

4D printing: Technological developments in robotics applications

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Abstract

The idea of four-dimensional (4D) printing is the formation of intricate stimuli-responsive 3D architectures that transform into different forms and shapes upon exposure to environmental stimuli. 4D printing (4DP) of smart/intelligent materials is a promising and novel approach to generate intricate structures for biomedical, food, electronics, textile, and agricultural fields. Nowadays, soft robotics is a growing research field focusing on developing micro/nanoscale 4D-printed robots using intelligent materials. Herein, recent advancements in 4DP of soft robotics, actuators, and grippers are summarized. This review also highlights some recent developments in novel robotics technologies and materials including multi-material printing, electro-, and magneto-active soft materials (MASMs), and metamaterials. It also sheds lights on different modeling mechanisms including numerical models and machine learning (ML) models for fabricating highly precise and efficient micro/macro-scaled robots. The applications of shape-memory polymers (SMPs), hydrogels, and liquid crystal elastomers (LCEs)-based 4D-printed soft and intelligent robots in different engineering fields are highlighted. Lastly, this review incorporates current challenges which are hindering the actual utilization of 4D-printed soft robotics and their possible remedies.

Keywords: 3D printing, 4D printing, smart materials, shape memory polymers, soft robots, actuators

1. Introduction

In the contemporary era, additive manufacturing (AM), or three-dimensional (3D) printing, has drawn a tremendous attraction from engineers and scientists due to its excellent adaptability and ability to print intricate shapes [1]–[3]. This technique incrementally adds different materials including polymers, metals, composites, cermets, ceramics, and metamaterials to develop complex and precise geometries [4]–[6]. Consequently, 3D printing (3DP) technology is extensively applied in a wide range of industries including automotive, aerospace, biomedical, food, construction, and electronics [7]–[10]. According to American Society for Testing and Materials (ASTM) international, fused filament fabrication (FFF)/fused deposition modeling (FDM), inkjet printing (IJP), digital light processing (DLP), stereolithography (SLA), selective laser sintering (SLS), selective laser melting (SLM), direct ink writing (DIW), and continuous liquid interface production (CLIP) are different processes employed for the 3DP of a variety of materials [11]–[14]. Multi-photon lithography or direct laser writing (DLW) is another high-resolution 3DP technology, which is used for the nano-/micro scaled printing of intricate 3D objects [15]–[17]. This approach depends upon the multi-photon

technique using near-infrared femtosecond laser pulses firmly concentrated within the volume of the photoresist, thus, permitting the development of 3D-printed architectures using sub-micrometer resolution [18]–[20].

Despite the significant development in the 3DP field, the production of static solid objects is currently limiting its commercial utilization [21]–[23]. Therefore, novel materials that are programmed to change their properties, functionalities as well as their shapes are developed through newly emerged four-dimensional (4D) printing technology [24]–[26]. In other words, incorporation of life into 3D-printed objects through time dimension helps in developing 4D-printed adaptive and dynamic products [27]–[29]. These dynamic products are made-up of different smart materials including shape memory polymers (SMPs), hydrogels, and liquid crystal elastomers (LCEs) [30]–[33]. Compared to conventional subtractive manufacturing, 4DP technology possess myriad advantages like rapid prototyping, cost-effectiveness, accessibility, lower material consumption, excellent design flexibility and geometric complexity [34]–[36]. The shape transformation of dynamic products is mainly triggered by external energy inputs like ultraviolet (UV) light, heat, pH, or other sources [37]–[39]. Besides one-way change, reversibility is another feature of 4D printing (4DP) technology [40]–[42]. It is mandatory to select appropriate stimuli-responsive materials for developing intricate and reversible 4D-printed architectures [43]–[45]. Additionally, these materials are triggered through internal and external stimuli. External stimuli incorporate light, electric field, acoustic waves, and magnetic field [46]. On the other hand, chemical stimuli including pH and humidity are the internal stimuli [47]–[49]. The stimuli-responsive behavior of smart materials and endless shape possibilities upon exposure to stimuli expands their utilization in smart textile, mechatronics, self-folding food packaging, electronics, automotive, deployable structures, and healthcare systems [50]–[54]. Recently, 4DP technology is extensively applied for developing macro/micro-scaled soft robots for different engineering applications.

1.1.Scope of 4D-printed robots

Even in this modern world, classical manufacturing techniques are still used to fabricate conventional large robots for numerous industrial applications [55]. The wastage of materials and high energy consumption rates are some of the prominent limitations of these techniques. Thus, the world is continuously searching for innovative technologies for the development of soft robotics and other smart devices [56]. 3DP/4DP technology can also be applied to manufacture micro-/nano-scaled robots [57]–[59]. Conventional micro-fabrication processes exhibit certain limitations in terms of geometries, design, and material selection [60]. 4DP technology at a small scale has shown tremendous potential in developing soft robotics and actuators for mechanical engineering, material science, and biomedical engineering fields [61]–[64]. These robots use advanced integrated technology incorporating control systems, smart/intelligent materials, acoustics, and chemistry at micro-/nano levels [65]. This technology incorporates main features such as repeatability, reproducibility, and controllability of 3DP along with supporting technical developments including modeling of novel 3D-structured soft robots [66]–[68]. The primary objective of this review article is to highlight research advancements and current challenges in 4D-printed soft robotics. This will be helpful for developing a variety of robots for a wide range of applications.

Figure 1 shows three different steps involved in the transformation of 3D-printed product into the 4D-printed intelligent object. Smart/intelligent/programmable materials upon exposure to certain stimuli change their shapes, aesthetics, or color through bending, twisting, swelling, and deswelling [69]–[71]. These soft structures developed through smart materials have

tremendous potential for developing sensors, actuators, and other smart devices for biomedical, haptics, adaptive optics, and microfluidics applications [72]. 4DP technology can impart different properties such as in-homogeneous, homogeneous, and functionally-graded as well as develop mono- or multi-material printed architectures [73]. 4DP uses same 3DP processes to print stimuli-responsive materials (SRMs).

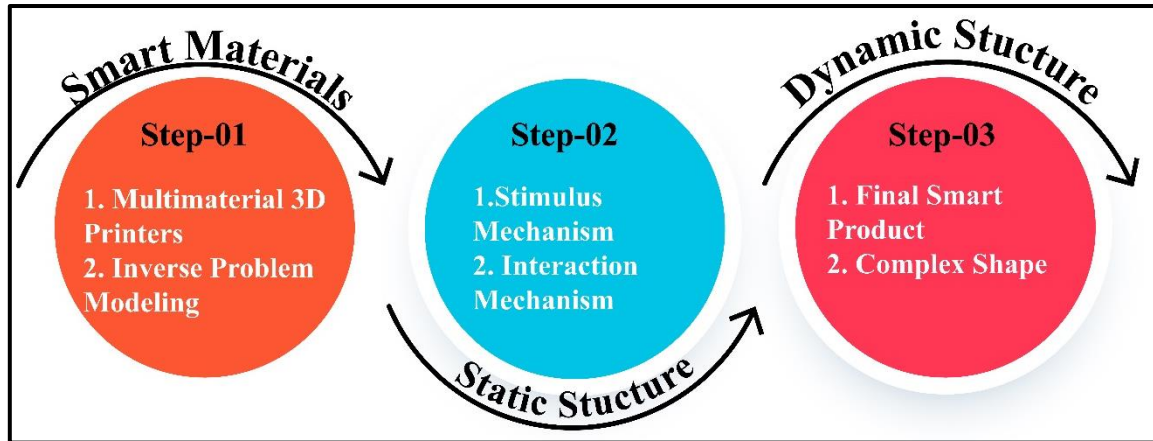


Figure 1. Different important steps in 4DP

2. 4D Printing technologies

4DP is an emerging and technical approach that integrates both SRMs and 3DP technology to develop 4D-printed dynamic architectures [74]. 4DP technology uses the same FDM, DIW, IJP, SLA, SLS, DLP, SLM, and DLW techniques for the printing of SRMs [75]–[77]. However, all these printing techniques are not appropriate for developing soft robots, sensors, and actuators [78].

SLA and DLP use a laser beam and a UV light source, respectively to promote photopolymerization [79]. Both these light-based processes are highly suitable to develop soft actuators due to their extraordinary printing resolution and fast printing speed [80]. During the SLA technique, photochemical processing to cross-link photo-sensitive materials, thus, allowing the development of highly precise printed parts [81]. DLP technique provides excellent printability through an optical mirror to develop patterns [82]. The resolution of these two light-based techniques is lower compared to lithograph processes. For instance, a modified version of SLA known projection micro-stereolithography (PμSL) technique is highly advantageous to fabricate multi-material SMP architectures such as actuators, flowers, and grippers due to its high-resolution [83]. In this technique, a virtual photomask through a projector triggers photopolymerization, which develops multi-material and multi-scale intricate 3D structures [84]. Han et al. [85] proposed SLA-based 4DP for fabricating an emission model as presented in Figure 2(III) for measuring the volatile organic compound emissions from printing. The proposed model also effective for quantifying the shape programming, and shape recovery stages. Results showed that a 61.29 % reduction in emission yield was observed through varying the thermo-temporal conditions with exhibiting the acceptable shape memory performance.

In addition to light-based techniques, FDM/FFF is another vastly applied printing platform, which integrates the state transition of printing polymers by applying heat [86], [87]. Different SMP-based grippers and soft actuators are printed through this technology due to its versatility

and simplistic nature [88]. However, this technique is only limited to the printing of thermoplastic materials such as thermoplastic polyurethane (TPU), acrylonitrile butadiene styrene (ABS), polylactic acid (PLA), vitrimers, and polyamides [89]–[92]. For instance, Choi et al. [93] illustrated the shape memory and reshaping mechanism of engineered vitrimer which was applied as a functional "ink" for FFF 4DP. Figure 2(I) depicts that rolled and twisted shapes were programmed as temporary shapes and restore to their shapes as a result of cooling. The programmed films recovered to the permanent shapes under heating stimulus. Due to the intrinsic cross-linked architecture of the vitrimers, these printed structures have gained significant attention in 4DP applications. Additionally, the availability of this technique for 4DP is low due to high porosity and low compatibility with SRMs [94]. Nowadays, self-coiling and self-folding meta-materials have also been successfully printed through this technology [95].

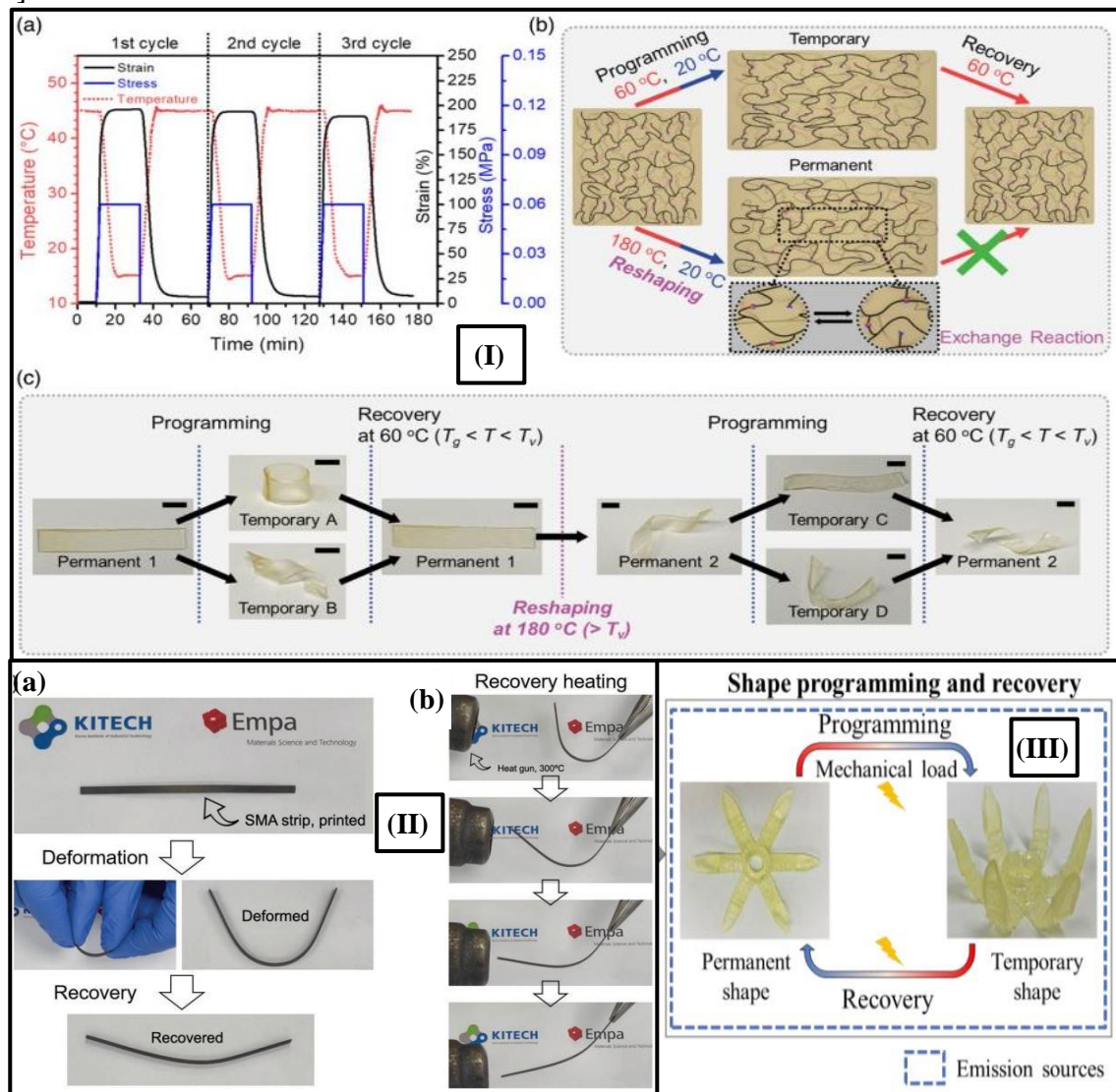


Figure 2. (I) Shape memory mechanism; (a) Consecutive shape memory cycle test; (b) Memory shaping and reshaping illustration; (c) Shape recovery behavior [93] (adapted with permission). (II) (a) Experimental procedures of shape recovery of a Fe-SMA strip in the horizontal direction and (b) Different stages of shape recovery through heating [96] (adapted with permission). (III) Shape programming and shape recovery stages under temperature stimulus [85] (adapted with permission).

IJP process, also known as material jetting (MJ), develops 3D architectures by directly extruding shear-thinning ink materials onto the substrate through micro-scale nozzles to form patterns [97]. The formation of droplets depends upon the physical parameters of inks like surface tension. Therefore, these physical parameters of inks should meet specific requirements [98]. This technique is highly suitable for developing micro-robots, micro-actuators, flexible electronics, optoelectronic devices, and microfluidic setups [99]. Furthermore, IJP offers the printing of less viscous materials which allows the deposition of a variety of materials including dyes, sol-gels, and polymers [100]. Insoluble NPs such as ceramics and metal oxides as well as organic materials like carbon nanotubes (CNTs) and graphene can also be printed through this technique [101]. Low cost and high precision make this technique in the micro-fabrication field [102].

PolyJet printing technique uses direct jetting of photopolymer ink droplets and subsequently solidified onto a build platform [103]. This technique is extensively applied to fabricate smart materials [104]. To date, 4D-printed structures developed by using polyjet printing are actuated through heat [105]. This technology has an added advantage to print multi-materials due to simultaneous printing with multi-printheads [106].

Similar to the FDM technique, DIW uses a variety of viscoelastic ink materials like hydrogels, thermoplastics, polyelectrolytes, and sol-gel oxides to print 3D structures upon high pressure [107]. Recently, DIW has emerged as a suitable and robust technique to develop a variety of soft robots, actuators, and other smart products [108]. Two-photon polymerization (2PP) also known as DLW, is widely applied for developing micro-scaled highly intricate, and precise 3D-printed robots [109]. The use of femtosecond pulsed laser develops architectures of sub-micrometer resolution [110]. This technique is normally used for the micro-printing of LCE and hydrogels, and other composites [111]. Kim et al. [96] developed complex 3D structures of Fe-based SMA through a 3DP using laser powder bed fusion technique. The 3D printed Fe-SMA hold much higher mechanical properties than SMP. The printed Fe-SMAs exhibited various material-inherent functional behaviors, self-healing and shape changing behavior (referring to Figure 2(II)).

Conventional composite manufacturing approaches including resin transfer molding, filament winding, pultrusion, autoclave, and prepreg are extensively applied for the development of polymer composites [112]–[114]. However, these conventional processes require mold for the development of highly precise and accurate polymeric composite-based products [115]–[117]. Additionally, the process parameters of these manufacturing approaches are difficult to control [118]. Nowadays, moldless composite manufacturing is usually done through 4DP technology and is called 4D printing of composites (4DPCs) [119]. These 4D-printed composites are usually hard materials due to the incorporation of long and continuous fibers into soft polymer resins [120]. These composites offer excellent mechanical characteristics, intricate 3D structures, and excellent design flexibility [121]. Different 4D-printed fiber-reinforced composites such as twisted structures, leaf springs, actuators, stiffeners, and flexible airplane wings are developed through moldless composite manufacturing [122]. In addition to this, different additives/nanoparticles (NPs) in composites can be easily embedded using this technique [123]. Furthermore, soft robots and actuators can also be developed through multi-material printing.

2.1 Multi-material printing

4DP technology is not limited to develop intricate architectures of single materials [124]. SMP- or hydrogel-based multi-material 4DP approach is widely employed for the development of actuators and soft robotics applications due to its excellent shape-morphing effect [125]. This approach uses a combination of different materials such as hydrogels and SMPs, to develop highly complex structures comprised of soft areas of functional materials as well as hard areas to develop the skeleton of the robot [126]. However, it is difficult to print multi-materials, and only a few printers ProJet MJP series by 3D systems and PolyJet Connex series by Stratasy are commercially available for printing these multi-materials [127]. These printers use photopolymer resins for the fabrication of intelligent architectures. DIW is another technique that is applied for the developing smart actuators by using multi-materials [128]. For instance, López-Valdeolivas et al. [129] developed LCE/polydimethylsiloxane (PDMS)-based soft actuator through the DIW technique. The authors observed excellent actuating properties upon thermal stimulus, as illustrated in Figure 3(a). Additionally, SMP and conducting polymer-based multi-material printing exhibit excellent features as well as enhance functionality to develop crawling robots or smart grippers [130].

Likewise, Wang et al. [131] fabricated SMP-based multi-material programmable objects containing gold nanoparticles (Au-NPs). The activation and shape morphing behavior of 3D objects was due to the presence of Au-NPs in acrylate-based printing which produced SMP with tunable transition temperatures. A low-cost LED light at a specific wavelength was used which demonstrated shape morphing behavior. The heat from the LED light triggers the shape transition of objects due to temperature exceeding above glass transition temperature (T_g) of SMP. Figure 3(b) depicts excellent shape fixity and recovery ratios of 95 % were observed. Thus, the proposed approach fabricates programmable light-activated 4D-printed objects capable of dual transition with tuning the concentration.

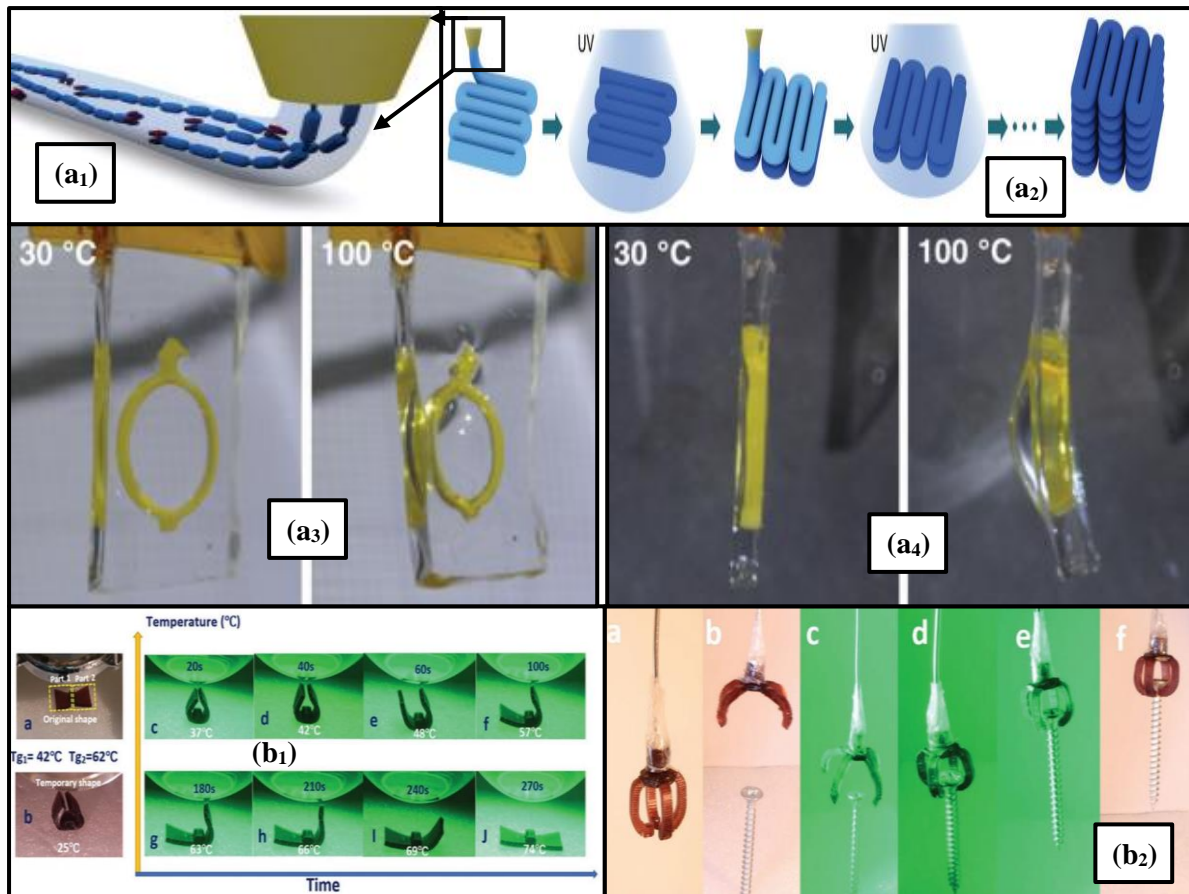


Figure 3 (a₁, a₂) A schematic representation of DIW technique; 4DP of PDMS/LCEs-based soft actuator which exhibited decrease in length upon exposure to temperature stimulus; (a₃) Oblique view; (a₄) Lateral view; (Adapted with permission from ref. [129] Copyright 2018, Wiley-VCH GmbH); (b₁) Shape-memory behavior of different objects under light stimulus at different temperatures and time; It shows flower shape object with its blooming and temporary shape of de-blooming; (b₂) Multi-material printed gripper under light stimulus effectively used as a soft robotic hand for grabbing screw like objects (Adapted with permission from ref. [131]. Copyright 2021, Wiley-VCH GmbH)

3. Programmable shape memory materials for robotics

In 4DP, smart materials incorporating different NPs/additives are loaded to develop 3D-printed objects [132]. These 3D-printed objects are transformed into programmed 4D-printed states if materials are programmed during the printing process and these materials exhibit the ability to undergo controlled time-dependent change [133]. Sometimes, the printed objects remain in unprogrammed states and are manually programmed after the printing process [134].

The shape memory effect (SME) is triggered through different stimuli including photo-responsive, thermo-responsive, pH-responsive, moisture-responsive, electro-responsive, magneto-responsive, and multi-responsive [135]–[138]. Materials that exhibit SME are known as shape memory materials (SMMs) [139]. SMMs are the next generation of smart materials for developing robots through 4DP technology [140]. Based on the morphological transformations, these materials are divided into one-way SMMs and two-way SMMs, as illustrated in Figure 4(a). One-way SMMs are programmable materials, which transform their shape in response to certain stimuli and remain even after the removal of stimulus [141]. For instance, Kumar et al. [142] investigated the influence of the one-way programming of the recycled polyvinylidene fluoride (PVDF) and polyurethane (PU)-based composites under a chemical stimulus. The results of the developed composites demonstrated that acceptable one-

way programming-based 4D properties were observed for self-healing applications such as repair of non-structural cracks in heritage sites.

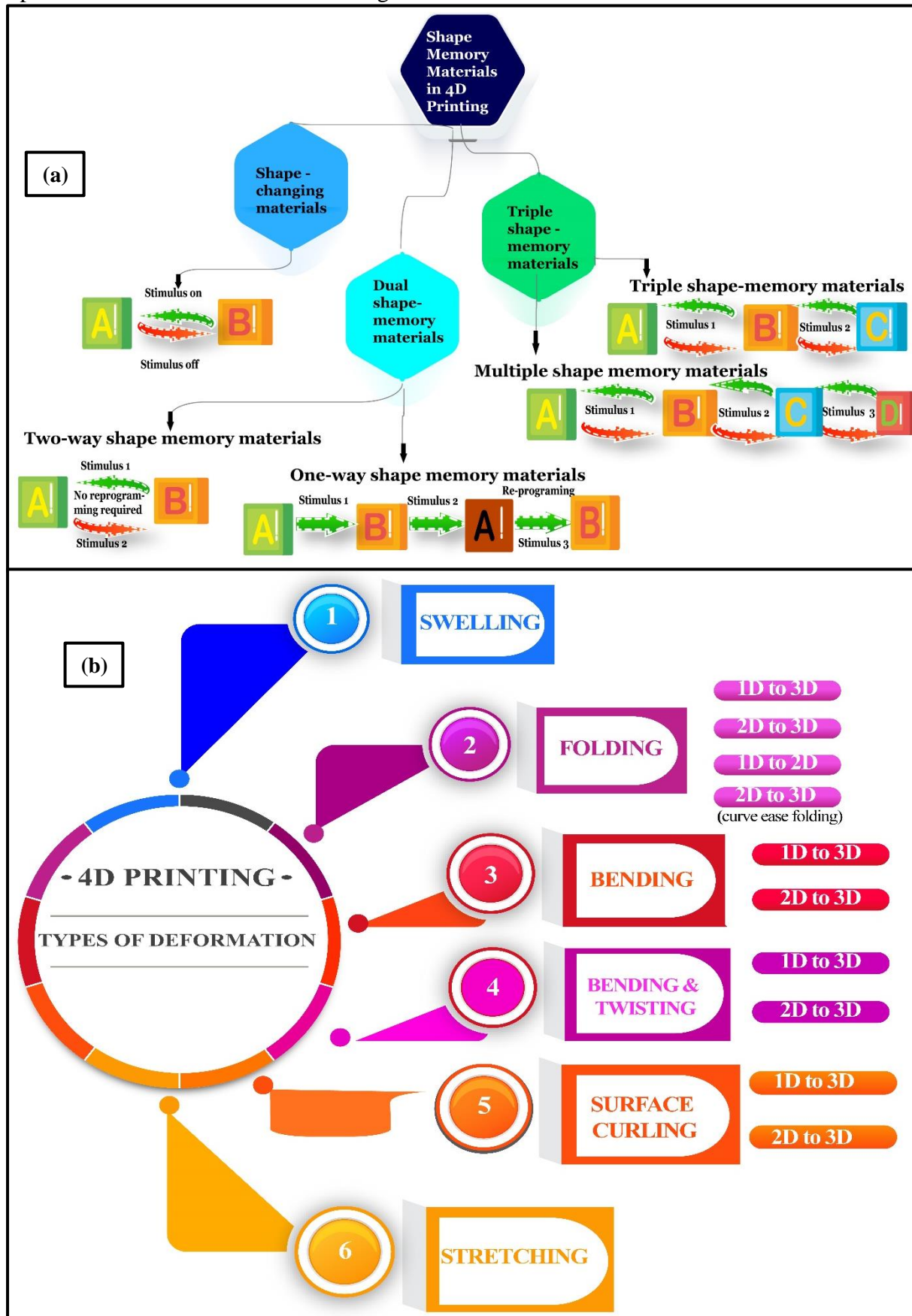


Figure 4. (a) Difference types of SMMs in 4DP (Adapted with permission from ref. [23] Copyright 2022, Wiley-VCH GmbH); (b) Illustration of deformation in 4DP.

Two-way SMMs reversibly morph between two shapes through the switching of stimulus, thus, developing an intricate biomorph mechanism [143]. These materials exhibit shape memory effects (SME) at high (heating) as well as at low temperatures (cooling) [144]. The programmed 4D-printed product is exposed to specific stimuli to achieve desired shape change. This object changes its function and remains in the new configuration permanently [145]. In an irreversible system, the printed object can be transformed into another desired shape through reprogramming [146]. Whereas, the printed objects adapt to temporary shapes and reverse to their native shapes upon restimulation through different stimuli such as cold temperature in a reversible system [147]. Additionally, these materials also track their transformation paths during this whole process [148].

SMMs are categorized into SMPs, shape memory composites (SMCs), LCEs, shape memory alloys (SMAs), shape memory hybrids (SMHs), and magnetoactive soft material (MASMs) [149]–[152]. Intelligent materials, especially SMP-based composites and hydrogels, are most commonly used for sensors, actuators, and soft robotics applications [153] and normally incorporate thermo-mechanical programming techniques after printing [154]. These techniques require partial melting at the intermediate melting temperature under actuation [155]. Additionally, the thermodynamically preferred configuration of polymer-network melts and dismantles the crystalline structure of polymers [156]. The degree of reversibility depends upon the intermediate temperature as well as the cross-link density of polymers [157].

Extortionate researchers have developed SMP-based soft robotics by using specific stimuli. For instance, Keneth et al. [125] developed various 3D printable SMP inks with controlled transition and melting temperatures. The difference in melting and transition temperatures were produced by tuning the ratio between the monomers and the diluents. The authors fabricated a 3D box with two lids that open and close at two similar, but well-distinguished temperatures due to two different stimuli including light irradiation and direct heating, as depicted in Figure 5(I). The proposed research has the potential to use in soft robotics and drug release applications. In soft robotics, remote actuation is accomplished through different modes of movement at two different temperatures, and in the case of controlled drug release, a box, which can simulate a valve or a pill. Likewise, Jeong et al. [158] demonstrated remote actuation of SMP under color-dependent selective light absorption and heating stimulus. The results showed that multistep actuation was operated by the color of light and duration of illumination on the hinged structures, as illustrated in Figure 5(II). Furthermore, 4DP can allow the fabrication of complex and multicolor geometries for tailored responses. Thus, multicolor 4DP of SMP-based composites possessed unique abilities for light-induced structural changes and remote actuation.

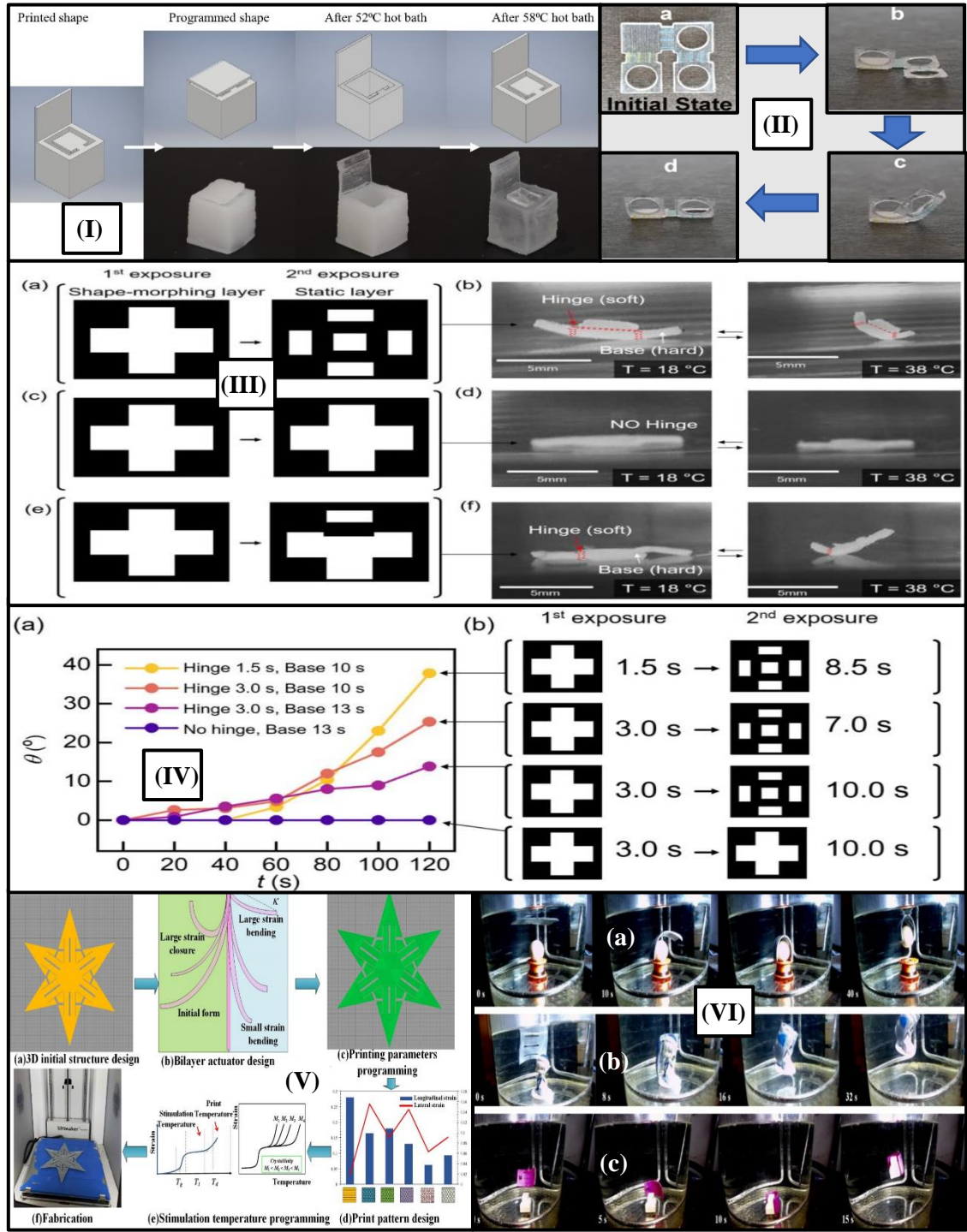


Figure 5. (I) Shape morphing behavior of 3D-printed box with two lids. The box opens and closes at 52 °C and 58 °C, respectively [125] (adapted with permission); (II) 4D-fabricated multicolor hinged structures under multistep actuation depending on the color of light and duration of illumination [158] (adapted with permission); (III) Shape-morphing behavior of four hinges object under different temperature; (IV) Optimization of the shape-morphing behavior [159] (adapted with permission); (V) Proposed methodology for producing bio inspired structures with programmable actuators; (a) Initial structure 3D design; (b) Actuator bilayer-based design; (c-e) Printing parameters programming and actual design making model; (VI) (a) Behavior of shell gripper of egg with time; (b) Irregularly surfaced model shell gripper response; (c) T-shaped structure model shell gripper response [160] (adapted with permission)

The selection of appropriate active materials is important to developing 4D-printed soft robots [161]. These active materials are SMMs which are 3D-printed through computer-aided design (CAD) files, which provide path instructions [162]. The incorporation of SMMs in the printing process resulted in the transition from 3DP to 4DP with the integration of robotics [163]. 4DP is possible only due to the unique features of these materials, including shape restoration or one-way time dependent deformation upon exposure to external or internal stimuli [131], [164], [165]. These materials inherently memorize and retain their programmed shape in response to specific stimuli. These chemical-, physical-, biological-, or a combination of multiple stimuli-based external and internal triggers develop plastic deformation [166]–[168].

Thermal stimulus-based SMMs are extensively applied in a wide range of engineering applications [169]. For instance, Aberoumand et al. [170] investigated different programming conditions such as loading temperature and holding time of polyethylene terephthalate glycol (PETG) and evaluated their effects on the shape memory performance. The results showed that lowering the printing temperatures and increasing the printing speed improved the self-bending and shape recovery performances. Furthermore, at a programming temperature of 75°C, the stress recovery ratio decreased by 47 % due to the increase in load holding time, and programmed samples exhibited a weak shape memory performance at 90°C.

The single-layer controllable response under external stimuli is quite challenging in 4DP. To overcome this problem, Lee et al. [159] demonstrated the programmed shape morphing of a single-layer through 4DP by patterning both the static and shape-morphing layers of four hinged structures. Shape-morphing layer and static layer were formulated for this purpose. A short-time (<3 s) illumination was set for the shape morphing layer. Likewise, for the static layer longer-time (>3 s) illumination was set under UV light, as demonstrated in Figure 5(III). Additionally, the behavior of folding angle with UV light exposure times was also observed for optimizing the shape morphing performance. The results showed the highest folding angles were achieved for the shape morphing layer and static layer at 1.5 s and 10 s upon exposure to UV light, respectively, as presented in Figure 5(IV).

The existence of coupling of multi-directional strain in actuator layout makes the deformation highly complex and unpredictable. To overcome this problem, Zheng et al. [160] designed a general unit and an actuator unit, which cause the transformation of closed-shell bioinspired structure. The complete layout of this study is depicted in Figure 5(V). The authors eliminated the transformation produced by the uncontrolled shape memory behavior of the general unit by introducing the shape mutual stress confrontation between the actuator and the general unit in the layout thermodynamic model. Finally, the adapted technique was adopted on a complex shell-like gripper structure, as illustrated in Figure 5(VI).

Magneto-active soft materials (MASMs) incorporating hard magnetic particles including superparamagnetic iron NPs, ferrites, or neodymium particles, are shaped-programmable materials, which are manipulated robustly, reversibly, and remotely under a magnetic field without electrical or pneumatic tethers [171]–[174]. MASMs are usually 4D-printed by using highly viscous inks through the DIW technique [175]. The magnetic actuation is harmlessly and easily perforated through different materials, which makes these materials highly appropriate for soft robotics, sensing, and actuating applications [176]. For instance, Zhu et al. [177] developed MASM-based 3D architectures through the DIW technique by incorporating iron NPs into the PDMS matrix. The developed composite ink exhibited low magnetic coercivity and can be applied to remotely develop tunable 3D terahertz photonic crystal with a rapid response rate. In another study, Wang et al. [178] studied the shape memory behavior of

soft magnetic composites fabricated through the electrohydrodynamic printing (EHDP) technique under the magnetic stimulus, as depicted in Figure 6(I). In this study, different soft prototype actuators had been designed and tested under different magnetization orientations and profiles including magnetically driven electrical switches, inchworm shaped-based deformable actuators for bionic soft robot applications, and dragonflies, as elaborated in Figure 6(II). Due to their fast response and untethered control, these deformable actuators are significant for harmless human-machine interaction-based applications.

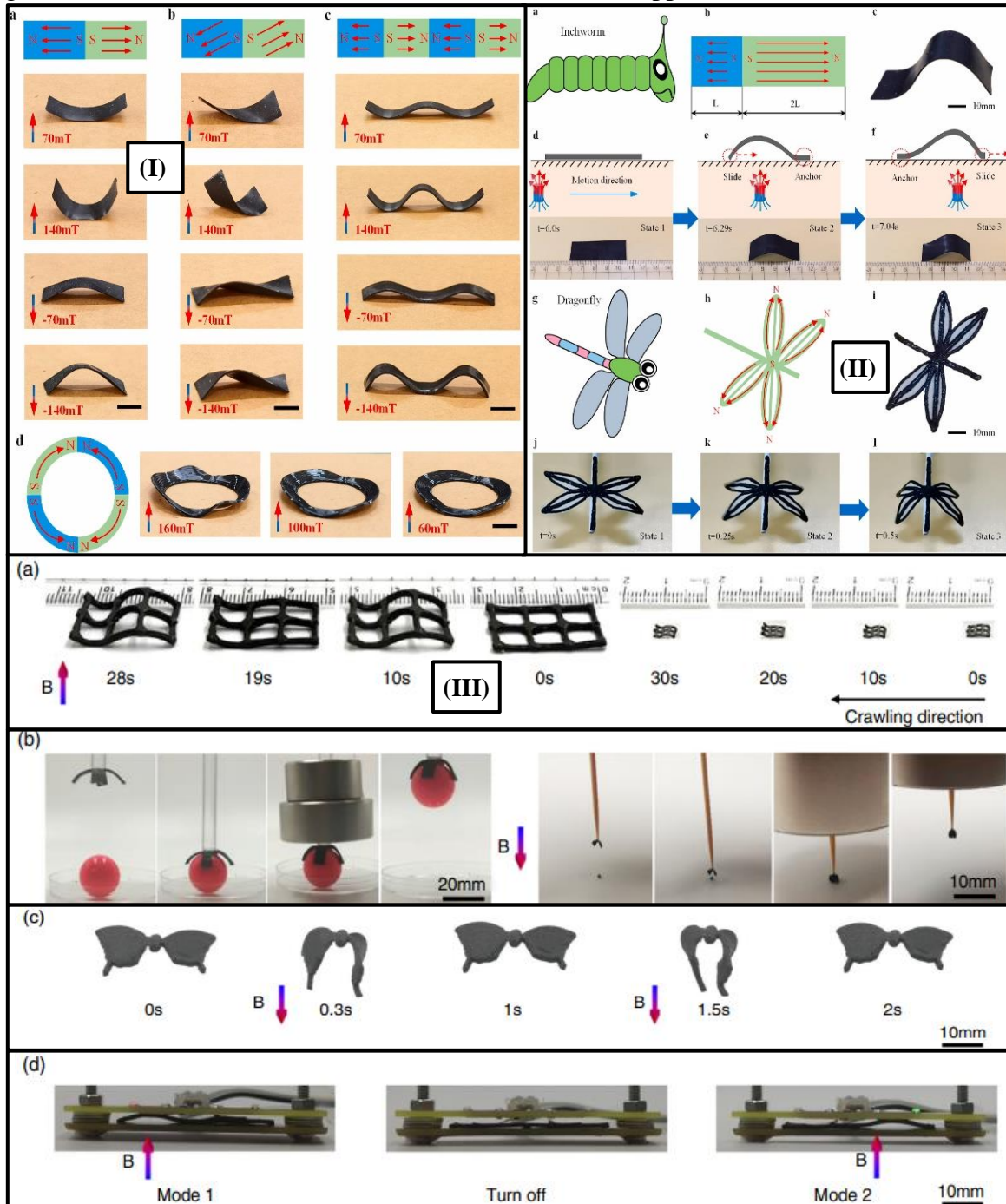


Figure 6. (I) Shape shifting behavior of soft magnetic actuators under different external magnetic fields and along with their magnetization profiles; (II) Different bionic soft robots (a-f) The inchworm-based robot prototype and their response under the magnetic actuation; (g-j) The dragonfly-based robot prototype and their response under the magnetic actuation [178] (adapted with permission); (III)

Demonstrations of MASMs; (a) Soft crawling robots; (b) Flexible actuator/gripper; (c) Bionic butterfly; (d) A multi-state magnetic switch [108] (adapted with permission)

Magneto-responsive characteristics of these intelligent materials originate from structural geometry and magnetic anisotropy [179]. However, it is a challenging task to develop micro-scaled materials by using programmable magnetic anisotropy [180]. Zhu et al. [108] developed a novel mechanically-guided 4DP technique to program magnetic anisotropy in intricate 3D small architectures, as illustrated in Figure 6(III). These micro-scaled soft materials can be applied to develop flexible grippers, soft robots, multiscale magnetic switches, and bionic butterflies.

Sometimes, 4DP technology is not a suitable option for developing the clever design of shape morphing structures [181]. Therefore, magneto and electro-active intelligent shape-morphing structures are constructed by combining 4DP with conventional techniques for achieving the successful design of soft robots [182]. SRM-based 4D-printed architectures are highly advantageous due to their time-dependent characteristics, shape memory effect (SME), and multi-functionality [183]. These materials induce shape change as well as properties [184]. In addition to this, 4DP is a single-step fabrication procedure, which has the ability to generate robots and actuators for different engineering and biomedical applications [185].

The electric field is extensively applied for producing actuation in 3D-printed smart systems [186]. For this purpose, electro-sensitive nanocomposites and conductive hydrogels are the most suitable materials for sensor design, soft robotics, and tissue engineering applications [187]. Electro-sensitive nanocomposites are printed by incorporating different conductive additives like carbon black, graphene, and CNT into polymeric resins [188]–[190]. The uniform distribution of these additives in the polymer matrix is essential to maintain homogeneity in the electrical properties of nano-composites [191]. For instance, Dong et al. [192] developed electroactive PLA/CNT-based composites for different smart devices with remote control capabilities through the FDM technique. The authors investigated shape memory mechanisms of different 2D and 3D printed SMC-based complex structures under the electric stimulus, as illustrated in Figure 7(I). The results showed that the thermal conductivity, electrical conductivity, and shape recovery ratio of SMC were increased with the incorporation of CNT content within a certain limit. High-performance SMAs exhibit excellent robotic functions like anchoring and bending motions [193]. However, most Ni- and Al-based SMAs lack biocompatibility [194]. On the other hand, SMP materials are light, flexible, and biocompatible and SMC-based soft robots/actuators can be developed through the integration of SMAs and SMPs [195]. For instance, Pyo et al. [196] fabricated a 4D-printed actuator using reversible SMC developed through SMA and SMP-based materials. The study showed that SMA demonstrated SME as a result of phase change between martensite and austenite phases, under temperature change and SMP showed SME due to changes in the proportions of hard and soft segments near the T_g , as depicted in Figure 7(III). In this study, Nylon 12 was used as 3DP material in filament form. Thus, SMC actuator properties can be improved by controlling the volume ratio and cooling time and have shown tremendous potential for stents and control valve applications.

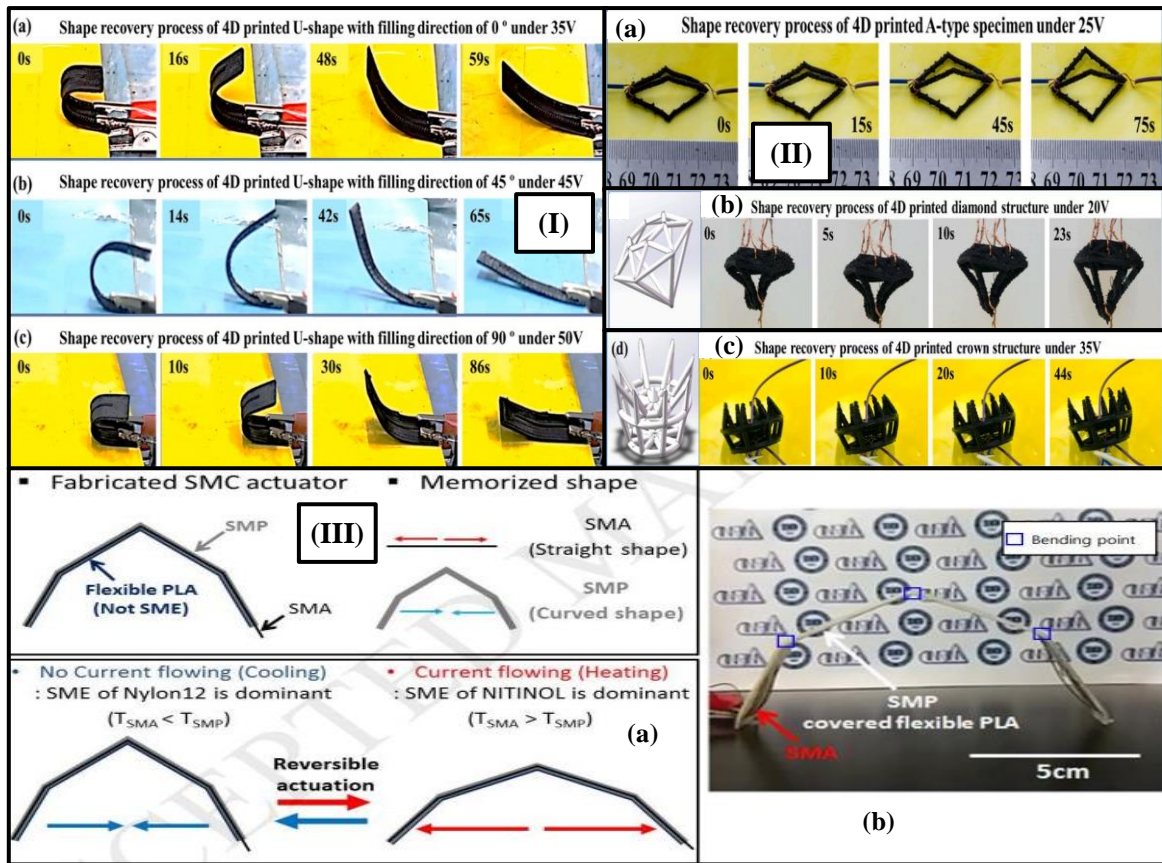


Figure 7. (I) Electro-active shape recovery behavior of U-shaped structures at different infill directions; (a) 0°, (b) 45°, (c) 90°; (II) Electroactive shape recovery behavior of the different types of specimens; (a) A-type specimen, (b) diamond-shaped structure, (c) crown-shaped structure [192] (adapted with permission); (III) (a) Actuation mechanism of SMA and SMP; (b) 4D fabricated SMC actuator mode [196] (adapted with permission)

Table 1 presents the shape-changing behavior of most recent research results of 4D-printed programmable composite materials. These actuating products possess different motions like bending, folding, swelling, rolling, stretching, and twisting.

Table 1. Summary of latest research results on 4DP composite materials in soft robotics along with their specifications

AM process	Dynamic material	Stimulation method	Printed objects	Shape change deformation/Programming	Ref.
DIW	Alginate/MC/ PAA	Magnetic	Actuator	Rolling / Jumping / bending	[134]
FDM	PETG	Heat	Multilayer samples for mechanical testing	Bending	[170]
FDM	PLA	Temperature	Bilayer structures / aka hinges	Bending/twisting	[197]
FDM	TPU/PLA	Temperature	Laminate structures, butterfly and flower models	Bending	[198]
-	Collagen fibers	Heat	LAA occlusion device	Sequential shape change	[199]
FFF	CF/PLA	Temperature	Horseshoe lattice structures	Uniform patterns / compression	[200]
DLW	PEGDA/EEC	Humidity	3D cross-shape pattern	Swelling	[201]

FDM	SMP	Heat	Meta-surface structure	Uniform patterns	[202]
Extrusion	EC	Humidity	3D strips	Swelling	[203]
FFF/DIW/SLA/DLP	Silicones	Magnetic field	3D surface patterns	Uniform patterns	[204]
EHDP	PDMS	Electric field	Grasping device	Stretching	[205]
DLW	PEGDA/EEC	Hydrothermal	3D strip-shaped object	Swelling	[206]
-	Hydrogel	Humidity	3D star-shape structures	Swelling	[207]
SLS	Diels–Alder bond (PUDA)/CNT composites	Light	Miura origami structures	Sequential shape change	[208]
-	PLA	Temperature	Bilayer structures	Folding	[209]
Extrusion	PNIPAM hydrogel	Temperature	Capsules	Sequential shape change	[210]
FDM	PLA/TPU/CNT	Temperature	3D wave structures samples	Length changing	[211]
Extrusion	Alginate/MC hydrogel	CaCl ₂ and deionized water solution	3D structures (tube, helix and flower-based shapes)	Swelling	[212]
Extrusion	Polyester/CNC composite	Heat	Tree-like design model	Bending	[213]
Polyjet Printing	Ag-NP paste	Heat	3D strips	Stretching	[214]
DLP	WPUA/acrylamide/AA/NaCl	Ethanol/water solution	Soft lattice structures	Stretching and compression	[215]
Extrusion	ABS	Heat	3D annular frames	Non-uniform	[216]
FFF	Wood/PLA composites	Heat	3D structures	Bending and folding	[217]
DIW	PTMPAC/graphite	Temperature	Scaffold	Bending and folding	[218]
FDM	-	Temperature	Bilayer structures	Bending	[219]
2PP lithography	HPPA/BPA/TPO	Temperature	3D grid structures	Compression	[220]

4. Design, modeling and simulation of 4D-printed smart materials

The modeling of 4D-printed soft materials is another milestone achieved by the researchers due to its robustness and conceptual phase designing [221]. This section illustrates different types of modeling mechanisms adopted by the researchers for smart materials.

4.1 Numerical modeling of 4D-printed smart materials

In the contemporary world, intelligent materials have spellbound the world and are widely applied for printing soft robots and actuators for myriad engineering applications [222]. The development of constitutive modeling is essential to evaluate the stress recovery and strain recovery performance of soft materials [223]–[225]. These constitutive models are employed as numerical tools for different engineering applications. Constitutive models for smart materials are mostly developed through phase transition modeling and thermo-viscoelastic modeling [226]. These models are highly suitable for predicting the properties of multi-way smart materials [227].

Smart/intelligent materials are integral constituents of 4DP and the primary source of 3D-printed object functionalities. These functionalities depend upon the spatial distribution of intelligent materials or a combination of conventional and intelligent materials [228]. Design distribution requires computational modeling and topology optimization for obtaining programmable 4D-printed objects. Design optimization of 4D-printed structures through topology also been a great interest [229]. Both modeling and topology optimization approaches demonstrated several variations such that multiple targeted displacement optimization results in the form of bending and twisting transformation [230]. This approach was successfully adapted by Pakvis et al. [231]. These authors applied modeling and topology optimization

techniques on bilayer polymer structures with different layer heights and later validated these structures through experimental results. The results as depicted in Figure 8(Ia) showed that the numerical data had a good agreement with experimental results including different environmental factors such as gravitational effect for uniform bilayer structures with layer sizes of 0.5 and 1 mm.

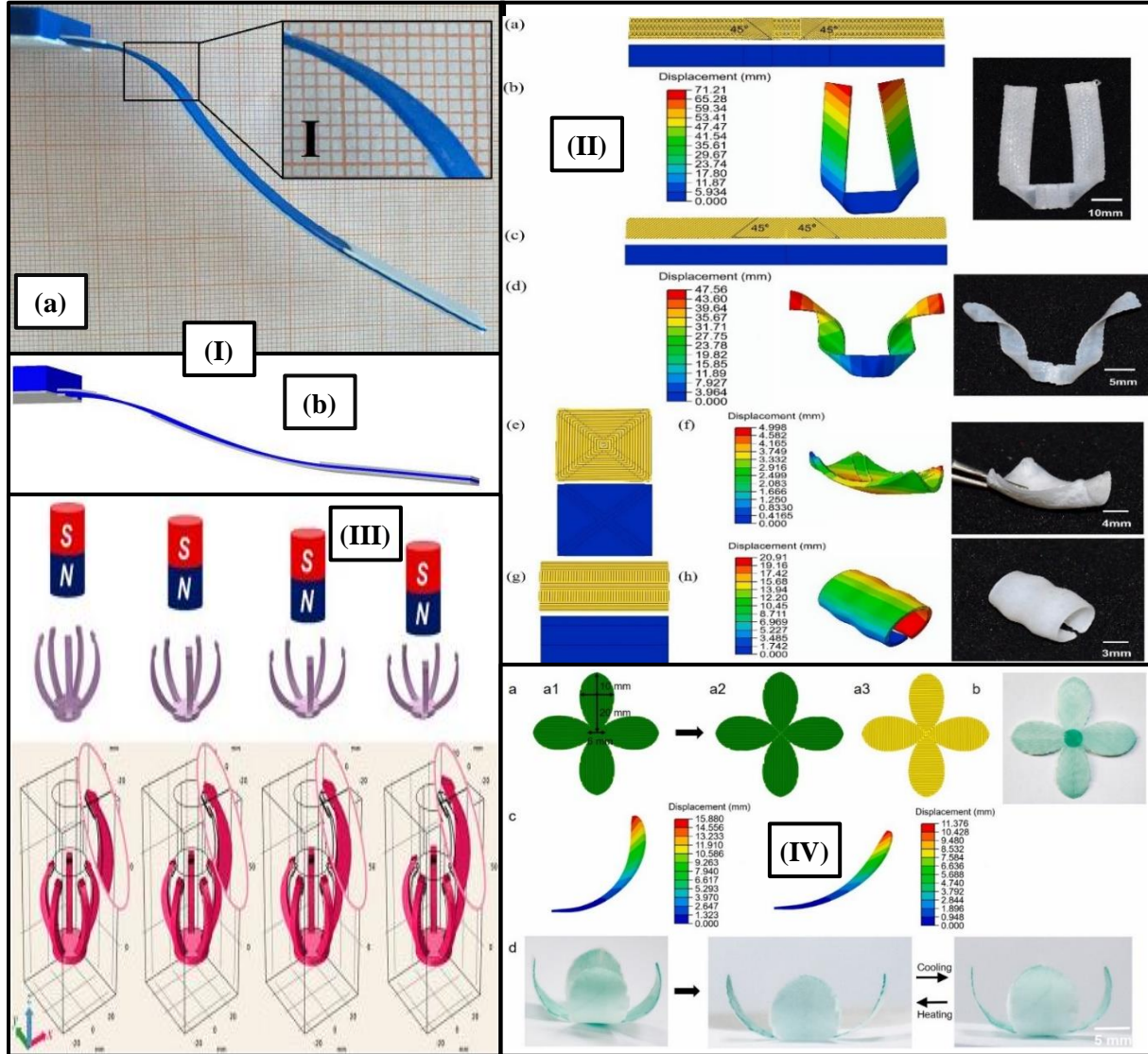


Figure 8. (I) Comparison of sample deformation; (a) Experimental; (b) Simulation (including gravitational effects) [231] (Adapted with permission); (II) Different experimental and simulation deformations of the 4D-printed models with the composite printing paths; (a-b) Shape of the letter “U”; (c-h) Spiral shape, wave-edge bowl, and tubular shape along with a spacer pattern [232] (adapted with permission); (III) (a) Schematic and numerical simulated models of TPU/Nd₂Fe₁₄B-based composites along with the deformation process. The gripper with a magnetic particle content of 40 wt.% as an external cylindrical magnet approached and moved away under the repulsive magnetic force generated by the external magnetic field [233] (Adapted with permission); (IV) Numerical simulation study on PLA/TPU laminates; (a) Smart flower shape different reversible unidirectional bending deformation; (b) Actual 3D-printed flower model through FDM; (c) Simulation-based programming results of reversible flower model for original and expanded shapes; (d) Experiment deformation results after reversible deformation recovery reversible flower model [198] (adapted with permission)

In another study, FDM-printed monolayer SMP models were numerically developed through ABAQUS software [232]. The numerical simulating tool used the constitutive function of SMPs to describe their thermo-mechanical parameters and later validated them for various deformation processes in pre-programmed architectures. External temperatures were used in stimulating different models for their deformation and also configured the anisotropic pre-strain stored in the printed model. The results showed the deformation of the SMP models was consistent with the simulation results, as demonstrated in Figure 8(II). Additionally, the simulation technique successfully predicted deformation angle and direction under thermal stimulation. Similarly, Wang et al. [198] studied the mathematical model of reversibly deformable structures derived from PLA/TPU and experimentally fabricated through FDM. The authors analyzed the flexibility of both combinations of materials and their requirements. For characterizing the thermal viscoelastic response, a one-dimensional (1D) SMP constitutive model as demonstrated by Tobushi was used and modified for the 3D constitutive model. All simulation results of the composite laminate structures are consistent with the experimental deformation results for different flower shape models, as depicted in Figure 8(IV).

Wu et al. [233] produced magnetically driven grippers, by SLS processing of TPU- and magnetic $\text{Nd}_2\text{Fe}_{14}\text{B}$ -powdered materials. The actuation mechanism was systematically investigated through both experiments and numerical simulations using COMSOL Multiphysics® software. The magnetic powder was assumed to be distributed uniformly in the gripper shape during simulation. Horizontal and vertical magnetizations were used for creating the magnetic induction intensity and scalar magnetic potential distribution around the grippers. Both experimental and simulation studies revealed that magnetic force increased with the increase in magnetic particle content and with the decrease in the distance, as illustrated in Figure 8(III).

Constitutive models of smart materials under the thermo-viscoelastic approach consider inherent characteristics including molecular chain motions, cross-links, relaxation time, intermolecular chain interactions, and movement of contact surfaces [234]–[236]. These models accurately illustrate the shape memory behavior of smart materials [237]. For instance, Zeng et al. [238] applied a novel micromechanics-based thermo-viscoelastic constitutive model on 4D-printed SMCs. In this micromechanics framework, an equivalent viscoelastic stiffness tensor of the composite was obtained through energy-based effective strain theory and the Mori-Tanaka homogenization scheme, as presented in Figure 9(I). Later, the proposed scheme for the 3D viscoelastic constitutive model of PLA-based SMPs was implemented through ABAQUS software. Stress relaxation tests were performed to govern the model parameters at different temperatures. Different experimental tests related to SMCs including uniaxial tension, stress relaxation, and shape memory cycling under the effect of several fiber layup angles were performed and compared with simulations results, as depicted in Figure 9(III). The simulation results effectively predicted the uniaxial tensile curves, stress relaxation phenomena and shape memory behavior of SMCs, as illustrated in Figure 9(II) and Figure 9(IV).

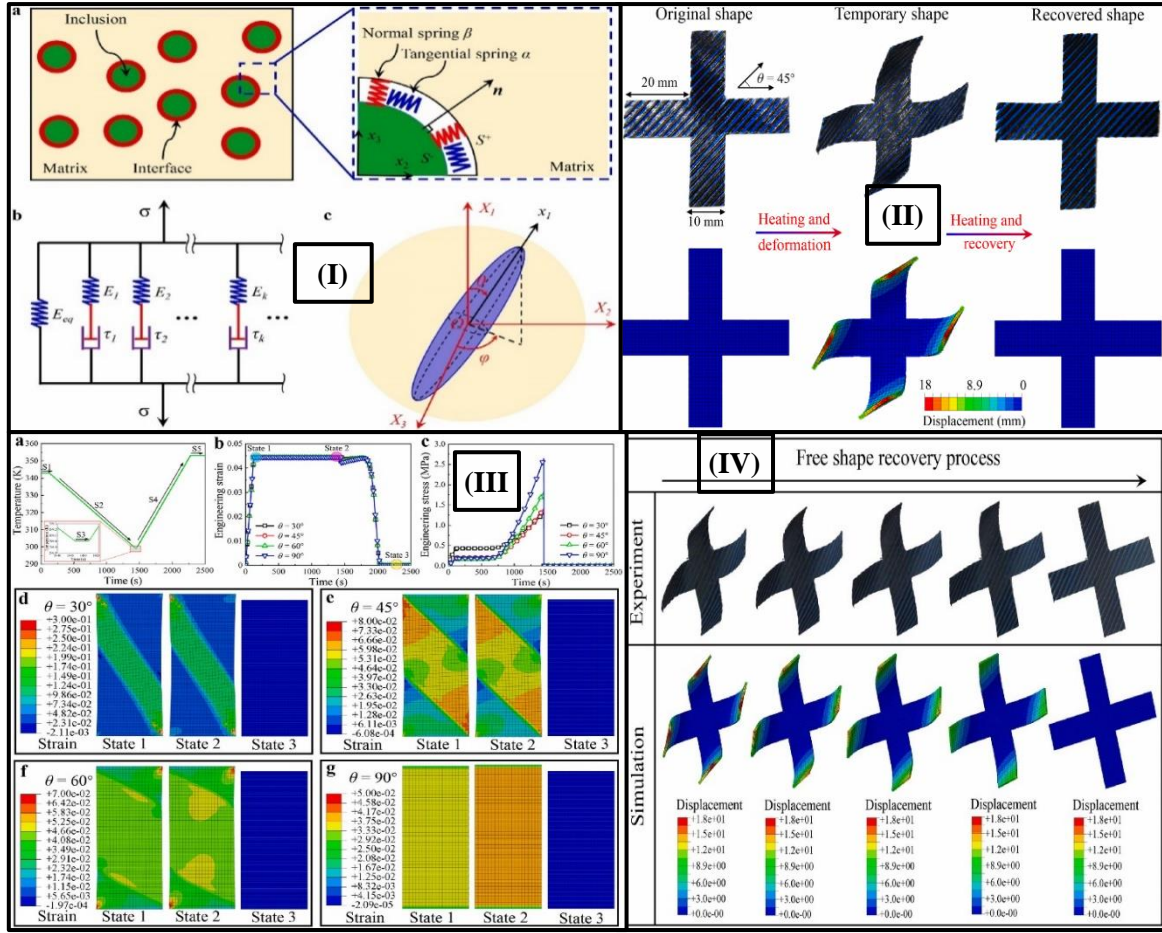


Figure 9. (I) Micromechanical equivalent model, schematic diagram for equivalent spring model and viscoelastic model of SMP matrix; (II) The shape memory behavior of 4D-printed cross-shaped SMC as demonstrated by both experiment and theoretical simulation studies during the shape memory cycle; (III) Simulation results of SMCs with different fiber lay-up angles; (IV) Free shape recovery behavior of cross-shaped SMC member as demonstrated by experimental and simulation studies [238] (adapted with permission).

Metamaterials are novel engineered materials, which are manufactured and designed with functions and physical properties that do not exist in natural materials [239]. The unit structures of these materials are properly designed and assembled to customize physical characteristics and functions, including acoustics, optical, mechanical, electromagnetic as well as thermal properties [240]. Recently, metamaterials with extraordinary mechanical characteristics and functions are printed through 4DP technology [241], [242]. Additionally, metamaterials are developed through stacking origami unit structures with distinct characteristics including large deformation, negative Poisson's ratio, lightweight, adjustable thermal expansion, and programmable stiffness [243]–[245]. These origami-based metamaterials exhibits unlimited potential in developing intelligent soft robots and flexible electronics [246]–[248]. Extortionate researchers have evaluated the energy absorption capability of metamaterials [244] by using finite element analysis (FEA) [249]. For instance, Namvar et al. [250] investigated different numerical models, including re-entrant auxetic, hexagonal, and AuxHex unit cells for characterizing the energy absorption, reversible shape memory properties, and structural mechanical behavior of PLA-based 4D-printed lattice metamaterials by using ABAQUS software, as depicted in Figure 10(I). The mechanical behavior of metamaterials was studied under quasi-static compression loading and the results were compared with the experimental

study. The simulation results shown in Figure 10(I) revealed that metamaterial with re-entrant auxetic unit-cells demonstrated superior energy absorption capacity compared to other cell topologies.

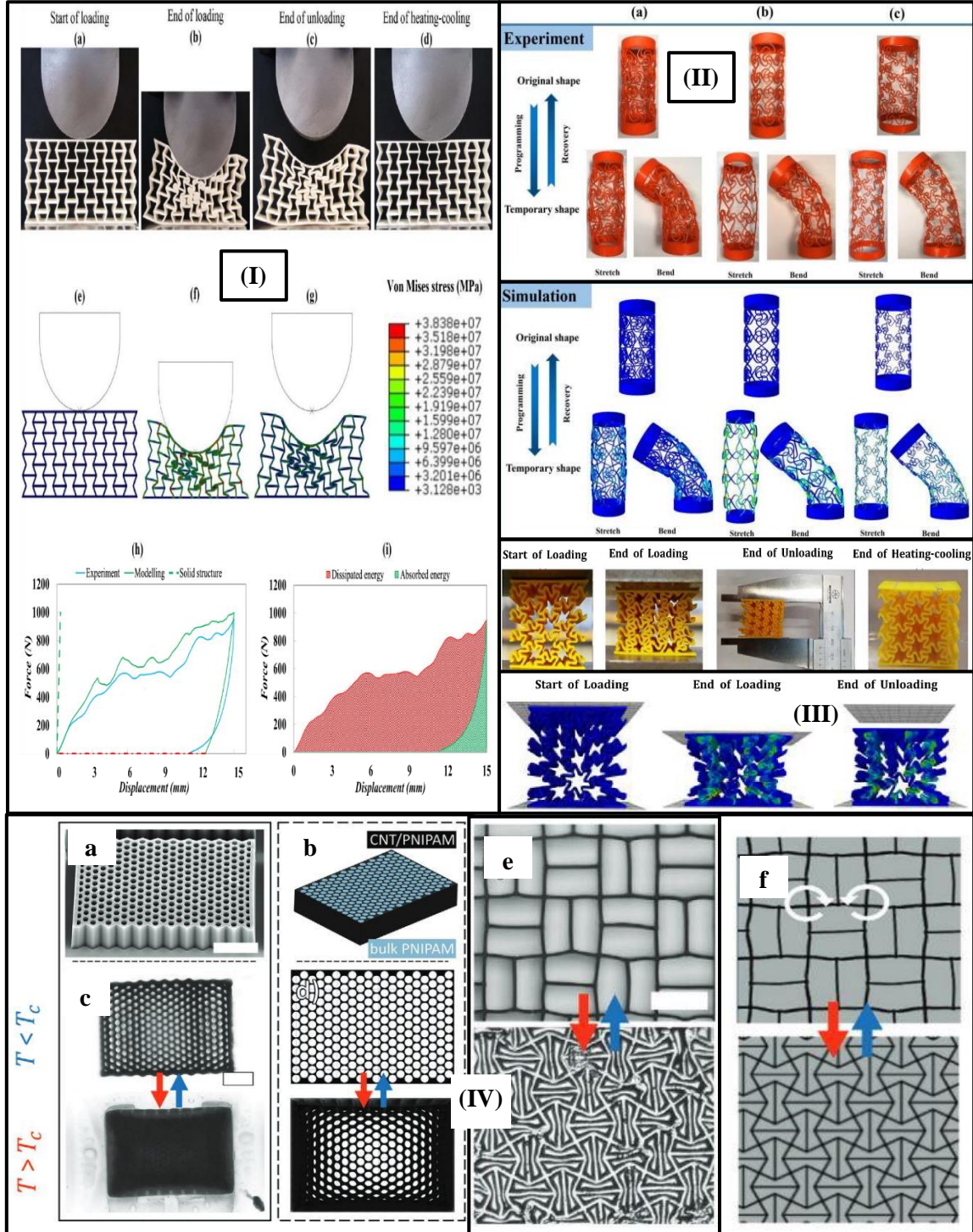


Figure 10. (I) Re-entrant auxetic structure metamaterial; (a)–(g) Compressive deformations behavior for both experimental and FEM simulations; (h) Force-displacement response under experimental and simulation studies; (i) Absorbed and dissipated energies [250] (adapted with permission); (II) Comparison of experimental and simulation results; Shape memory behavior for the cylindrical shells under axial stretch and bending loading for (a) Triangular (b) Square, and (c) Honeycomb lattice metamaterials [251] (adapted with permission); (III) Comparison of experimental and simulation-based studies deformation behavior results for sandwich structure [252] (adapted with permission). (IV)

(a-b) Images of pristine carbon nanotube structure employed for micro actuator (a) Actual SEM image (b) Finite element simulation model. (c-d) CNT/PNIPAM composite hydrogel micro actuators reversible actuation of surface bound, (c) Experimental results showing transparency switch from opening and closing of inner cells. (d) Finite element simulation results of CNT/PNIPAM hydrogel actuators demonstrating contraction under deswelling that almost similar to result in (c). (e-f) Actuation behavior of lattice unit cell of CNT/ hydrogel (e) microscale change in shape (f) finite element simulation results, presenting through shape change via local rotations [253] (adapted with permission).

In another study, Wan et al. [251] investigated triangular-, square-, and honeycomb-shaped 4D-printed programmable metamaterials. The experimental and FEA-based studies were adapted for different topological angles and radii. The simulation results confirmed that the triangular and square lattice metamaterials demonstrated large deformation and auxetic behaviors, as depicted in Figure 10(II), due to SMEs. Likewise, Serjouei et al. [252], 4DP of smart sandwich structures with the potential of energy absorption was explored through experimental and numerical procedures. FEM of horseshoe sandwich structures successfully predicted the functional behavior during compression analysis, as depicted in Figure 10(III). Both numerical and experimental results showed that the compressive load and energy absorption rate were increased due to different process parameters. Similarly, Gregg et al. [253] showed micro actuators behaviors of CNT/PNIPAM nanocomposites. Different shape changing behaviors such as opening and closing holes, shape changes of each lattice unit cell, and transparency changes were successfully predicted through finite element simulation techniques as elaborated in Figure 10(IV). Experimental and simulation results confirmed that the addition of CNTs to PNIPAM hydrogels improves actuation time under both light- and heat-stimulus.

4.2 Machine learning models for 4D-printed smart materials

It is often difficult to model and predict the motions of actuators and soft robots due to the non-linearity of the materials [254]–[256]. Furthermore, linear analytical models cannot precisely predict the actuation mechanism of 4D-printed robots, and FEA incorporating non-linear models helps in improving robots' efficiency [257]–[259]. In the contemporary era, deep learning (DL), artificial intelligence (AI), and machine learning (ML) are extensively applied in the field of smart materials and system design. ML model learns from the provided data sets which are fed as input to predict the future outputs [260]. During design, the ML approach is a versatile and effective tool to predict the actuation mechanism of 4D-printed robots, which could help in saving time [260]. It is an alternative approach used for optimizing the design of high-performance smart materials without investigating the entire design space [261]. Recently, different researchers have employed ML-based approaches for developing soft robots and actuators. For instance, Sun et al. [262] proposed a novel ML- and evolutionary algorithm (EA)-based technique for designing 4D-printed active composites, as illustrated in Figure 11(a₁). For predicting the forward shape-change, a recurrent neural network (RNN) based ML model was developed, which was trained through a dataset by FEA. For different target shapes with multiple complexities and optimal design, the ML-EA technique demonstrated high efficiency. Finally, the proposed ML-EA technique has significant use in various hand-drawn lines and transformed into various 4D profiles under the swelling stimulus, as highlighted in Figure 11(a₂).

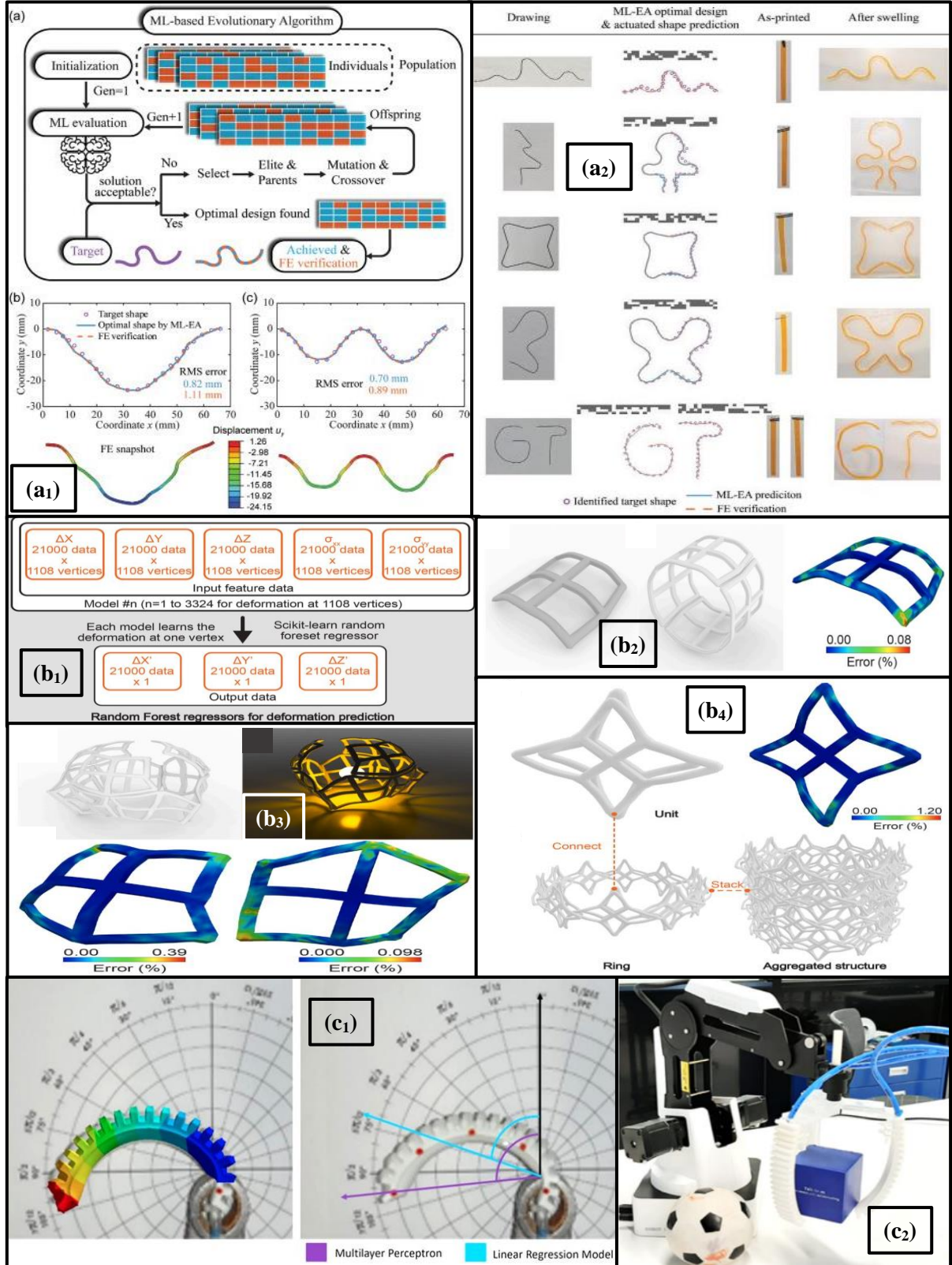


Figure 11. (a₁) A schematic illustration of ML-EA optimization approach which for optimized different shapes and corresponding FE shape; (a₂) Illustration of different models (from left to right) line drawing by hand, ML-EA approach designs and predicted optimal shapes, 4D-printed shapes, and shapes under stimulus response [262] (adapted with permission); (b₁) ML model and error distribution between prediction and ground truth for different design model; (b₂) Arm band; (b₃) Lamp cover; (b₄) Stacked

aggregation sculpture [263] (adapted with permission); (c₁) Actual experimental deflection result of rectangular SPA superimposed with both FEM and ML model (neural network prediction); (c₂) 4D-printed SPA grasping box at different bending positions [261] (adapted with permission)

FEA has shown different disadvantages. For example, error due to geometric approximation and the FEA technique is a time-consuming computational analysis due to the high degrees of freedom. ML-based approaches can resolve these problems. One such approach is used by Yu et al. [263] who proposed a hybrid design using both FEA and isogeometric analysis (IGA) elements, which produced accurate simulation results due to a lower degree of freedom. To modify further this FEA-IGA in terms of faster deformation predictions, they further introduced the ML model through a polycube-based random forest regressor as depicted in Figure 11(b₁). Results showed that the ML approach for different complex shapes as illustrated in Figure 11(b), provided 20 times faster simulation results than hybrid IGA-FEA simulations with an error of less than 0.11%. In another study, Zolfagharian et al. [261] studied ML and (finite element) FE-based models for accurately estimating the specific bending angle of a 4D-printed soft pneumatic actuator (SPA), as presented in Figure 11(c). FEM was used in for the simulation of experimental actuation using a nonlinear hyperelastic model, which later trained thousands of data for the ML modeling. An ML model was successfully developed for predicting bending angle, pressure, and shape. Results showed that the rectangular-shaped actuator possessed the highest bending angles among the circular and triangular geometries under different variables such as the bellow shape, width, height, thickness of the bottom layer, and air pressure. ML models have significant potential for optimizing hyperparameters along with 94.3 % accuracy.

5. Applications of 4D-printed smart materials

4DP of intelligent/smart materials has been utilized to develop soft devices such as sensors, actuators, robotics, and grippers for a wide range of engineering applications, as illustrated in Figure 12. The main goal of 4DP is to develop multi-functional and smart devices, for these applications [264]. This section incorporates various engineering and biomedical applications of 4D-printed robotics.



Figure 12 Applications of 4D-printed smart/intelligent materials

Origami structures: Intelligent origami structures are extensively applied to develop 3D spherical architectures from 2D parylene C thin films [265]. Active origami structures are vastly applied to the development of self-folding and self-soiling equipment such as robots [266]. Origami-based 4D-printed soft actuators and robots exhibit tremendous potential for classifying and sorting food and agricultural products [267]. The climb or crawl-like motions of these actuators are applied for pipe inspections, inaccessible environments, rescue missions, and drug delivery smart devices [268]. Origami-based soft robots/actuators permit variable stiffness and high efficiency, compared to conventionally fabricated robots. These robots can also be applied in endoscopy, laparoscopy, and autonomous surgeries [269]. For instance, a hybrid hinged structure was able to expand and deform like an origami structure even after 4DP and was investigated by Yamamura et al. [270]. This 4D-fabricated hybrid hinge exhibited high durability and elasticity, as it recovered to its original shape without failure after 500 cycles of folding, as illustrated in Figure 13(Ia). Additionally, Miura-ori and origami compliant mechanism grippers were also fabricated, and these origami structures deformed below the T_g of the SMP. These 4D-printed parts have excellent perspectives in developing self-folding actuators and robots, which involve origami deformation.

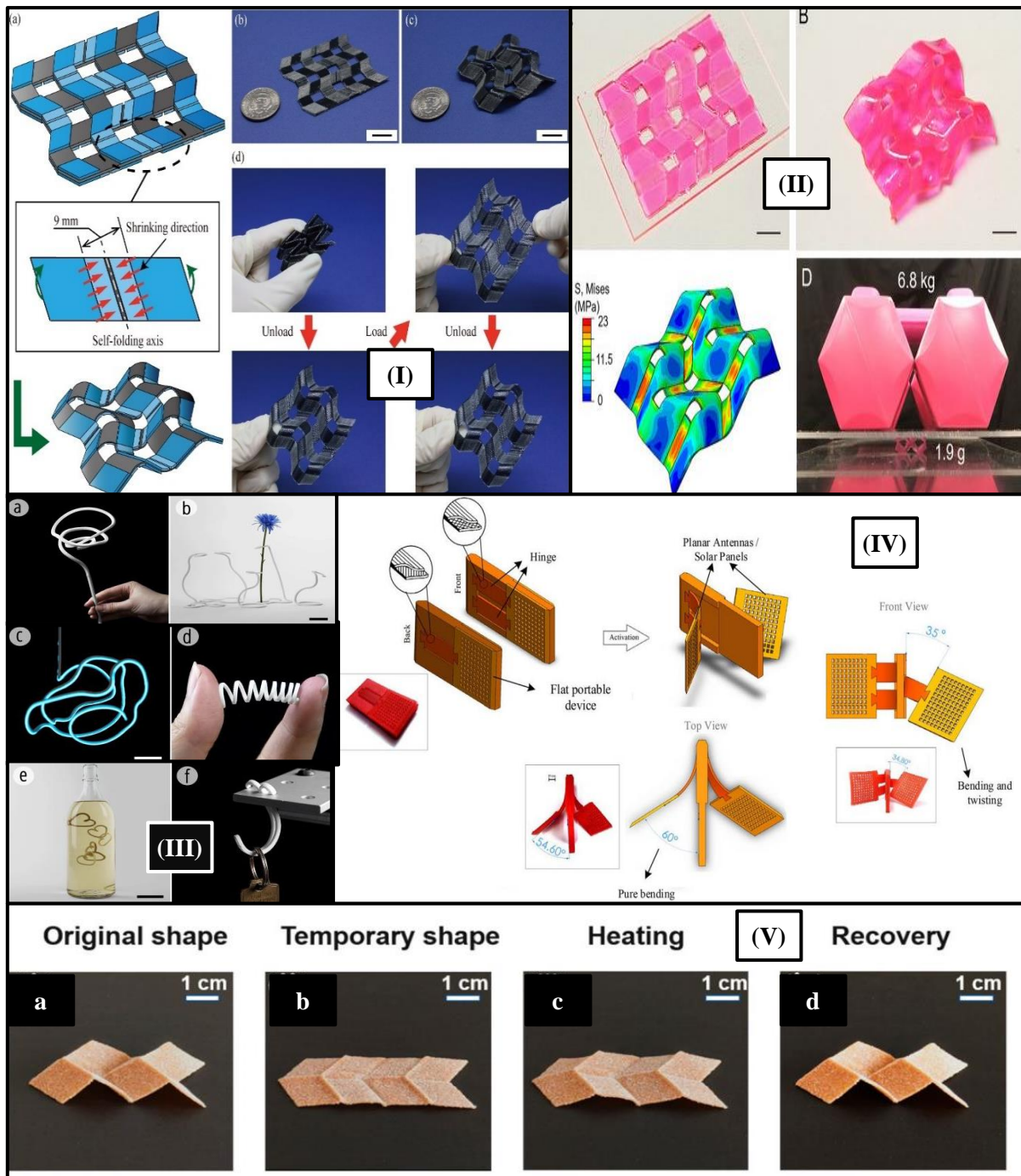


Figure 13.(I) (a) Complex origami structure with various hybrid hinges fabricated through 4DP; (b-c) Pictures of origami structure before and after 4DP; (d) Folded and unfolded deformable shapes of origami structure under an external force [270] (adapted with permission); (II) Printed and simulated Miura-origami structures for high load bearing applications [271] (adapted with permission); (III) 4D-printed different A-line elements for different applications; (a) Rose; (b) Vases; (c) Lamp; (d) Spring; (e) Self-deployable wishing heart; (f) Self-locking hook [272] (adapted with permission); (IV) 4D-printed hinges in structures for different applications [197] (adapted with permission). (V) 4D fabricated Miura origami structures and their shape memory behavior under near-infrared (NIR) light stimulus [233] (adapted with permission).

Deformation mechanisms like origami have also been applied to printed expandable and self-deployable structural components. For instance, Weng et al. [271] manufactured self-morphing structures with large deformation and high modulus through DIW using photocurable polymer

resin, short glass fibers, and fumed silica-based composite inks. The results showed that the proposed technique for printed composite structures had excellent mechanical properties, especially in morphing lightweight structures with load-bearing capabilities. For instance, a Miura-ori structure, as depicted in Figure 13(II), can hold a load of 6.8 kg which is ~ 3580 times its weight.

Likewise, Wang et al. [272] fabricated different 3D morphing shapes through 4DP technology and designed A-line elements using thermoplastic material. These A-line elements can be applied for designing in line sculpting, self-deploying, compliant mechanisms, and self-locking structures, as illustrated in Figure 13(III). Similarly, Aka hinges have the potential to be used in many structural engineering applications. These aka hinges are entirely possible due to the 4DP of fat self-bending structures. Nezhad et al. [197] developed aka hinges through 3DP of fat structures. SME of these 3D-printed bilayer structures is effective in controlling and predicting the shape-shifting behavior of these hinges, as demonstrated in Figure 13(IV). Ouyang et al. [233] developed SLS-based 4D printed Miura origami structure using PUDA/CNT methylene diphenyl diisocyanate materials. These complex architectures showed excellent self-healing, and shape memory performances under the NIR light stimulus as presented in Figure 13(V).

Electronics: 4DP technology has gained exceptional importance in electronics and is used to print electronic devices like sensors and actuators [273]. Energy sources and electronic controllers can be embedded inside these stretchable/flexible products to develop highly versatile soft robots [274]. However, conductive ink materials are key requirements for developing these electronic embedded devices [275]–[277]. Different authors developed 4D-printed electronics sensors for various engineering applications. For instance, Li et al. [205] developed a 4D-printed resistive transparent strain sensor-based flexible transparent electrodes (FTEs) device through liquid substrate electric-field-driven microscale printing. The real application of the developed sensor was successfully demonstrated in measuring the strain variables at different positions in the human body, as depicted in Figure 14(Ia). The results showed that these thermally driven 4D-printed structures with resistive transparent strain sensors are remarkable in sensing different human expressions and for sensing positioning of joints, as illustrated in Figure 14(I). Additionally, these FTEs-based optoelectronics devices exhibited excellent mechanical stability and environmental adaptability.

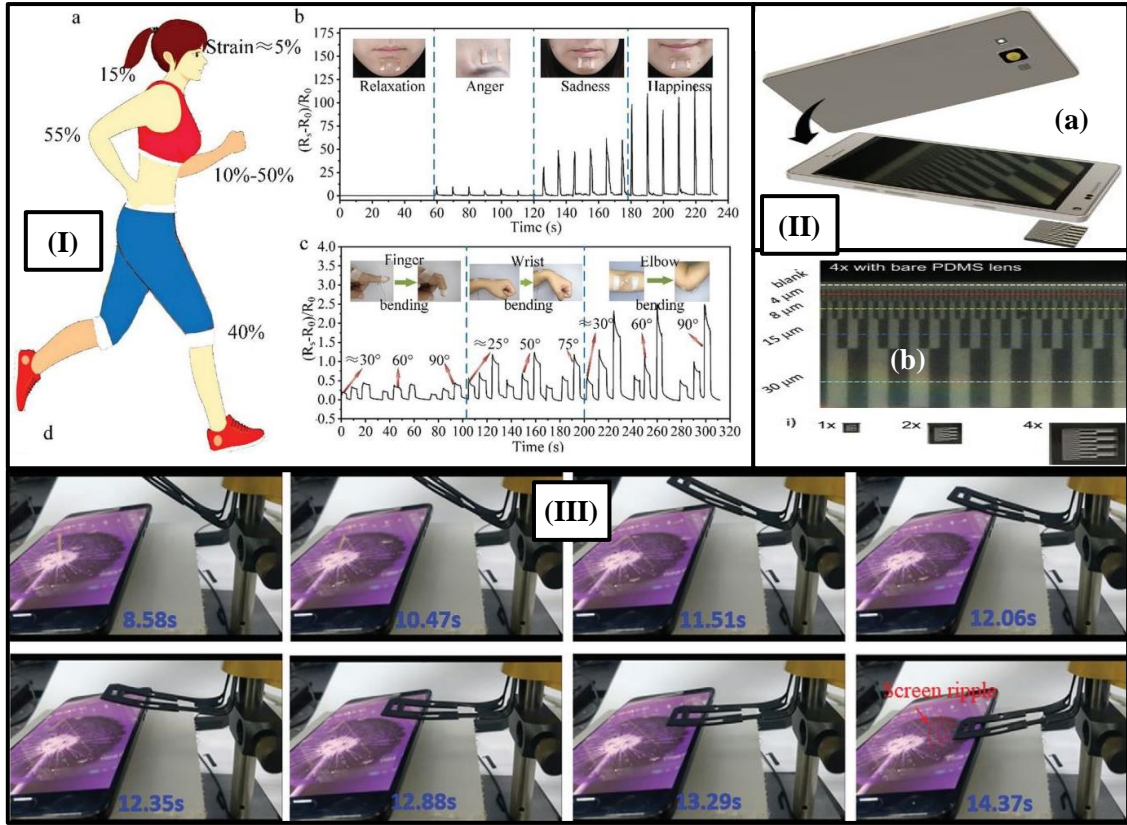


Figure 14. (I) (a) 4D-printed stress variables sensors for different positions of the body; (b) The strain sensors for measuring the facial expressions responded in real time; (c) The strain sensor for measuring human joints responds in real time under different bending angles [205] (adapted with permission); (II) (a) PDMS lens-based smartphone sketch for imaging applications; (b) Images of chromium-on-quartz test pattern including lines of various widths obtained from a PDMS lens adhered to the camera of a commercial smartphone (At a focal length of 2.8 mm and 4× digital zoom) [278] (adapted with permission); (III) Schematic figures of the PISA that mimicked the finger of an active touching mobile phone and reflected the touching information through changing resistance at different times [279] (adapted with permission).

Chen et al. [279] adopted a 4DP bioinspired microstructure strategy for designing a high-performance integrated sensor-actuator demonstrating both actuation and sensation at a time. Thermal stimulation and strain sensation was applied by combining nanocarbon black/PLA composites with bioinspired gradient micro-gap structures. Figure 14(III) depicts a printed integrated sensor-actuator (PISA) that can actively touch objects initiated by thermal stimulation and self-sense the touching state through the resistance change. Thus, PISA actively touched the phone screen and triggered the screen saver, creating a ripple. In another study, Mariani et al. [278] investigated the behavior of a PDMS magnifying lens programmed with a tunable plasmonic rejection filter through 4DP. Ag- and Au-based NPs were employed for integrating nanometer-thick plasmonic filters on the lens surface through in situ synthesis of NPs with programmed density. The purpose of the filter was to reject and pass the light at the specific plasmonic resonance wavelength of the NPs through the swelling of PDMS in hexane and ether. This was responsible for changing the NP density and transmittance properties of the lens. Later, the proposed methodology was employed on a commercial smartphone with the plasmon-encoded lens. The potential applications of 4D-printed NP-decorated PDMS lenses in different fields such as color tuning, light-filtering, and on-field

microscopy for detecting different bacteria and pathogens on the wound surface, and food, including live monitoring of the quality of water through identification of protozoa and protists.

Wireless sensors: 4DP of intelligent materials has the potential for developing wireless sensor systems and radio communications antennas due to the self-assembly nature [280]. For instance, Jeong et al. [202] proposed a 4D-printed metasurface for memorizing the absorption and reflection functions to improve wireless communication, particularly in millimeter-wavelength regimes. The experimental results showed that these 4D-printed metasurface structures exhibit good absorptivity and peak absorption values. Thus, this metasurface can be used in various smart electromagnetic wave control systems in tunable intelligent surfaces, and wireless sensing systems. Similarly, Wu et al. [281] fabricated a thermally deformable bowtie antenna using nylon and carbon fiber laminated composite through 4DP. The bowtie antenna was placed on the composite material surface where carbon fiber was energized and heated, which was responsible for the thermal deformation of the substrate to reconfigure the antenna feature. The results showed that 4D-fabricated bowtie antenna structures, as depicted in Figure 15(II), offer a wide frequency range and radiation characteristics and can also be employed in 5G communications, flexible electronics, and wearable devices. Furthermore, the control of antenna beamwidth and gain was attained by implementing electrically controllable deformation of the 4D model.

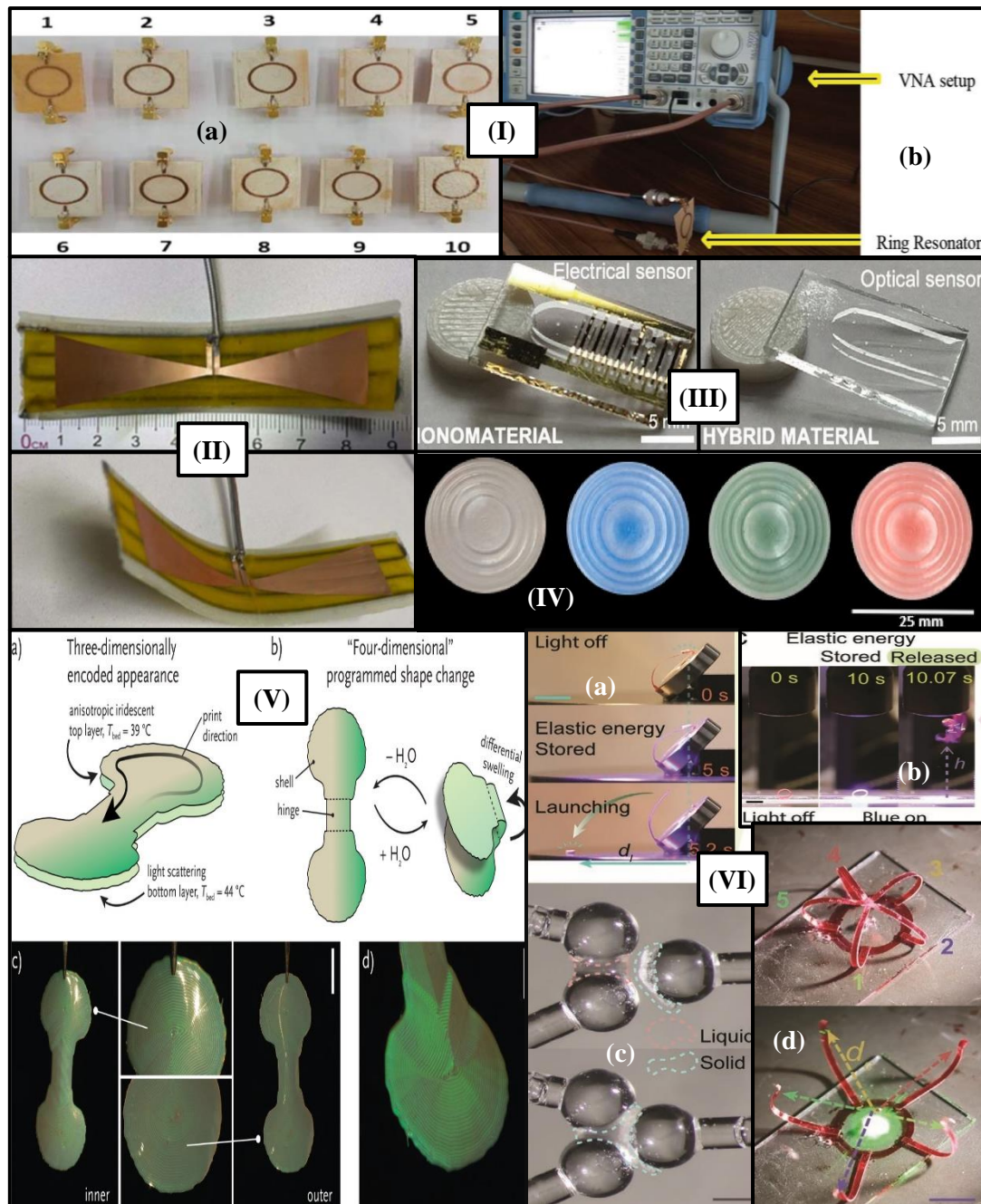


Figure 15. (I) (a) ABS ring resonator prototypes models; (b) Testing of prototype through VNA and an exact 2.45 GHz was observed through the proposed prototype model [282] (adapted with permission); (II) 4D-printed model of the deformable bowtie antenna [281] (adapted with permission); (III) Electrical and optical sensors derived from thin films [283] (adapted with permission); (IV) 4D-fabricated Fresnel lenses for focusing a particular color and temperature sensing applications [284] (adapted with permission); (V) (a) Figure depicting the bilayer design of the object printed with different temperatures; (b) 4D behavior of the actuator; (c-d) Pictures displaying the “inner” and “outer” layers and close up of the photonic actuator when illuminated from overhead and oblique. The actual actuation behavior when relative humidity was changed at isothermal conditions ($T \approx 22\text{ }^{\circ}\text{C}$). Differential swelling for bending developed through acidic treatment of the backside of the clam's hinge [285] (adapted with permission); (VI) (a) Images of light-based catapult motion; (b) Images of light-controlled jumping; (c) Schematic illustration of bonding process with the liquid-crystalline adhesive; (f) Snapshots of the integrated LCE strips before (top) and after (bottom) launching [286] (adapted with permission)

Jain et al. [282] reported radio frequency (RF) characteristics of recyclable ABS substrate-based sensors using 3DP technology while demonstrating 4D capabilities under two different chemical and thermal-based stimuli. The simulated response of 3D ring resonator-based prototypes was confirmed with a vector network analyzer (VNA)-based experimental study results, as highlighted in Figure 15(I). The reported results showed that the prototype sensor has the ability to detect a minimal frequency shift, thus, exhibiting great promise in Bluetooth applications.

Optical sensors: To date, different smart intelligent electrical and optical sensors have been created through 4DP technologies and have exhibited the excellent potential to be used in many optical sensor applications [287]. For instance, Esteves et al. [283] developed different optical and electrical sensors for humidity sensing and controlling humidity interference in volatile organic compounds through the design of gelatin-based ionomaterials, as depicted in Figure 15(III). The results showed that the developed sensors have a wide range of applications in e-nose sensing arrays and wearable devices effective under room conditions. Likewise, Ali et al. [284] DLP-fabricated Fresnel lenses-based photonic devices by introducing spectral color variation as the 4th dimension to 3D-printed lenses under temperature stimulus, as depicted in Figure 15(IV). In this study, thermochromic pigment powders were added to liquid monomer resins. Normalized power intensity and transmission spectra measurement setups were used for elaborating the printed lenses focusing ability and thermal sensing. These sensors have the potential to detect temperature changes remotely on surfaces of interest and can also measure strains, stresses, and pressures on certain surfaces.

In another study, Sol et al. [285] synthesized a humidity-responsive photonic actuator through the DIW technique and cholesteric liquid crystal oligomer ink, which exhibited a multicolored appearance. Hydrochromic coating was deposited onto a 3D-printed beetle and with the atmospheric humidity imparts multicolor appearance characteristics. Furthermore, with the insertion of 3D-printed beetle in aqueous acid, the beetle showed vibrant color shifts across the visible spectrum. Figure 15(V) depicts an acid-treated bioinspired scallop actuator upon exposure to humidity showing the reversible “opening” and “closing”.

Snapping is an important phenomenon through which structures rapidly transform from one stabilized form to another as a result of some mechanical instability developing in the system. A sudden release of stored elastic energy is often required for snapping. Guo et al. [286] attained similar snapping phenomena using responsive materials, which are triggered by external stimuli. The authors studied a light-fueled snapping-like launching with excellent control over the elastic energy, which was prestored in a light-triggered LCE-based actuator. Figure 15(VI) depicts that a 4D-printed actuator enabled different motions like catapult and jumping, and can be employed for multiple launchers and soft robotics applications.

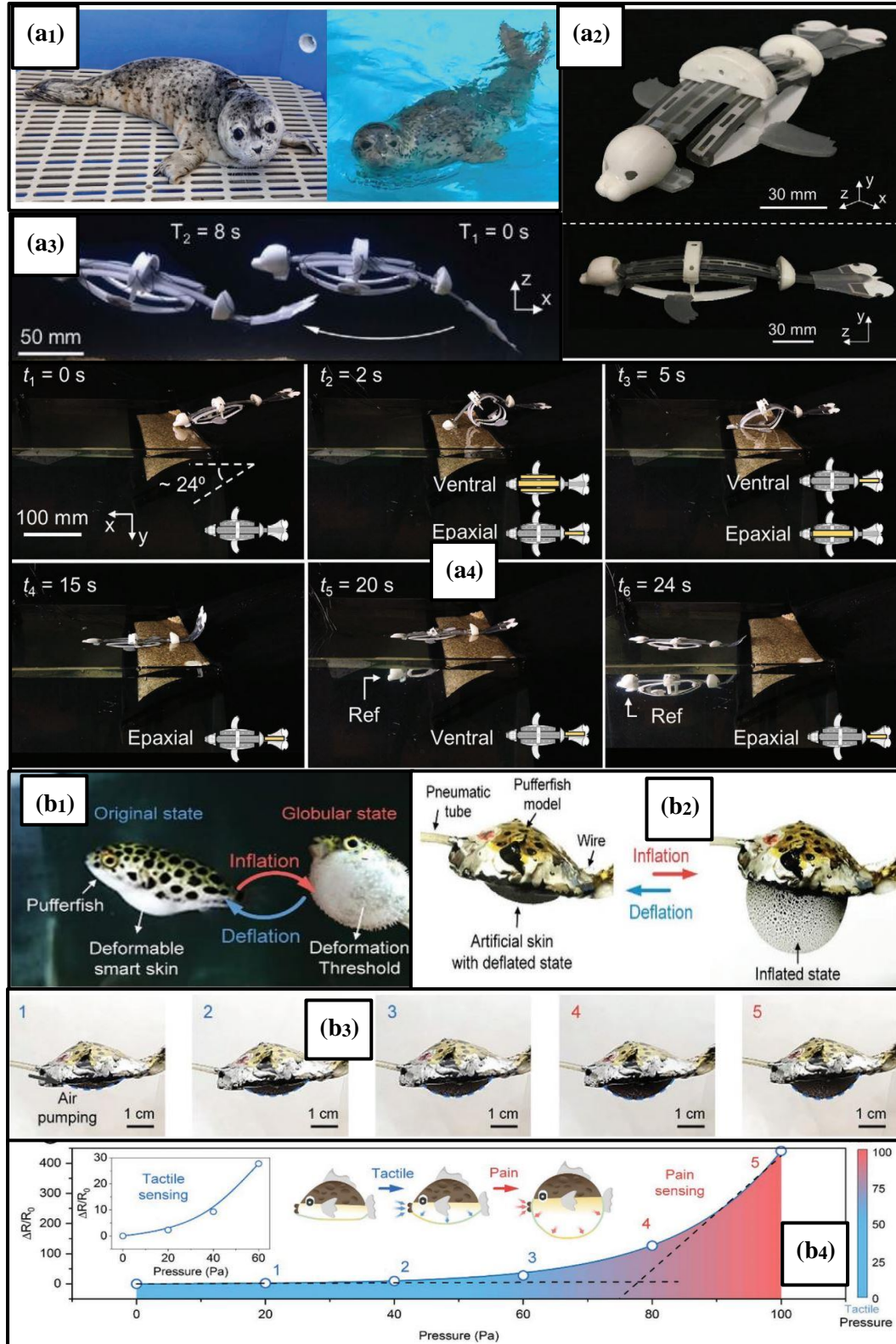


Figure 16. (a₁) Terrestrial and subaqueous locomotion of Crawling seal pups. Different views of soft amphibious robot; (a₂) perspective and side views; (a₃) SEALicone swimming underwater; (a₄) Demonstration of transition behavior of soft robot from one environment (terrain) to another (water) [288]. (b₁) Pufferfish photo in inflated and deflated conditions; (b₂) Artificial pufferfish model; (b₃) Images of actuated artificial pufferfish model; (b₄) Outstanding color mapping of pain warning as obtained from unit pressure for pain warning through the graph of normalized resistance versus pressure curve with sharp improvement of the normalized resistance [289].

In another study SMA-based soft amphibious robot named as SEALicone, capable of demonstrating multimodal locomotion both aquatic and terrestrial was developed (referring to Figure 16(a)). The robot was composed of the backbone actuator (wave-like deformation) and support actuator (fusiform deformation). The fabricated soft robot actuators were capable to produce different locomotion styles that were inspired by crawling seals. The potential applications of the SEALicone in shoreline exploration [288]. Similarly, in another study strain-perception-strengthening (SPS) mechanism which triggers the dynamic transformation was captured by designing , a bionic pufferfish with synthetic skin. Tiny airflow and finger touch-based different external mechanical stimuli were applied and later inflated itself to a 3D deformation determined by the SPS effect as presented in Figure 16(b). The proposed methodology of soft skins with SPS effect have promising applications in safe human-machine interaction, soft robotics and smart prosthetics [289].

Li et al. [290] developed a novel multifunctional MPDMS/MXene/PTFE-based soft actuator capable of producing stable heat and generate large and quick bending deformation under low voltage stimulus. Results showed that the printed actuator has potential to use in construction of numerous intelligent devices as presented in Figure 17(a) for bionic motion, such as heating the irregular objects along with various intelligent crawling robots features. Similarly in another study [133] nanocomposite smart hydrogel-based smart robots that can respond to both temperature and magnetic field was fabricated. The double-layer structure was synthesized using 4DP the driving unit to understand bending deformation of various amplitudes under thermal stimulus. A shellfish-like robot and the leptoasteria-like robot were fabricated with the ability of active cargo transport as presented in Figure 17(b). Roach et al. [291] studied liquid crystal elastomers for their large, reversible mechanical actuations under temperature stimuli through a novel ink formulation. The multiple advantages were associated with the proposed technique such as it permits the 3D printed liquid crystal elastomers to be integrated with other 3DP techniques and materials to form more complex shape changes as demonstrated through, a soft robotic gripper (referring to Figure 17(c₁)), and a printed hand for sign language referring to Figure 17(c₂). Lately a silicone-based embedded 3DP technique, referred as Rapid Liquid Printing (RLP) was considered for the fabrication of soft pneumatic actuators. The advantage of RLP technique was demonstrated through designing a complex, multi-chambered from the actuator geometry capable to perform various soft robotics applications such as bending and grasping as presented in Figure 17(d).

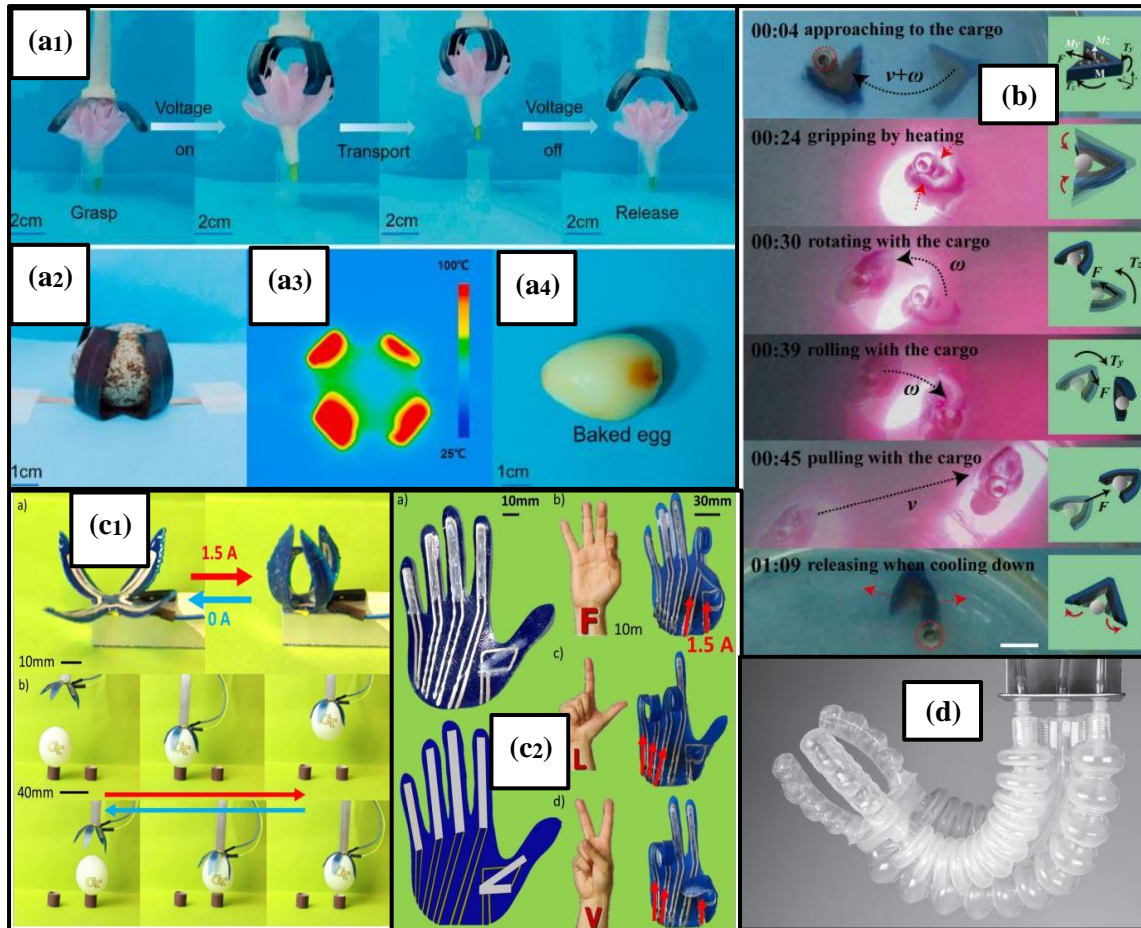


Figure 17. (a₁) The intelligent gripper actuated by voltage drives grabbing a flower; A quail egg wrapped by four fingers actuator (a₂) actual view and corresponding (a₃) infrared thermal image views; (a₄) Image of quail eggs cooked via four fingers actuator [290]. (b) A shellfish shape-based gripper demonstrating its complete cargo delivery [133]. (c₁) Four hinge soft robotic gripper picking and placing a ping pong ball under current stimulus; (c₂) Schematic of the hand with five different conductive paths for prescribed bending through five hinges hand [291]. (d) soft robotic printed arm fabricated through rapid liquid prototyping demonstrating bending and gripping behavior [292].

Biomedical: There is a rapid increase in the adoption of 4DP technology in the biomedical field and this technology presents an infinite potential in biomedical engineering [293]–[295]. For instance, 4D-printed heart and brain can prove to be beneficial, as these artificial organs precisely fit at the locations under specific stimuli [296]–[299]. 4D-printed intelligent material-based actuators play an important role in the development of biocompatible medical robotics [300]. These robots exhibit extraordinary features like adaptability, biodegradability, and biocompatibility [301]–[303]. Nowadays, micro/nano-scaled robotic swimmers are designed through 4DP technology for conveying biomedical functionalities throughout the human body [304]. It requires an appropriate power supply for generating locomotion. However, it is essential to use an acceptable environment for the actuation of smart robots. Bozuyuk et al. [305] developed a chitosan/iron oxide (Fe₂O₃)-based micro-swimmer for drug delivery applications. The results suggested that printed micro-swimmer exhibited excellent drug-releasing properties upon light stimulus. This highly efficient and versatile light-triggered drug delivery micro-robot can also be triggered under multi-stimuli.

4DP technology has huge potential to print pH-responsive soft robots, biocatalysts, valves, actuators, and devices for drug delivery [306]–[308]. For instance, Wu et al. [309] fabricated a

cost-effective pH measurement device through the FDM process by incorporating poly(4-vinylpyridine) into ABS thermoplastic filaments. The 4D-printed pH sensing claw revealed linearity between the pH values from 5.0 to 8.6. Thus, the printed device exhibited a geometrical change in the form of swelling in the presence of a strong buffer solution, as depicted in Figure 18(a). Similarly, Wang et al. [310] developed bionic asymmetric micro-actuators through femtosecond laser 2PP technique by using 2-(dimethylamino)ethyl methacrylate (DMAEMA)-based pH-responsive monomer. This flytrap-like bionic micro-actuator was employed for capturing of microspheres with a deformation time of 1.2 s and recovery time of 0.3 s. Figure 18(b) demonstrates the release behavior of multiple microspheres by manipulating and employing different simultaneous release processes and successive release strategies. The proposed micro-actuators have potential applications such as soft robotics, bionic devices, and precision sensors. Likewise, Malachowski et al. [311] developed a micro-gripper for sustained release of drugs in the gastrointestinal tract, and Azam et al. [312] fabricated a drug delivery polyhedral device using thermo-responsive PCL and SU-8 faces. Additionally, 4D-printed soft robots also possess tremendous potential in performing minimally invasive procedures [313].

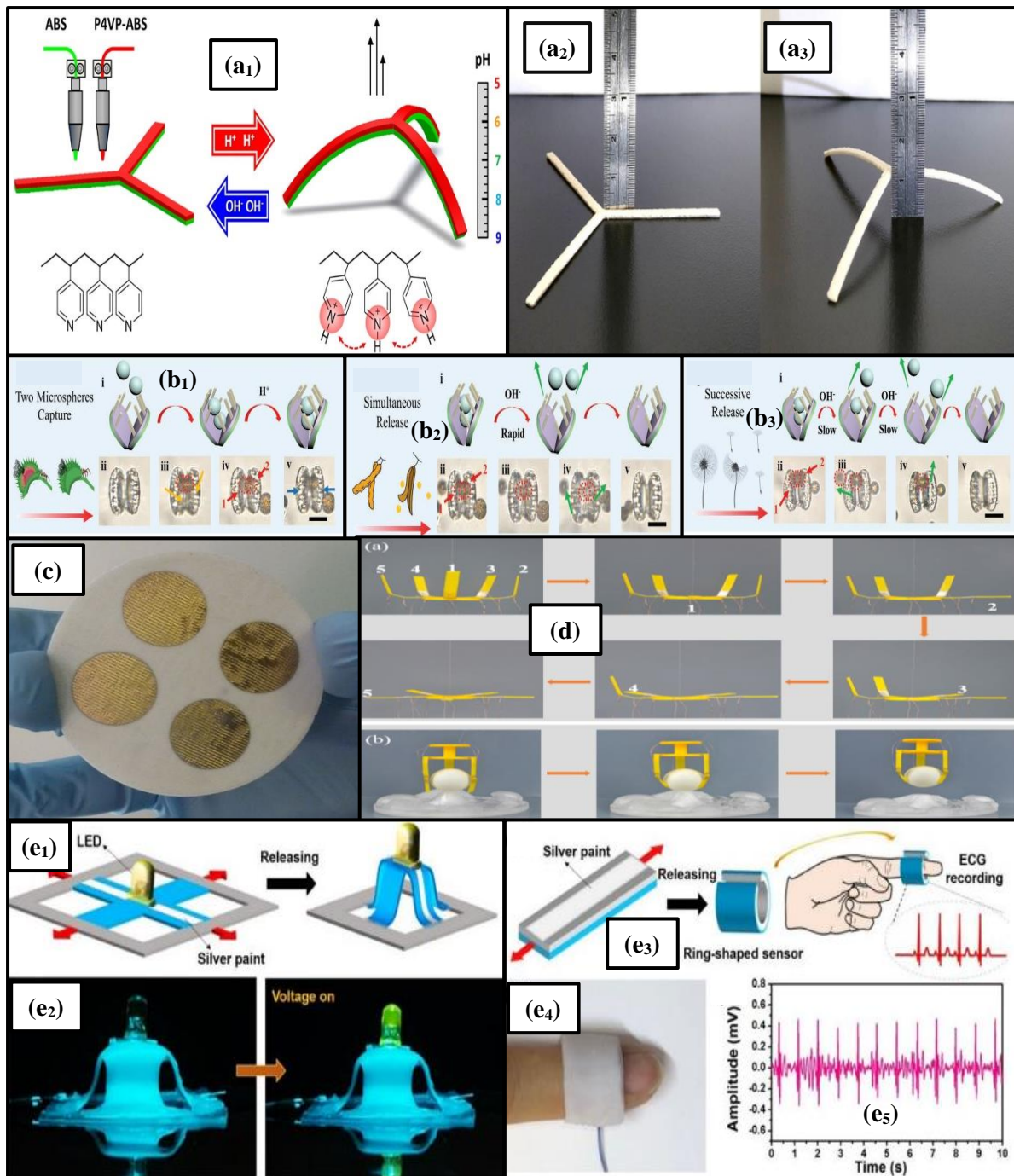


Figure 18. (a₁) Schematic diagram of pH sensing claw highlighting three cuboids extending through the center and its [H⁺]-responsive shape-programming. Image of the 4D-printed pH sensing claw; (a₂) Before its immersion; (a₃) After its immersion in buffer solution [309] (adapted with permission); (b₁-b₃) Illustration capture and release behaviors of multiple polystyrene microspheres by the bionic flytrap micro-actuators through various schematic diagrams as well as bright field images [310] (adapted with permission); (c) 4D-fabricated piezoelectric composite [314] (adapted with permission); (d) 4D-printed biomimetic flowers with five petals and each petal expands independently under the applied voltage. The gripper can hold a ball under extreme environmental conditions [315] (adapted with permission); (e₁- e₂) Schematic diagram showing 3DP of electronics and its function to light up as light-emitting diode; (e₃) Schematic illustration of manufacturing of a ring-shaped ECG sensor; (e₄) ECG sensor wrapping on a human finger; (e₅) Actual ECG measurement [316] (adapted with permission)

A few researchers have also developed biosensors through 4DP technology to detect various physical activities of metabolites and cells for diagnostics [317]. For instance, Grinberg et al. [314] 4D-printed piezoelectric composite-based sensor with exceptional smart sensing capabilities. The reported results demonstrated that high sensitivity and a good linear relationship between the resulting electric charge and the input mechanical excitation were observed for the piezoelectric composites shown in Figure 18(c). These piezoelectric composites have excellent perspective in a knee prosthesis for high-fidelity measurements of the mechanical strain. In another study, Deng et al. [316] fabricating multi-stable shape-morphing 3D structures which were programable with applied strains using phase change wax microparticles in the elastomer matrix. Multi-stable buckling modes were observed due to the varying applied strain directions in the same elastomer-based composite films. Thus, 4DP of the elastomer composites demonstrated the functionalities in the assembly of 3D electronics and adaptive wearable sensors for measuring real-time electrocardiogram (ECG), as highlighted in Figure 18(e).

Intelligent materials are also applied to develop bioactuators through 4DP technology [318]. These 4D-printed bioactuators undergo actuation for delivering therapeutic agents at the specified locations under some external stimulus [319]. For instance, Shao et al. [315] created novel electrically driven 4D-printed PLA-based composites by incorporating silver nanowires (Ag-NWs). These Ag-NWs act as a “heater” for simulating the local deformation of PLA under an applied voltage. A biomimetic flower with five petals was fabricated with each petal controlled independently, as depicted in Figure 18(d). The printed model can also be employed for different kinds of SMPs and have potential applications to be applied as bioactuators. Table 2 summarizes the most recent works done by the researchers to develop soft robots for different engineering applications.

Table 2. Summary of recent works on various functions/applications of soft robots fabricated through 4DP

Year	Smart materials	Stimulus	Soft robotics functions	Applications / features	Ref.
2022	MPDMS/ MXene / PTFE	Voltage magnetic field	Crawling, gripping and dragon fly motion	Intelligent crawling robot	[290]
2022	-	-	The robot comprises two types of SMA-based soft actuators: The support actuator with fusiform deformation and the backbone actuator with wave-like deformation.	Soft amphibious robot having SEALicone capabilities generates multimodal locomotion (terrestrial and aquatic).	[288]
2022	NIPAM / Laponite nanoclay / NdFeB	Temperature magnetic field	A catheter with a multi-segment magnetic head, a shellfish-like robot, and a leptasteria-like robot have slipping, rolling, gripping, and climbing functions.	The 4D-fabricated different robots such as shellfish-like and leptasteria-like robots have the ability of active cargo transport on the wrinkled surface of the stomach model.	[133]
2022	LCE	Light	Climbing and controllable steered motion	Helps in fabrication of miniaturized devices for taking more tasks with wireless.	[320]
2022	-	Magnetic field	Multimodal locomotion	The developed magnetic micro/nanorobots generate reactive oxygen species in vivo to induce tissue/organ damage.	[321]
2022	PDMS	Magnetic field	Across the different length scale a remotely, and reversibly controlled responsive behavior was observed.	Potential to use in various applications such as soft	[108]

				crawling robots, , bionic butterfly, flexible grippers and multistate magnetic switch.	
2021	LCE	Heat	Untethered self-propelling soft robot exhibited tactile perception	4D-fabricated soft robot had strong transportation ability.	[322]
2022	Ferromagnetic PLA	Heat	Precise multi-step actuation and remote actuation	Potential application in deployable structures.	[169]
2019	Gelatin/mercapto-ester PU	Temperature	Multiple shape-memory properties, such as expansion and stretching	Manufacturing of 4D stent-like medical smart devices.	[191]
2019	P(DMAAm-co-SA)	Water	Transformation of the flat flower shape to the 3D blooming state	4D-fabricated soft robots demonstrated for gripping transportation applications.	[266]
2018	PDMS	Heat	Shape changing phenomenon from circular to square shape, and rotation of the frame	Temperature steered light transmission devices, polarization monitoring, and polarimetry.	[129]

5.1. Role in COVID-19 Pandemic

Nowadays, 3D-printed products are highly advantageous to healthcare systems due to minimal training, rapid prototyping, as well as minimal assembly time, and operations [323]. Additionally, this technology has demonstrated its potential in developing medical equipment under pandemic situations such as Coronavirus Disease 2019 (COVID-19) [324]. COVID-19 pandemic has terrified people due to the huge number of deaths and the post-COVID world has completely changed the lives of the people. During the pandemic, different 3DP technologies help in fabricating low-priced medical equipment, as depicted in Figure 19. Figure 19(a-j) depicts different 3D-printed equipment like nasopharyngeal swabs, Y-connector and T-connector for ventilator machines, face shields, and hand-free door openers for the pandemic. Similarly, different companies have also developed drones for contactless delivery. Additionally, these articles of healthcare equipment are mostly fabricated using polymeric materials.

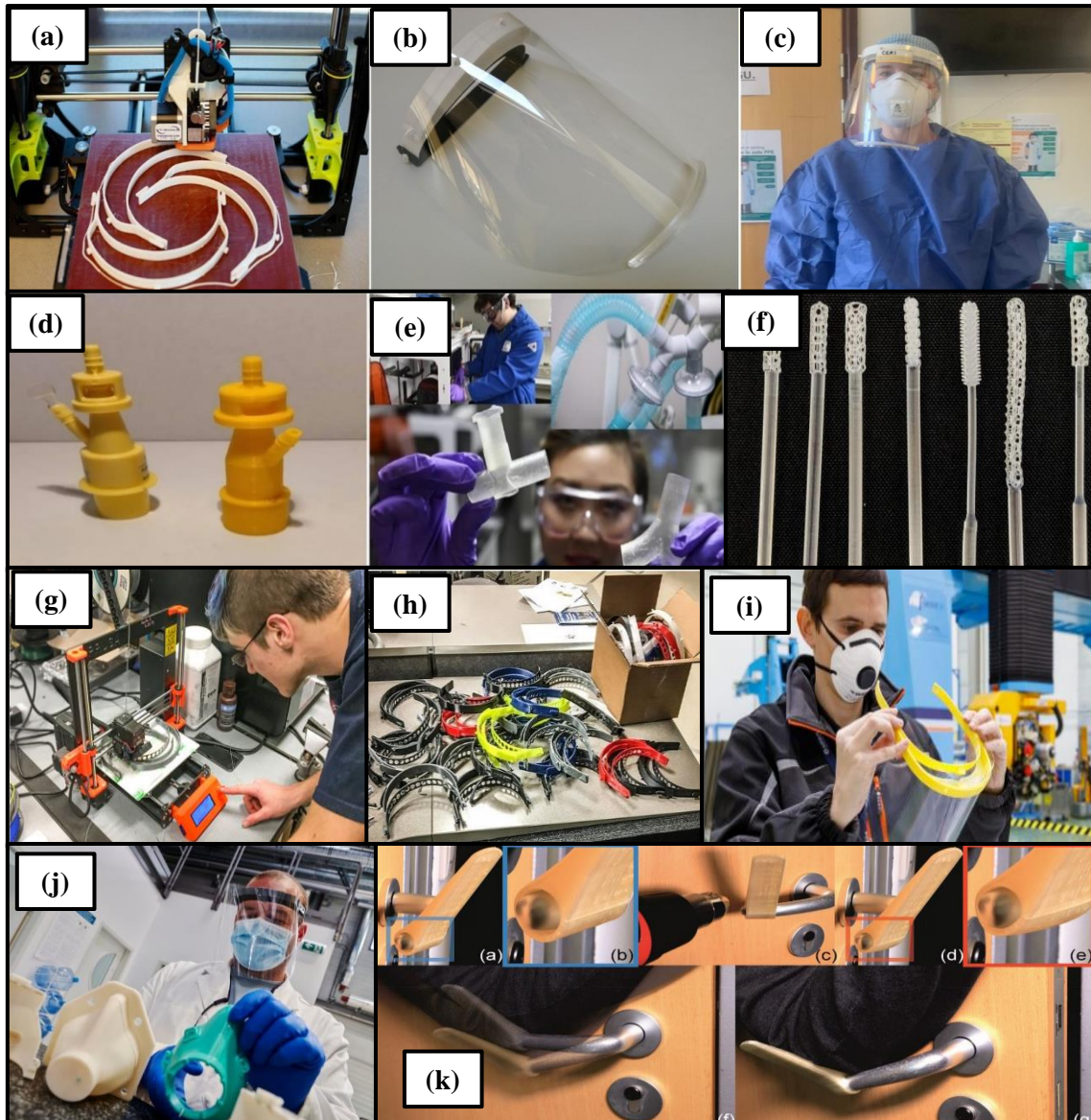


Figure 19. (a) FDM printer produces frames for face shield; (b) Face shield with different components, such as bracket, frame/ headband, and visor; (c) 3D-printed face shield worn by medical consultant; (d) 3D-printed ventilator valve perfectly similar to original valve fabricated from conventional manufacturing techniques; (e) Different T-connector, Y-connector ventilator parts fabricated from 3DP used by multiple patients with using a single ventilator ((a-e) adapted from [325]). (f) 3D-printed testing swabs [326] (adapted with permission); (g) 3DP setup by a technician at Somerset Community College for fabricating personal protective equipment; (h) Partially 3DP completed different face shields. (Photo courtesy Somerset Community College) [327] (adapted with permission); (i) PLA-based 3D printed visors used by majority of Airbus sites in Spain (Copyrights Airbus) [328] (adapted with permission); (j) 3D-printed silicon- based washable face mask developed by CERN [329] (adapted with permission); (k) 4D-printed hand free protective (door handle made from (poly(ether urethane) (PEU)) can be effective in preventing COVID-19 [330] (adapted with permission).

4D printing technology greatly helps in confronting the COVID-19 like crisis by providing smart devices such as soft robots [331]–[333]. This technology can further help healthcare systems by developing soft material-based protective visors, soft robotics, and actuators, which would protect the patients and doctors from pandemic-like situations [334]. These soft robots are extensively applied as smart devices for therapies and surgeries, body-part simulators,

artificial organs, drug delivery systems, and prostheses, which mimic the human body [335]–[338].

6. Summary and future perspectives

4DP, a rapid prototyping and futuristic technology, incorporates intelligent materials and 3DP technology to develop highly precise macro-/micro-scaled robots and actuators [339]. In the last few decades, the soft robotics field has been continuously developing at a rapid pace and these micro/macro-scaled robots can be applied in unlimited engineering applications [340]. However, there is a need to address a few concerns and Figure 20 summarizes the major research hotspots for the 4D-printed robotics field. In the following paragraphs, current challenges and future perspectives are outlined.

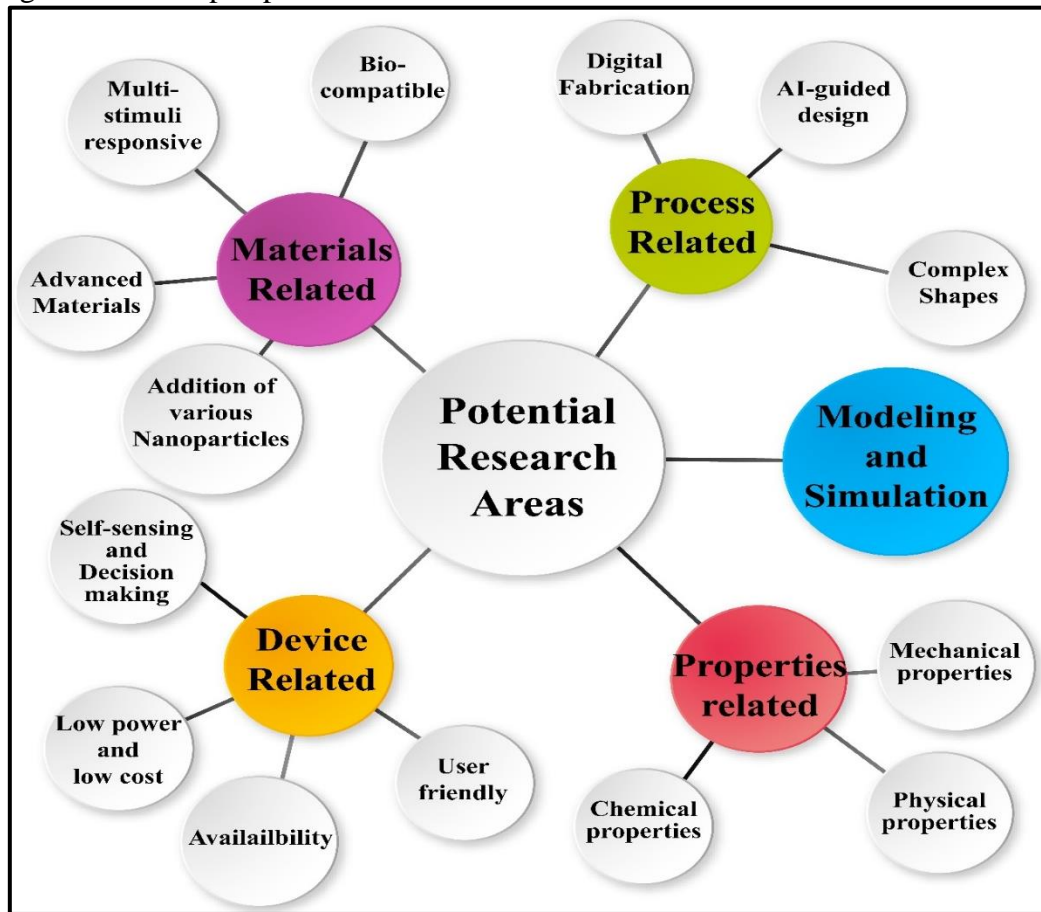


Figure 20. Potential research hotspots and challenges of 4D-printed soft robotics

In the future, the successful development of robotic actuators can be done by generating arbitrary shapes using two-way responsive materials. Furthermore, the integration of different SRMs like light-responsive or magneto-responsive micro-/nano-scaled structures for producing 4D-printed SMC-based components will provide future directions for smart robotic actuators. Thermal stimulus provides an excellent perspective to develop optimized waveguide architectures [341]. The development of novel materials for heater elements along with the optimization of the location and shape of these elements will be helpful in controlling deforming timing. This will improve the remote actuating ability upon heating. Additionally, additional heating approaches like induction heating and microwave heating can be helpful to assess a variety of combinations of stimulus and smart materials.

In the present era, the 4DP approach exhibits unlimited potential in robotics, gripping, and actuating applications [342]. Additionally, these 4D-printed robots are preferred due to the attainment of large structural deformations compared to the traditionally manufactured robots [343]. 4DP technology can also be utilized for fabricating smart devices for underwater applications due to the excellent functionalities of smart materials [344]. Furthermore, SMC-based 4D-printed grippers and actuators have exhibited great potential for holding hot and cold devices, and optical sensors, as well as finding applications in the electronics, telecommunication, and biomedical fields [345]. These composites have also shown excellent piezoelectric properties and can be applied to the knee prosthesis.

In the future, hydrogel-based microrobots can be fabricated through 2PP and photolithography and their synergistic responses to thermal and magnetic stimuli make these robots highly flexible and ultimately prevent leakages during drug delivery [346], [347]. Furthermore, the incorporation of a variety of 4D-printed materials in smart robotics systems such as the set of arms with end-effectors or micro/nano-scaled components assembly will unearth new opportunities. It is a challenging task to integrate manifold sensing and driving modules in the micro-/nano-scaled robots. These 4D-printed robots exhibit an active bionic deformation similar to those of small animals and micro-organisms [348]. These micro-organisms use soft tissues to transform their shapes for predation and locomotion [349], which shows that these 4D-printed soft actuators and robots must possess biocompatibility and remote-control capability for their utilization in clinical applications.

Although tremendous progress has been made in the development of 3D-printed soft actuators and robots, which will enable miniaturization. However, 4D-printed smart materials have not been commercialized yet [350]. In the future, 4D-printed micro-scaled robots will help in developing surgical tools for minimally invasive procedures, micro-stents, smart scaffolds, and adaptive drug-releasing reservoirs. However, the evolution of 3D- to 4D-printed robotics is still in the nascent stage, keeping in view the biomedical perspective. In the future, focused research is required on material aspects of SRMs to improve the morphological behavior of actuators and sensors. Additionally, high-performance multi-functional coupling-actuated soft actuators need to be further explored due to their potential to be used in different emission control methods and their effectiveness in reducing or removing airborne pollutants.

The utilization of 4DP technology is limited in wireless communication due to the limited control over the high-frequency bandwidth complexities [351]. The optimized positioning of 3D-printed actuators and sensors as voxel arrays can improve the performance of the controller. Figure 21 incorporates strengths, weaknesses, opportunities, and threats (SWOT) analysis of 4D-printed robots and actuators.

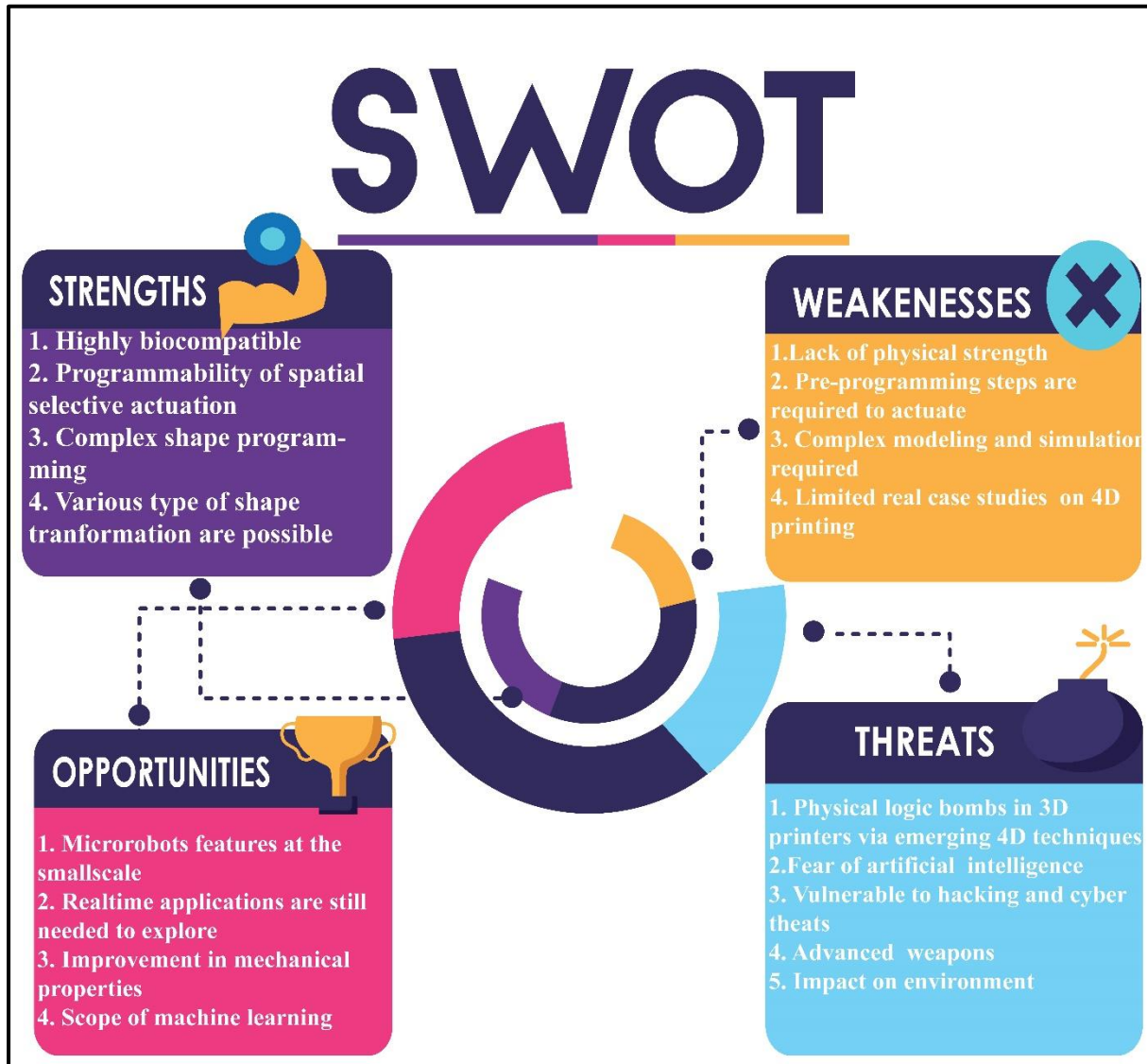


Figure 21 . SWOT analysis of 4D-printed robots and actuators.

The exploration of multi-physics numerical simulations, ML algorithms, and other different control platforms for 4DP technology will permit the adoption of 4D-printed systems in a dynamic environment. Sometimes, SMP-based soft materials undergo bending, which may cause failure/rupture of these materials [352]. There is a need to develop novel SRMs for soft robotics applications for adaptive 4D-printing systems. ML is an effective, powerful, and versatile tool, which is another research spot for the 4DP research community [353]. The integration of ML-based models in the 4D-printed robotics field will further accelerate smart/intelligent materials design. Thus, ML-assisted fabrication technologies will enable the fabrication of multi-functional and adaptive SRMs, smart devices, robots, and grippers for different engineering applications. Furthermore, these models can also sense, adapt, and assess the printing parameters [354], which will be helpful in eliminating the common problems in manufacturing. These ML-assisted 4DPs will enable bioprinting of tissues and organs as well as develop highly efficient soft robotics.

A lot of work is required for soft robotics applications in the future to make 4DP a ubiquitous technology, similar to 3DP technology, by exploring novel and multiple-SRMs and micro-

fabrication methods. Finally, 4D-printed soft robots can change the application prospects of robots in the fields of bioengineering and medical treatment. It is perceived that 4DP technology is a futuristic technology and focused exploration will help in unearthing new potential in the soft robotics field, within the next few years.

Conflict of interest statement

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