [Figure 2B] and made the conclusions that test specimens post-cured under intense ultraviolet light exhibited a uniform shrinkage, while thermal post-curing resulted in higher shrinkage strains.

The aforementioned works successfully analyzed the shape distortion in SLA-printed parts and some design rules for optimizing printing parameters were proposed to increase shape accuracy. However, most of these studies only consider shape distortion at the structural element scale, such as a long strip or a thin layer, and ignore the complex features of the dynamics of photopolymerization. A recent study conducted by Westbeek *et al*.[49] overcame these limitations by developing a multi-physical modeling framework to predict the deformed geometry of SLA-printed parts on a full-component scale. Multiple samples were printed for comparison with the simulation and the results showed that the distorted geometry depends on the irradiation profile and the polymerization and solidification characteristics [Figure 2C][49]. In another study[50], the same authors investigated the SLA printing process where particles are filled in resins by using a multiscale process simulation framework. In this work, the influence of the filler particles on the light scattering, conversion characteristics and resulting effective thermal and mechanical properties can be determined.

DLP is a printing technique that also uses photopolymerization to solidify materials and the internal stress therefore mainly comes from volume shrinking as well. Different from SLA that patterns one volume element at a time through point-source exposure, the printing method of DLP relies on the dynamic mask patterning technology to illuminate one layer of the liquid resin at a time[29,30]. This greatly speeds up printing but also separates the printing process from the SLA and requires new analytical approaches, as well as new design guidelines to reduce shape distortion. Wu developed a theoretical model where modulus and shrinkage are related to cure to understand the curing process for DLP. A finite element analysis that incorporates the model was then performed to calculate the dynamic evolution of stresses during the layer-by-layer fabrication[51].

Wang *et al*.[52] conducted a coarse-grained molecular dynamics simulation to model the continuous liquid interface production (one form of DLP) 3D printing technique [Figure 2D] and simulations showed that the quality of the shape of the 3D-printed objects is determined by a fine interplay between the elastic, capillary and friction forces. The authors identified the source of the object shape deformations and developed a set of rules for calibrating the parameters to meet the accuracy requirements. Very recently, Zhang *et al*.[53]

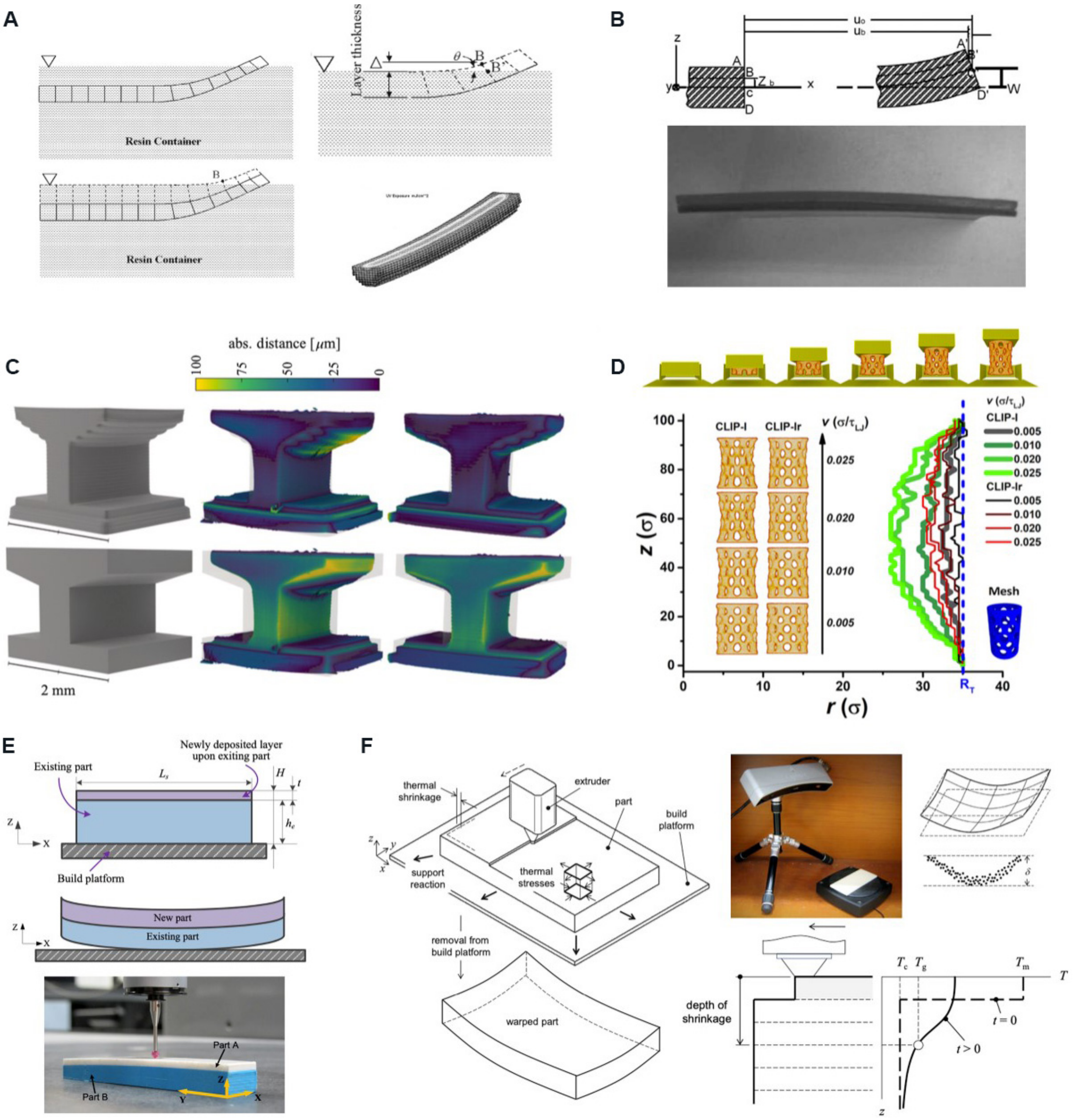

Figure 2. Mechanical designs for the shape distortion of additive manufactured soft materials. (A) Schematic of the rezoning technique in the dynamic element method simulation of SLA-printed parts[47]. (B) Schematic and experiment result of the deformation of a two-layer resin laminate[40]. (C) Comparison of the simulated geometry with the printed samples and the target geometry with the printed samples for a T3-shape[49]. (D) Coarse-grained molecular dynamics simulation of continuous liquid interface production and dependence of the shape and radius of the object on the printing speed[52]. (E) Part distortions occurring during the material adding process[57]. (F) Warpage in the FDM (Fused Deposition Modeling) process and warpage evaluation[55].

investigated the volume shrinkage-induced distortion of DLP-printed parts by conducting experiments, theoretical modeling and finite element analysis simulations. Design rules that include adjusting the layer printing time, the layer thickness, as well as the content of photoabsorbers and photoinitiators, were given to reduce shape distortion of DLP-printed structures. For shape distortion induced by the post-curing process, Wu *et al*.[54] theoretically investigated the shape distortion of DLP 3D-printed structures during ultraviolet post-curing by combining photopolymerization reaction kinetics and the Euler-Bernoulli beam theory. The results showed that the evolution of the mechanical behavior of the printed samples during the post-printing process was dependent on the printing parameters, which provides fundamental understanding for optimizing the final shape of 3D-printed structures[54].

Another common source of internal stress is thermal deformation, which can occur in 3D printing technologies that require heat to process materials. FDM is one such printing technology and thermal deformation-induced shape distortion is a difficult problem for FDM. FDM printing relies on melting thermoplastic materials by an extruder and then depositing the material onto the printing plate for cooling and solidifying. During a FDM printing process, the material can experience multiple thermal cycles [Figure 2F][55]. After printing (with the part peeled from the building plate), these residual strain drive the printed part to curl at the corners. Warpage in FDM-printed parts can depend on many factors related to material properties, part geometry and process parameters. Towards estimating the warpage built with different process settings, many modeling approaches have been proposed in either one-dimensional (1D), two-dimensional (2D) or 3D frameworks and validated by experimental tests. According to some basic hypotheses and simplifications, Wang *et al.*[56] proposed a 1D model of the prototype warp deformation and quantitatively analyzed many variables, including the deposition layer number, the stacking section length, the chamber temperature and the material linear shrinkage rate. Moreover, improved methods for reducing the warp deformation were proposed.

A similar mathematical model was proposed by Zhu *et al.*[57] to identify the influential parameters that contribute to part distortions and the parameters were used in their experimental designs [Figure 2E]. For 2D models, Liu *et al.*[58] adopted the theory of elastic thin plates in thermoelasticity to describe the deflection field in the x-y plane and the warpage estimated by the model was verified on 81 test specimens with a portable 3D laser scanner. For more complex problems, Zhang *et al.*[59,60] conducted a 3D finite element analysis to simulate the mechanical and thermal phenomena in FDM. The method was further used for simulations of residual stress and part distortion. From the simulations, the short-raster tool path results in higher residual stresses and thus possibly larger distortions than the long-raster and alternate-raster patterns. The same authors also conducted a follow-up study to evaluate the effects of the deposition parameters on residual stresses and part distortions, where the parameters include three factors and three levels. Shape distortion has also been analyzed for other polymer-based AM processes by considering specific shrinkage effects, as well as further process-dependent mechanisms. Based on models and tests, design rules are proposed for selective laser sintering[61,62] and binder jetting[63].

Based on the above discussion, strategies for reducing the degree of shape distortion are highly dependent on the specific process mechanisms. The general goal is to reduce the residual stress during the manufacturing process, which can usually be realized by adjusting the printing conditions, including the light intensity, layer thickness for photopolymerization-based printing, printing speed, printing temperature and printing path for thermal-based printing. The useful strategies for reducing distortion in some soft material-based additive manufacturing techniques are summarized in Figure 3.

MECHANICAL DESIGNS FOR INTERFACIAL STRENGTH AND ANISOTROPY

In most additive manufacturing techniques, soft materials are layer-by-layer deposited or part-by-part piled up. This process creates many internal interfaces inside a soft structure, for both single- and multi-material printing. Before printing, mechanical designs should be carried out to eliminate the influence of these interfaces on the final mechanical performance. The existence of internal interfaces also results in macroscopic anisotropy, for example, layer-by-layer printed structures perform similarly to composite laminates. Mechanical designs of the printing path and processing conditions are needed to effectively control this type of anisotropy.

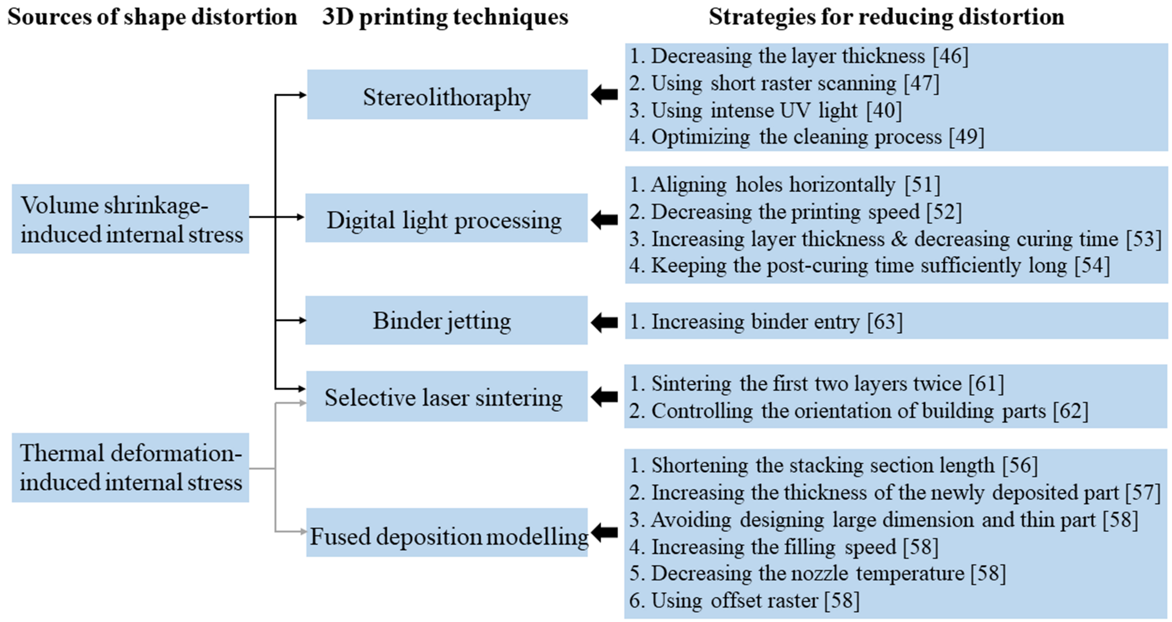

Figure 3. Strategies for reducing shape distortion in various soft material-based additive manufacturing techniques.

Weak interfaces in additive manufactured single materials come from inconsistencies in the reaction history or processing sequence. In the FDM printing of polymers or soft composites, thermoplastic precursors are sequentially heated up and deposited onto the structure, during which materials on two sides of a certain interface experience different and periodic thermal histories[64,65]. The strength of such an interface is dependent on molecular diffusion and chain entanglement near the interfacial region[66], which can be quantitatively correlated with the equivalent thermal history of the interface. Thermal viscoelastic models incorporating the Williams-Landel-Ferry equation[67], as well as multiscale thermal mechanical models considering the evolution of polymer chains[68], can be utilized to predict interfacial strength and to optimize the macroscopic thermal history[69]. In DLP-based additive manufacturing, interfaces are created when liquid resin is layer-by-layer cured. Due to the inhomogeneity of the photopolymerization reaction, materials near an internal interface have weaker mechanical properties and sometimes might be as low as half those of the base material[70]. Based on delicate mechanical models considering details of the reaction kinetics, researchers have found that the heterogeneity near the interfacial region can be quantitatively controlled by the printing parameters[71,72] [Figure 4E]. It should be noted that recently developed continuous[73] and volumetric DLP printing techniques[74] have significantly reduced the amount of internal interfaces, but there is still intrinsic heterogeneity inside the soft material. The internal interface in DIW is a combination of the aforementioned two types of interfaces because many DIW systems utilize partially cured precursors that will continue to react after been deposited on the structure[26,75].

Another important category of interface is that between different materials, which is common in multi-material additive manufactured soft structures. Early multi-material additive manufacturing mainly utilized chemically similar materials, for example, the acrylate resin used in Polyjet printing[76] and DLP printing[77]. Desirable interfacial strength could be obtained, although interlocking structures are sometimes designed to further improve the interface under large deformation[78]. For chemically dissimilar materials, either chemical treatments or physical designs become necessary. The group of Lewis designed a microfluidic nozzle to homogeneously mix different materials and to create gradient interphase regions with both

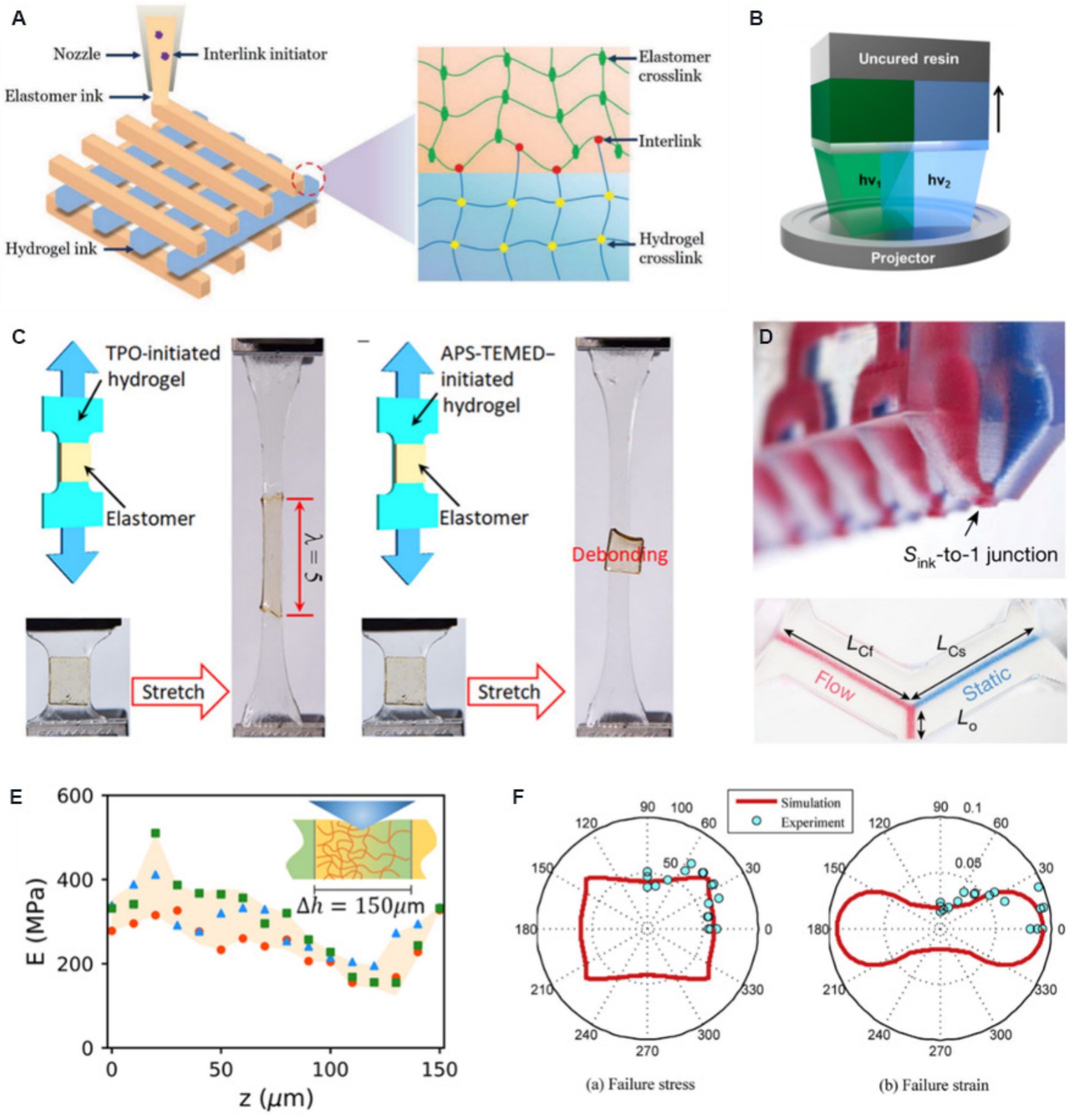

Figure 4. Mechanical designs for the interfacial strength and anisotropy of additive manufactured soft materials. (A) Improving the interfacial strength between chemically dissimilar materials by adding initiators to trigger interfacial reactions[81]. (B) Improving the interfacial strength by selective wavelength irradiation to create interfacial penetration[83]. (C) Improving the interfacial strength with a water-soluble initiator[82]. (D) Improving the interfacial strength by material mixing[79,80]. (E) A weak interface and the origin of macroscopic anisotropy in DLP-printed materials revealed by indentation[71]. (F) Anisotropic failure stress and failure strain of additive manufactured polymers determined from experiment and simulation[94].

chemical and physical connections [Figure 4D][79,80]. Yang *et al.*[81] supplied additional initiators into the precursor, which promoted the formation of interfacial covalent bonds during printing [Figure 4A]. Ge *et al.*[82] used water soluble 2,4,6-trimethylbenzoyl diphenylphosphine oxide to increase the interfacial strength between polymers and hydrogels [Figure 4C]. Dolinski *et al.*[83] utilized selective wavelength irradiation to create interpenetrating networks within the interphase region, which could be used in solution mask liquid lithography [Figure 4B]. These chemical approaches have the intrinsic drawback that they are only suitable to special material systems.

For general chemically dissimilar materials, a combination of the chemical and physical approaches would be more helpful. Multi-material interfaces in nature generally have structured geometries and specific geometric parameters exist when the interfacial strength is optimized[84,85]. Roach *et al.*[86] and Wu *et al.*[87] employed blunt and buzzsaw interfaces to improve the interfacial strength between elastomers and thermosets that could not be chemically bonded together. Rivera *et al.*[88] further incorporated laminate designs near the structured interface, by which the interfacial strength of the additive manufactured connection was almost doubled.

The existence of internal interfaces results in macroscopic anisotropy, even for single-material additive manufactured soft structures[89]. In the DLP printing of polymers, Monzon *et al.*[90] found that the elastic modulus of vertically-printed materials could be 300 MPa higher than that of horizontally-printed materials. In path-dependent FDM printing, the anisotropy in modulus and failure strength became more apparent[91] but could be intentionally regulated by sequentially aligning the printing path[92]. Many additive manufactured soft structures adopt lightweight porous infills and isotropic lattice designs would be beneficial for such structures[93]. To quantitatively predict the anisotropy of additive manufactured materials, Zhang *et al.*[94] developed a phenomenological hyperelastic-viscoplastic model that could precisely capture the anisotropic failure stress and failure strain [Figure 4F]. Chen *et al.*[95] considered the anisotropy arising from porosity evolution to develop a viscoelastic-viscoplastic model for Multi Jet Fusion-printed polymers. Liu *et al.*[96] incorporated material anisotropy into the topological optimization of self-support structures. For future developments, details of the origin of the internal interface can be included in these theoretical approaches to quantitatively correlate the mechanical behavior with the manufacturing process.

MECHANICAL DESIGNS FOR BIOINSPIRED COMPOSITES

Natural materials, such as bone, nacre, wood, silk and hair, have outstanding mechanical performance in resisting impacts and tolerating damage because of their unique microstructure architectures across multiple length scales[97-100]. The building blocks that constitute these materials are rather simple and most of them do not have remarkable mechanical properties. It is instead the complex architectures that endow the materials with superior performance. This has inspired scientists and engineers to emulate designs in natural materials and create novel materials with bioinspired structural features. However, due to the complexity of these architectures, it is difficult for conventional manufacturing techniques (e.g., casting) to reproduce the design patterns observed in natural materials. In contrast, state-of-the-art 3D printing techniques allow digital materials to be placed into almost arbitrary positions in a 3D space at a "voxel" level[39] and have thus become suitable platforms for designing, modeling and manufacturing various bioinspired materials. Currently, 3D printing-enabled bioinspired materials can be roughly classified into three categories, namely, composites reinforced by complex-oriented fibers or particles, composites composed of two material components arranged in a brick block-like layout and composites consisting of gradient material constituents.

Balsa wood is a cellular natural composite and achieves its remarkable properties from the hierarchical structure of biological polymers. Inspired by balsa wood, Compton *et al.*[101] created a cellular, lightweight composite with controlled alignment of multiscale, high aspect ratio SiC fiber reinforcement through DIW [Figure 5A]. In DIW, because fiber alignment occurs in the direction of the print path, the mechanical properties, including stiffness and toughness, of balsa wood-inspired composites can be designed and optimized by reasonably planning the print path. By additionally adding a rotary union into the standard DIW platform and using the so-called rotational direct-ink writing technique, the orientation of the extruded fiber reinforcement can be decoupled from the prescribed print path, such that each voxel of the printed material can itself be spatially varied for locally controlling fiber orientation, which allows the

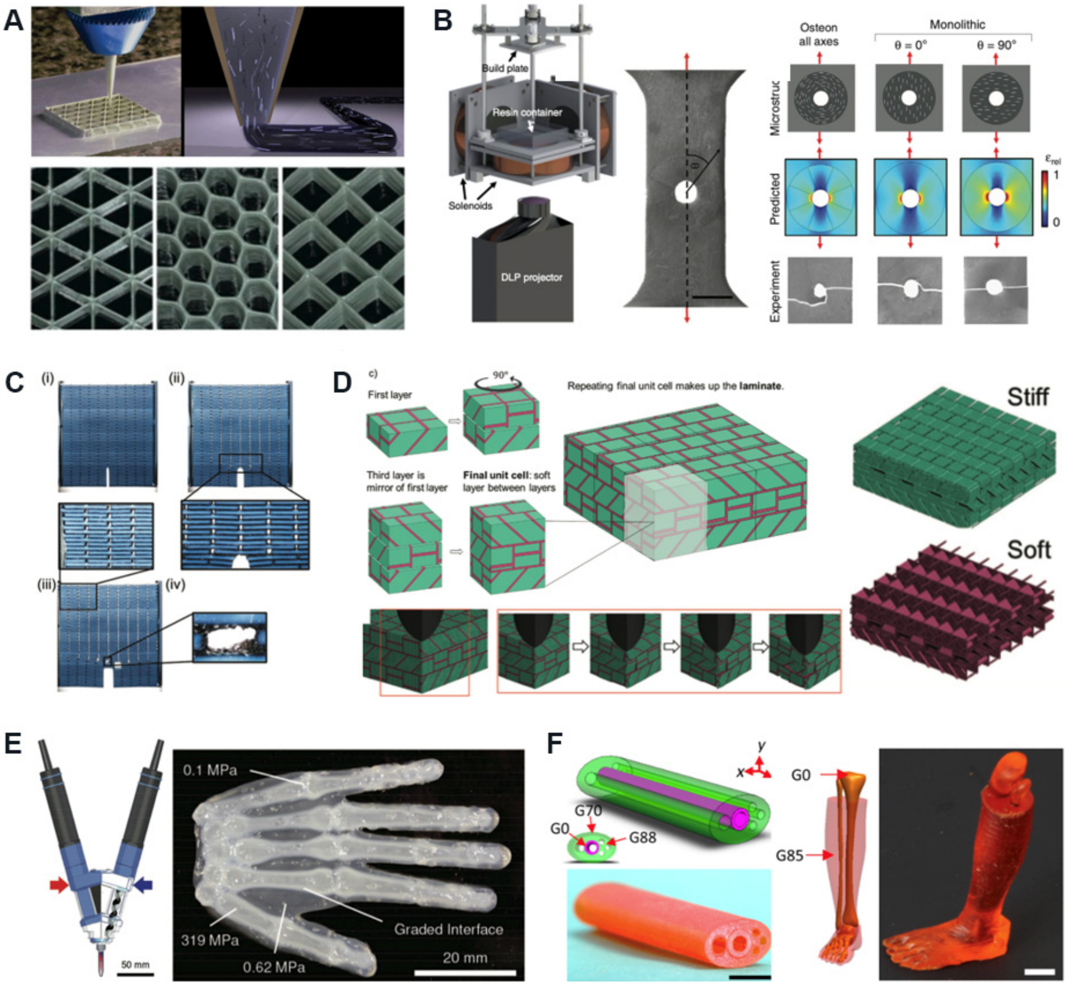

Figure 5. Mechanical designs for bioinspired soft composites. (A) Design and printing of various honeycomb structures using SiC-filled epoxy materials[101]. (B) Microstructural design of samples with circular defects using 3D magnetic printing that can program the orientation of stiff platelets[105]. (C) Snapshots displaying deformation and fracture mechanisms for a 3D-printed specimen with a brick block-like material layout[111,112]. (D) Design of material arrangement in a conch shell-inspired structure and the mechanisms of crack propagation in the structures[117]. (E) Schematic of a two-component dispenser and a printed hand-like structure with gradient materials[120]. (F) Design and print of a limb-mimic structure using gradient digital light processing[121].

mechanical response of the resulting material to be site-specifically optimized[102]. The inspiration of this work comes from natural composites that use helical fiber arrangements to enhance damage tolerance, such as the hammer-like stomatopod dactyl club and wood cell walls.

Complex-oriented fiber reinforced materials can also be designed based on multi-material inkjet 3D printing and topological optimization. Boddeti *et al.*[103,104] developed a general framework that integrates the workflow for the optimal design and fabrication of novel multiscale laminated composites with spatially varying microstructures. The developed approach in their work allows for design automation, material compilation and voxel-based digital fabrication. Apart from the above methods, external field-assisted 3D printing techniques have also been utilized by researchers to fabricate bioinspired composite reinforcement architectures. Martin *et al.*[105] [Figure 5B] and Kokkinis *et al.*[106] proposed the use of magnetic 3D printing to allow for the rapid assembly of magnetized stiff platelets in rather complex orientations. Materials that replicate the microstructural composition of abalone shell, peacock mantis shrimp and mammalian cortical bone were successfully designed and fabricated. In addition to short fiber or particle reinforcement, long or continuous fiber reinforcement is also chosen by some natural materials, such as bone, to simultaneously achieve excellent stiffness and toughness. Mueller *et al.*[107] borrowed this design inspiration and created a cellular composite composed of flexible epoxy core-elastomeric silicone interfacial layer-brittle epoxy shell motifs through a multicore-shell, 3D printing technique. The produced material is found to exhibit both high stiffness and toughness. Boddeti *et al.*[108] applied their approach previously developed for short fiber reinforced composites to continuous fiber reinforced composites and realized the creation of laminated material with variable stiffness. Bioinspired materials have also been achieved by other printing techniques, such as FDM[109].

In nature, some structural materials exhibit fracture-resistant properties by arranging two dissimilar constituents in a brick block-like layout. Such a material layout can greatly promote the deflection of crack propagation and energy dissipation[110]. A typical example is nacre, which is composed of a stiff and a compliant phase with a structured brick-and-mortar arrangement. Dimas *et al.*[111,112] computationally designed two types of nacre-like composite materials with predicted fracture response based on multi-material 3D printing [Figure 5C]. It was shown that the synthesized composites exhibit an order of magnitude larger toughness and modulus than its fundamental building blocks. Going beyond simple tension or compression loadings, Gu *et al.*[113] created nacre-like composites with superior impact performance compared to their monolithic stiff phase. In their design, two base materials with vastly different properties were firstly assembled in a ply with an architecture similar to nacre, these plies were then stacked with orientation angles of 0° and 90° to form a laminate construct. In another study, the same authors proposed an approach that can generate the optimized material layout starting from a composite system with a distinct set of unit cells. This approach combines machine learning with a self-learning algorithm and can design hierarchical materials with tougher and stronger properties[114].

In addition to nacre, cortical bone is another natural composite material that adopts a staggered layout of stiff and soft phases. Libonati *et al.*[115] designed and tested synthetic composites with a pattern inspired by the microstructure of cortical bone and the results demonstrate that the bone-inspired design is critical for toughness amplification and balance with material strength. In their design, the soft phases were used as inclusions while the stiff phases were isolated by the soft inclusions. Although this design can efficiently increase toughness, it limits the level to which the modulus of the composite can be elevated. Lei *et al.*[116] avoided this limitation by using stiff phases as inclusions and creating a different enhancement design for the staggered composite. In addition to 2D patterns, a 3D hierarchical pattern for material layout was proposed by Gu *et al.*[117] to obtain a composite with enhanced impact resistance, the inspiration of which derives from biological conch shells [Figure 5D].

Many biological tissues in the human body are composed of gradient material constituents. For example, the skeletal muscle is divided into muscle belly and tendons. The muscle belly is made up of muscle fibers and is red and soft with contractility, while tendons are made up of dense connective tissue and are white and hard with no contractility.

The 3D printing of gradient materials has found biomedical applications in fields covering tissue engineering[118] and prosthetic replacement[119]. Kokkinis *et al.*[120] developed a 3D printing platform to fabricate elastomer gradients spanning three orders of magnitude in elastic modulus. To demonstrate the potential applications, they designed a structure that resembled a hand using graded materials for the interface and three other different materials for distinct regions in the "hand" [Figure 5E][120]. Kuang *et al.*[121] conceived the concept of using grayscale digital light processing to 3D print highly functionally graded materials. One of their demonstrated examples included a structure that mimics a human limb where the soft muscle region was printed using G85 light while the stiff bone region was printed using G0 light [Figure 5F][121]. Bioinspired graded composites can also be designed based on other 3D printing techniques, such as multi-material inkjet printing[122,123] and extrusion-based multi-material printing[124,125].

MECHANICAL DESIGNS FOR 4D SHAPE-CHANGING STRUCTURES

4D printing is a novel concept where additive manufactured structures can change their shape or function upon external actuation[126]. Currently, most 4D printing research is based on soft materials, including polymers, hydrogels and biomaterials[127], whose mechanical programming is the most crucial design step. In 4D printings based on active materials, mechanical design is required to predict the deformation pattern and actuation speed. In 4D printings based on structural innovation, the mechanics of beams, shells or frames should be utilized to provide information regarding the geometry, deformation route and stability.

The design strategy for 4D printed active materials is similar to the mechanical design of classical materials that respond to external stimuli. Firstly, the constitutive model of the material coupled with the external stimulus field is developed to describe the fundamental rule of deformation. The constitutive model is then incorporated into theoretical calculations or numerical simulations to precisely predict actuation[128,129]. The optimal design configuration can be obtained by iterative theoretical designs and experiment verifications. Special concerns in designing 4D printed active materials include the correlations between their responsive properties and the manufacturing process. For example, changing the printing parameters leads to either residual tensile stress or residual compressive stress in additive manufactured shape memory polymers, whose evolution rule could be included in the mechanical design theory[130-132] [Figure 6A]. Another example is additive manufactured hydrogels whose local swelling ability is correlated with the printing process, so the prediction and pre-definition of local material heterogeneity are necessary for programmable deformation after the hydrogel structure is immersed in solution[133-135] [Figure 6B].

In 4D printed liquid crystal elastomers and magnetoactive polymers that have anisotropic actuation behaviors, the microscopic analysis of particle alignment and the macroscopic planning of the printing path are both critical design issues[136,137]. Recently, many data-driven topological optimization and inverse design methods have been proposed and have realized the fast customerization of 4D shape-changing soft structures and increased the design space for complex deformation patterns[138-140] [Figure 7A-D]. Wu *et al.*[138] and Hamel *et al.*[140] used an evolutionary algorithm to design the actuated shape of a bi-material beam, where the designed configurations could be directly voxelized for additive manufacturing. Sossou *et al.*[139] developed a computational design tool that integrated the design, modeling and inverse optimization of complex 4D shape morphing structures. Zhang *et al.*[141] used a convolutional neural network to quickly predict the large deformation of digital active materials. In the work of Su *et al.*[142], machine learning was adopted to establish the correlation between manufacturing parameters and experimentally obtained curvatures of shape morphing SU-8 strips.

In addition to 4D printing based on active materials, structural mixing and hierarchy is another strategy that can induce automatic shape changing even for non-active materials. Here, we briefly introduce some deformation modes that are commonly used in 4D printing designs. Bending based on laminate or gradient design is probably the most widely adopted deformation mode and its theoretical foundation has been well established[143]. When designing 4D shape-shifting soft structures, residual stress and material heterogeneity related to the manufacturing process should be included in the beam model[144,145]. The shape of many 4D printed soft structures might also evolve dynamically and in this case, the dynamic beam model with the inertial term can be adopted[146,147]. It should be noted that bending is actually different to "folding", which

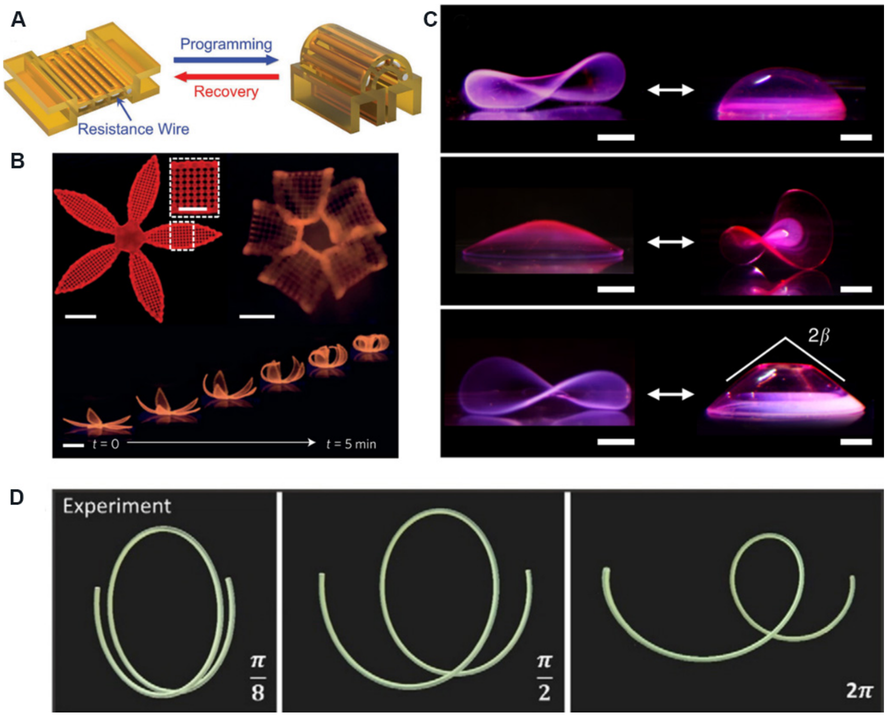

Figure 6. Mechanical designs for 4D shape-changing structures. (A) A 4D printed shape memory hinge that can be repeatedly programmed and automatically deformed upon electric heating[130]. (B) A 4D printed hydrogel flower that gradually closes due to swelling dynamics[133]. (C) 4D printed reversible shape-changing soft structures based on the buckling of a non-Euclidean plate[153]. (D) 4D printed soft actuators with programmable twisting and bending deformation[155].

always creates plastic creases or microscale damage, and two adjacent plates can be perfectly overlapped together[148,149]. However, if the bent region is relatively small compared with the overall dimensions, folding deformation can be approximated by bending[150,151].

The second type of commonly adopted deformation mode is buckling, which comes from the instability of potential energy[152]. Non-Euclidean plates that can deform from a planar configuration to a 3D surface[153,154] [Figure 6C] and twisting ribbons that automatically rotate upon actuation[155] [Figure 6D] are typical examples of 4D buckling designs. Non-developable 3D surfaces can be easily realized in 4D printing through a combination of buckling and computational geometry but the transformation of such surfaces should be further helped by bending designs[156-158]. In the theory of stability, an equilibrium state might not be stable, thus some 4D buckling structures exhibit different deformation patterns before they finally reach the stable state to minimize their potential energies[159,160]. Another common deformation mode is the large-scale motion of flexible mechanisms. In this case, the macroscopic shape changing upon actuation is neither driven by individual bending elements or by local instability but is instead driven by the shifting of the overall energy barrier. For example, Chen *et al.*[161] additive manufactured a shape memory robotic frame that can automatically propel upon heating and a reduction in the energy barrier. Wang *et al.*[150] found that an additive manufactured and stably folded square-twist origami became unstable when the energy barrier was decreased by softening the flat origami panels.

PERSPECTIVES AND CONCLUSIONS

Additive manufacturing has broken the shackles of traditional manufacturing technology and brought new bottom-up manufacturing concepts, thereby offering researchers numerous opportunities to design soft

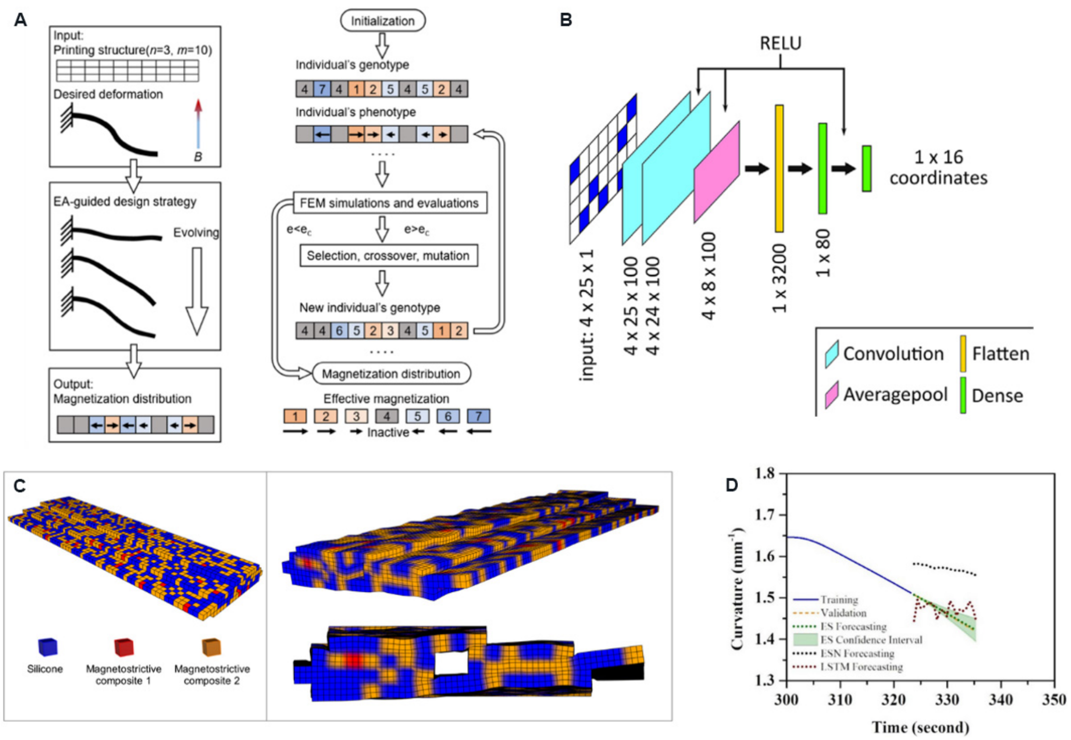

Figure 7. Data-driven inverse design for 4D shape-changing structures. (A) Evolutionary algorithm for 4D shape-morphing beams[138]. (B) Convolutional neural network for 4D shape-morphing beams[141]. (C) 4D shape morphing wing designed by evolutionary algorithm[139]. (D) Machine learning prediction for the dynamic curvatures of SU-8 shape morphing strips[142].

materials with superior structural and functional properties. With state-of-the-art of additive manufacturing, materials that mimic the mechanical properties of biological materials and materials that can actively change shape after been manufactured have been created, which permit applications in biomedical engineering, multifunctional electronics and the aerospace industry. However, there are also some challenges underlying current additive manufacturing techniques. Soft materials are known to easily deform compared to metals and ceramics and shape distortion is therefore commonly observed in the printed parts using various polymer based-printing techniques. Efficient models and simulations must be developed for better understanding these manufacturing process to optimize printing parameters and improve shape accuracy. In addition, the issue of interfacial strength in printed parts with different materials has also aroused concern from researchers. With the rapid development of multi-material 3D printing, it is significantly important to develop corresponding approaches for controlling or even optimizing various material interfaces during the printing process. The examples highlighted here only cover the mechanical designs for additive manufactured materials from limited aspects and are thus far from the entire content of additive manufacturing-based designs. In the future, it can be imagined that designs will be fully integrated with the additive manufacturing process to create more novel materials with superior properties.

DECLARATIONS

Authors' contributions

Designed and wrote the original draft: Zhang Q, Zhao Z

Reviewed and revised the manuscript: Shi Y

Availability of data and materials

Not applicable.

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Conflicts of interest

All authors declared that there are no conflicts of interest.

Ethical approval and consent to participate

Not applicable.

Consent for publication

Not applicable.

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