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Review

3D printing of magneto-active smart materials for advanced actuators and soft robotics applications

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ABSTRACT

In the contemporary era, novel manufacturing technologies like additive manufacturing (AM) have revolutionized the different engineering sectors including biomedical, aerospace, electronics, etc. Four-dimensional (4D) printing aka AM of smart materials is gaining popularity among the scientific community, which has the excellent ability to make soft structures such as soft robots, actuators, and grippers. These soft structures are developed by applying various stimuli such as pH, temperature, magnetic field, and many combinations onto soft materials. Stimuli in 3D printing permit various shape-morphing behaviors such as bending, twisting, folding, swelling, rolling, shrinking, origami, or locomotion. A wide variety of soft magnetic structures can be fabricated through the incorporation of soft or hard magnetic particles into soft materials resulting in magneto-active soft materials (MASMs). With this integration, magneto-thermal coupling actuation allows diverse magneto-deformations, facilitating the development of personalized devices that are capable of enhanced deformation. In this review, guidelines are provided on the 3D printing for MASMs such as magneto-active polymers (MAPs), magneto-active composites, and magneto-active hydrogels (MAHs) on the booming development of various smart and flexible devices such as soft robots, wearable electronics, and biomimetic devices. Moreover, 3D-printed soft robotics have an outstanding capacity to adapt to complicated situations for many advanced actuating applications. Finally, some current challenges and emerging areas in this exciting technology have been proposed. Lastly, it is anticipated that technological advancements in developing smart and intelligent magneto-active structures will have a significant impact on the design of real-world applications.

1. Introduction

Under constant evolution, the ambition to drive and pursue modern technologies has significantly improved today's living standards. This has happened because of scientific progress, leading towards transformation in many areas including materials, their synthesis techniques, and properties characterization, thus, opening a new paradigm for many novel applications [1]. Three-dimensional (3D) printing or additive manufacturing (AM) is regarded as a novel and emerging manufacturing technique for many materials and it is now being imposed in scale-up on an industrial scale [2–6]. 3D printing is also drawing attention from researchers due to its ability to produce complex parts with higher accuracy, adaptability and availability all over the world [7–10]. Various 3D printing techniques such as ink-based, light-based and laser-based are introduced [11–13] and performed significantly for various materials such as polymers [14], elastomers [15], metals [16], and polymer composites [17]. Ink availability, balancing printing quality including layer thickness, and layer height are some of the important design criteria in 3D printing [18–20]. From a sustainability perspective, 3D printing has so much to offer, for instance, various natural biomaterials [21–23] can be used as a potential ink source for exciting applications without creating any waste [24–26]. Moreover, 3D printing of composite materials has improved mechanical properties than traditional composites [27–29]. This technology has provided the opportunity for multi-material printing which includes two or more different

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Nomenclature

2D	Two-dimensional
2PP	Two photon polymerization
3D	Three-dimensional
4D	Four-dimensional
AAc	Acrylic acid
ABS	Acrylonitrile butadiene styrene
ALG	Alginate
AM	Additive manufacturing
BJ	Binder jetting
CA	Cellulose acetate
CIP	Carbonyl iron particles
CNF	Cellulose nanofiber
CNT	Carbon nanotube
DMAA	N,N'-dimethyl acrylamide
DLP	Digital light processing
DIW	Direct ink writing
DLW	Direct laser writing
FDM	Fused deposition modelling
FFF	Fused filament fabrication
FePt	Iron platinum
FASMC	Flexible anisotropic soft-magnetic composite
GelMA	Gelatin methacryloyl
LCE	Liquid crystal elastomer
MAHs	Magneto-active hydrogels
MAPs	Magneto-active polymers
MASMs	Magneto-active soft materials
MC	Methylcellulose
MJ	Material Jetting
MPs	Magnetic particles
MRE	Magnetorheological elastomers
MNPs	Magnetic nanoparticles
MWCNTs	Multiwalled carbon nanotubes
NdFeB	Neodymium-iron-boron
NIR	Near-infrared
PA	Polyamide
PAA	Poly(acrylic acid)
PAAM	Polyacrylamide
PBF	Powder bed fusion
PCL	Polycaprolactone
PDA	Polydopamine
PDMS	Poly(dimethylsiloxane)
PEEK	Polyether ether ketone
PEG	Polyethylene glycol
PEGDA	Polyethylene glycol diacrylate
PETA	Pentaerythritol triacrylate
PEU	Polyester urethane
PHB	Poly-hydroxybutyrate
PLA	Polylactic acid
PLMC	Poly(D,L-lactide-co-trimethylene carbonate)
PNIPAM	Poly(N-isopropylacrylamide)
PP	Polypropylene
PTMC	Poly(trimethylene carbonate)
PU	Polyurethane
PVA	Poly(vinyl alcohol)
PVC	Poly(vinyl chloride)
SDM	Shape deposition manufacturing
SEM	Scanning electron microscopy
SF	Silk fibroin
SME	Shape memory effect
SMP	Shape memory polymer
SMPC	Shape memory polymer composite
SLA	Stereolithography
SL	Sheet lamination
SLM	Selective laser melting
SLS	Selective laser sintering
SPIONs	Superparamagnetic iron oxide nanoparticle
rGO	reduced graphene oxide
TPR	Thermoplastic rubber
TPU	Thermoplastic polyurethane
UV	Ultraviolet
VP	Vat Photopolymerization

materials as well as solid material into a medium, creating a suspension for desired ink for any geometry [30–32]. Many complex structures such as helical coils, origami, and kirigami-inspired structures, and functionalized micro-architectures can be printed with extreme accuracy [33–38].

3D printing has opened up many interesting avenues for real or practical applications as well as continuously thriving for new platforms for incorporating many emerging materials including nanomaterials for achieving wide goals for a broader community perspective [39–42]. Recently, during the coronavirus disease 2019 (COVID-19) pandemic, 3D printing also played its part by fabricating personal protective equipment [43–45]. Other biomedical applications of 3D printing include patient-specific models that can be used to train medical staff and improve patient consent and understanding, wearable devices such as orthotics and prosthetics [46–48], tissue engineering [49–52], drug delivery systems [53–55] as well as gadgets to make life easier [56–58].

The use of 3D printing is growing in almost every field including analytical chemistry [59], microfluidic devices [60], and detection of analytes for medical diagnosis [61], electrochemical sensors [62], and system health monitoring [63,64]. However, the cost, limited print materials, the need for post-processing of devices [65], and the need for higher resolution still limit the broader application of this technology. One of the significant drawbacks is that printed functions remain static after 3D printing which limits its applications in some of the novel areas where many printed functions, such as self-healing ability, elastic conductivity, and shape-morphing mechanism in many devices (e.g., wearable electronics, soft robots, and flexible biosensors) performances are required [66–69]. Among all these 3D-printed drawbacks, the shape-morphing behaviors of printed materials have paramount importance in advanced engineering applications [70–72]. Lately, an improved form of AM relatively inspired by shape-morphing behaviors in nature, the four-dimensional (4D) printing technique has been introduced [73–75]. 4D printing can also be defined as using smart materials for adopting external stimuli in the 3D printing research division [76–80]. Researchers have developed a 4D printing technique for gaining more accurate control of the shapes of printed parts such as shrinking, swelling, folding, bending, rolling, origami, twisting, or locomotion under various stimuli [81–86].

Recently soft actuators and robotics have been studied extensively [87]. Soft robotics have some unique capabilities in comparison to traditional robots such as constantly changing stiffness and shape morphing ability for performing specific tasks such as grasping and lifting toxic or hazardous objects under extreme environmental conditions [88–91]. In fact, shape compliance of soft actuators provides a viable avenue to address many unsolved problems of today [92–96]. The fabrication of soft robotics through the conventional synthesis route is tedious and time-consuming, and more importantly, its shape-morphing behavior is not satisfactory. To date, various synthesis routes have been used for fabricating soft robotics, including solvent casting [97], lithography [98], roll-to-roll technology [99], laser heating [100],

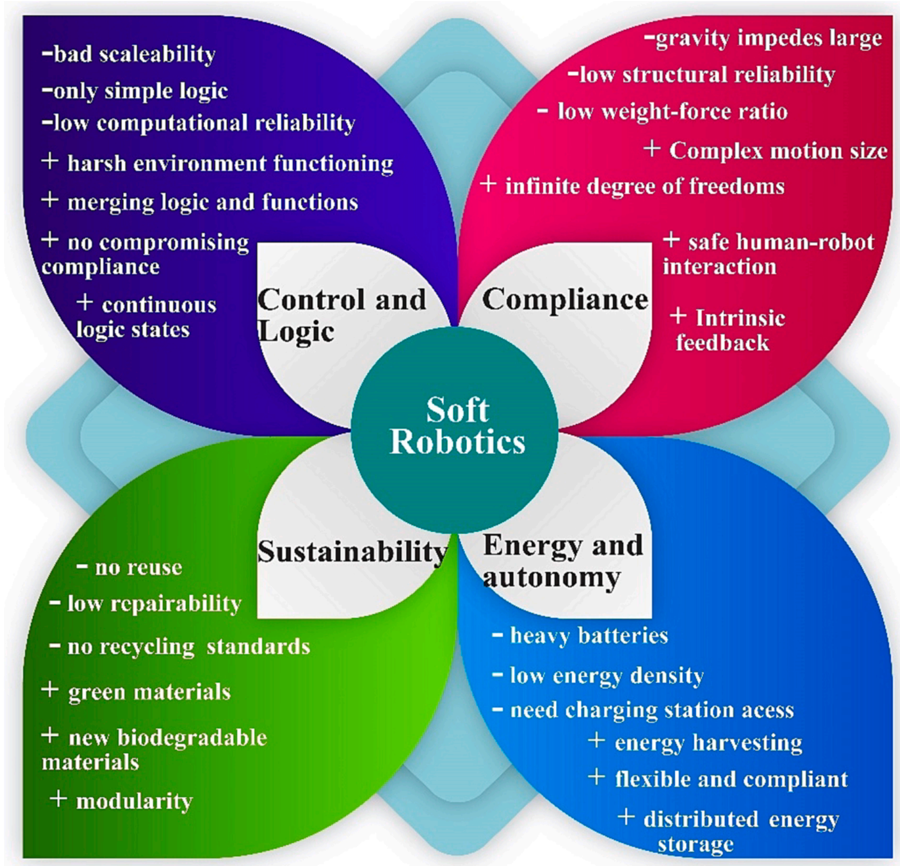

Fig. 1. Overview of recent soft robotics from various perspectives.

spraying with spin technique [101], magnetized modules assembly with dynamic covalent bonds [102], electron beam lithography of nanomagnets [103] and bonding agent [104,105]. Among them adding magnetic particles (MPs) to 3D-printed smart material is a promising and innovative way to achieve highly functionalized soft actuators [106–108]. To date, various magnetic materials such as electrical steel (FeSi), iron oxide (Fe_3O_4) and carbonyl iron particles (CIP) have been added to many shape memory materials. In the presence of magnetic field strength, the 3D-printed magnetic actuated soft robotics exhibited unique phenomena for changing their shape, structures as well and properties which are beneficial to many applications [109]. This unique 3D-printed magnetic actuation attribute with desirable performances is an ideal choice for practical application in the healthcare sector [110] like targeted drug delivery and tissue engineering [111–114]. Moreover, magnetically actuated actuators with remote magnetic steering capabilities have also proven their potential in minimally invasive medical procedures [115–118]. Fig. 1 summarizes the key features found today in soft robotics and their exciting role in many diverse applications.

1.1. Scope of review

Considerable progress has been made in the design of high-performance soft actuators. Herein we have provided some guidelines based on the latest research studies on how to use the power of 3D printing of smart materials in making high-performance novel devices such as smart grippers, wearable electronics, stretchable ionotropic devices, and many intelligent devices from AM techniques, and smart materials point of views. The broad aim of this review is to: i) Stipulate an exhaustive overview of 3D printing of magneto-active polymers, ii) Identify key smart materials employed for magnetic actuation and their key mechanisms for exciting applications, iii) Propose a series of guidelines for tackling future challenges and highlighting existing scientific and technological gaps in the field, and iv) Discuss potential opportunities for fabricating high-performance soft robotics towards practical applications. Fig. 2 shows the publication trends of 3D printing of smart materials under magnetic stimulus across the different years and significantly publication trends proving that there is a need for a systematic review to summarize the novel studies. Furthermore, we

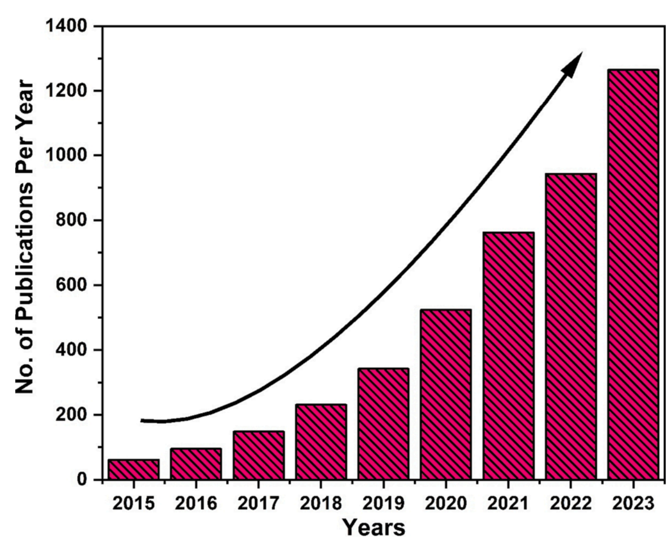

Fig. 2. 3D Printing of magnetic actuated materials publication trends across the different years ((Figure drawn based on the information from Scopus database using “3D printing”, and “magnetic responsiveness” as keywords).

Table 1
Brief comparison between current review and recent reviews on similar topics.

Major Discussion/aspect	Previous reviews					Present review
	Bastola and Hossain [119]	Lucarini et al. [120]	Khalid et al. [121]	Hedge et al. [122]	Yasa et al. [123]	
Discussion on 3D printing	–	✔	✔	–	–	✔
Discussion on Soft robotics	–	✔	✔	✔	✔	✔
3D printing under magnetic stimulus (only) for soft robotic applications	–	–	–	–	–	✔
Dispersion/synthesis of MPs in soft materials	–	–	–	–	–	✔
Magneto characterizations	✔	✔	–	–	–	✔
Sensing capabilities in soft robotics	–	–	–	✔	–	–

develop this review by highlighting the key aspects of various published studies related to this emerging field and adapting a systematic approach for balancing between the 3D printing technology and the performance of printed devices. Table 1 provides a brief comparison between a current review and recently published reviews on similar topics.

1.2. Smart materials for 3D printing

Smart materials can perceive and respond under normal conditions related to their surrounding environment; however, these materials are unable to improve or optimize their response when sudden change has happened in their surrounding environment [124]. Whereas intelligent

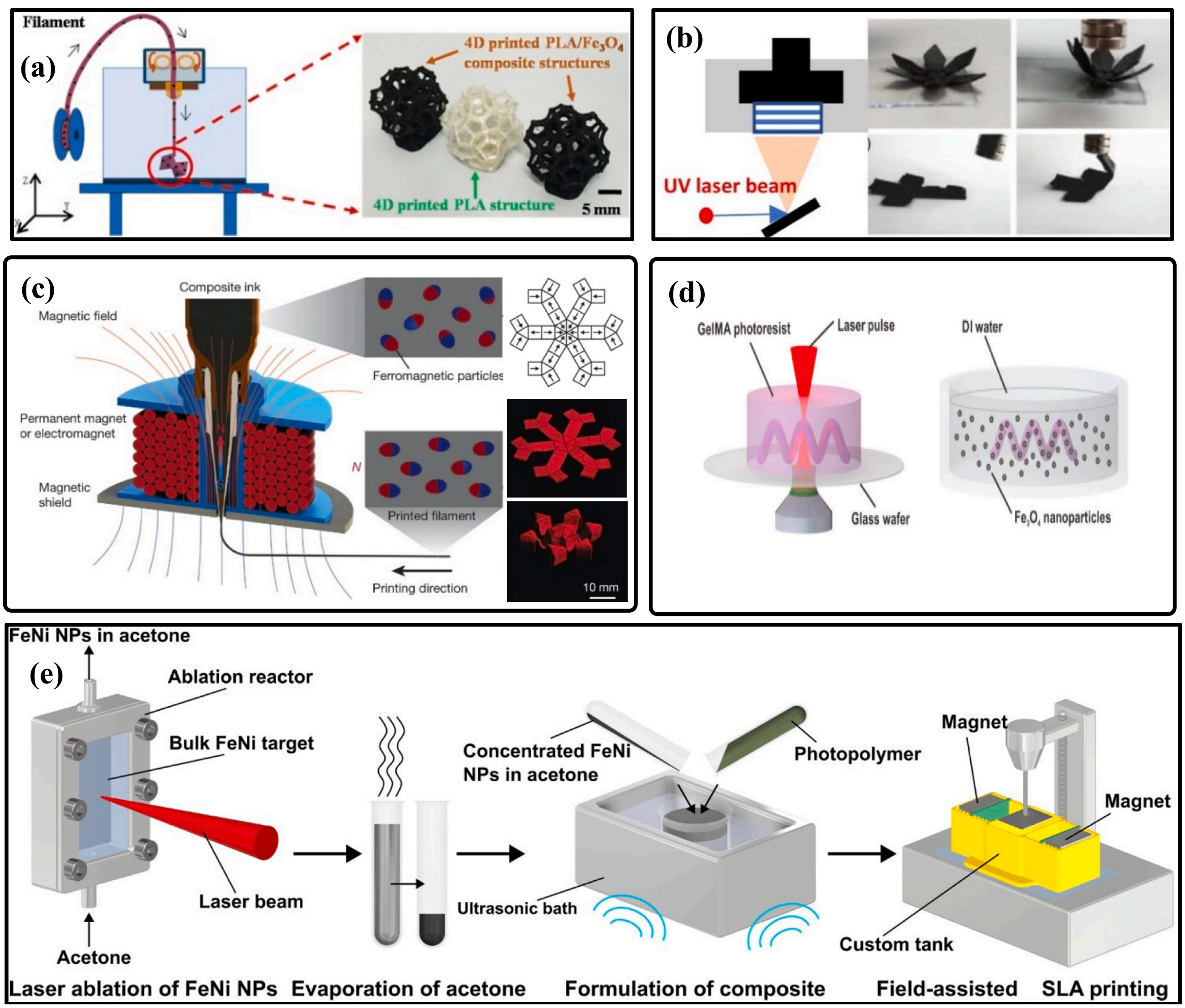

Fig. 3. Schematics of various 3D printing techniques; (a) fused filament fabrication of PLA-based magneto-active composites (adapted with permission from ref. [184], copyright 2019, Elsevier Ltd.); (b) DLP (adapted with permission from ref. [185], copyright 2019, WILEY-VCH Verlag); (c) Design of ferromagnetic domains in soft materials to develop magnetic composites using DIW (adapted with permission from [186], copyright 2018, Springer Nature). (d) TPP used to develop MASMs (adapted with permission from [187], copyright 2018, WILEY-VCH), (e) Masked type SLA technique used for the fabrication MAP structure (adapted from [188] under the terms of the Creative Commons Attribution license 4.0).

Table 2
Comparison of various 3D Printing methods, principles, materials, and cost.

AM processes	Printing principle	Typical polymer materials	Layer height materials	Resolution (μm)	Support structure	Printing cost	Ref.
DIW FDM	Plastic in melt form is extruded through a nozzle	Thermoplastics, hydrogel, liquid polymer, and colloidal suspension	0.050–0.400	100–600 100–150	Dependent on geometry, materials and dissolvable supports can be used	($300) low cost for home use and high for professional use ($2000–$8000)	[189] [190–192]
VP	Laser light or a projected image is used for curing liquid resin	Photocurable resin (acrylate-based resin or epoxy is used)	0.010–0.200	10–50	Dependent on model geometry and printer type	$2500 + for desktop models. $20,000-$200,000 for commercial printers	[193–195]
2PP	Laser light is used for curing liquid resin	Photocurable resins	–	0.1–5	Dependent on 3D geometry	up to $200,000	[196–198]
PBF	Sintering is done through heat-induced	PA, PCL powder and polystyrene	~0.100	–	No	$15,000-$30,000	[199–201]
MJ	Material jetting is done with UV solidification	Photocurable resin	~0.100	Up to 16	No	$100 k-$250 k	[202–204]
BJ	Drop-on-demand BJ	Acrylate-based powder (metal and sand) + bonding agents	~0.100		No	Typically, $200,000+	[205–207]
SL	Adhesive (layer by layer)	Bonding agents + polymer composites	~0.100	0.05–1 (diverse finish)	No	$30,000+	[208–210]

materials can adapt to those changes, and can respond well accordingly and purposefully, for improving and optimizing their response [125]. Smart and intelligent materials are under constant evolution for their applications in various artificial actuators. The motions of these actuators are inspired by nature such as life-like motions for bioinspired robotics [126–128]. Moreover, these materials can offer functionalities beyond traditional ones particularly for developing unique actuators due to their ability to adapt easily and deform according to the environment. Smart materials also include self-healing materials, self-transforming materials, their auxetic behavior, softening and hardening behaviors under compression and tension, action-at-a-distance phenomena and respond overtime to assemble into new compositions via bending, spreading, twisting, shrinking, and folding [129–131]. These dynamic functions of smart materials are teamed with the 3D-printed complex geometries of parts for soft robotics, advanced actuators, biomimetic devices, and self-deployable structures applications [132–134].

Shape-memory materials are the type of smart materials which trigger their response under the environmental stimulus, without relying on the application of an external force [135–138]. Different shape memory polymers (SMPs), liquid crystal elastomers (LCEs), hydrogels, and shape memory polymer composites (SMPCs) are effectively used for the fabrication of flexible devices through 4D printing [139–141]. It is worth mentioning that among all the SMP, SMPC, and the role of multifunctional hydrogels are highly effective in the development of novel smart structures [142–144]. Various two-dimensional (2D) materials such as graphene, and carbon nanotubes (CNTs) can further improve the shape memory effect (SME) of these smart materials [145].

2. 3D printing

In this section, the manufacturing techniques used for smart materials are reviewed according to their popularity, and working principles with pros and cons. Furthermore, 4D printing technology is correlated with 3D printing [146 147,148]. Thus, new possibilities in 4D printing will be created due to the development of 3D printing techniques [149–151]. Typically, 3D printing is considered a bottom-up manufacturing approach, and materials are deposited and patterned in a drop-on-demand manner [152–154]. This allows rapid design and manufacturing of many smart actuators-based various devices [155–157].

3D printing techniques are characterized by contact-based and contactless methods. Fused deposition modelling (FDM), material jetting (MJ), and direct ink writing (DIW) come under contact-based methods [158], whereas the photopolymerization process, powder bed fusion (PBF), and direct energy deposition, are common contactless technologies for 3D printing [159–161]. Of all these techniques, stereolithographic (SLA) and FDM are the most employed processes. FDM includes high-temperature nozzles for feeding the filament, and later depositing layer-by-layer sheets of a melted layer with high fabrication speed [162]. FDM also has significant advantages such as versatility and affordability for all types of structures (small to large) and less expensive 3D printing techniques [163–166]. Moreover, a wide variety of inks in DIW can be deposited onto arbitrary substrates with random or even complex geometries. Thus, sometimes it is interpreted as a powerful technique for fabricating advanced and sophisticated electronic equipment with high resolution [167–169]. However, the possibility of needle clogging during the low speed and high shear forces are some major drawbacks of FDM [170]. Fused filament fabrication (FFF) is also considered as simplest and most widely used 3D printing technology for a large variety of thermoplastic materials at low cost for multi-material 3D printing for various applications [171]. Another popular 3D printing commonly employed is SLA. It has customizability and the ability to print complex geometries through the step method of photo polymerization, scanning the liquid UV-curable matter with a laser [172–175]. This permits high print resolution and excellent speed that may be greater than FDM. Furthermore, SLA is extremely suitable for the fabrication of customized soft robotics for wearable applications [176–178]. Fig. 3 illustrates the working principles of various AM technologies, which are used to print MASMs. Moreover, increasing miniaturization and higher demand for microfabrication scale has diverted the attention of researchers towards micro and nano-printing techniques [179] such as two-photon polymerization (2PP) also referred to as direct laser writing (DLW) [180]. In this technique, a photo-reactive resin is exposed to high-energy femtosecond laser beams and provides excellent spatial resolutions in the range of 100 nm [181–183]. Table 2 highlights the key aspects of current AM technologies. Table 3 summarizes the key benefits achieved by soft robotics using AM technologies.

2.1. 4D printing

Considerable progress in 3D printing technology was achieved by MIT researchers in 2013 by introducing a shape-morphing capability into 3D-printed objects termed 4D printing [228]. It was made possible by the rapid expansion of smart materials, commercial 3D printers, and

Table 3
Some highlights/prominent works for soft robotic using 3D printing technology.

AM technique	Material(s)	Layer creation technique	Size	Soft robotics type	Highlights	Ref.
SLA	Glucose/CNT/PDMS	3D-printed PDMS substrate with CNT layer	15 × 15 × 5 mm^3	Soft wearable sensor like volcano sponge	The facile 3D-printed soft sensor successfully captures speech signals, pulse signals, tactile signals from a mechanical gripper, and gesture signals, for potential applications in medical diagnosis and soft robotics.	[211]
Inkjet Printing	Tangoblack	Multi-material layer by layer printing	14 × 9 × 7 cm^3	Bellows actuators, gear pumps, soft grippers and a hexapod Robo	The proposed 3D printing allows robotic components to be automatically built, with no assembly required.	[212]
Connex3 Objet350 3D printer	(TangoPlus FLX930), (TangoBlackPlus FLX980) and (VeroClear RGD810)	Multi material layer UV-curable	–	Soft gripper with embedded sensors	The proposed 3D-printed soft gripper with embedded sensors has resistive sensing capabilities directly into a pneumatic gripper.	[213]
FDM	TPU	Multi material layer	40 × 12 × 0.55 mm^3	Smart soft grippers	The proposed multi-material printing has enormous scope in the automation industry for fabricating on-demand smart universal gripper with variable stiffness and integrated sensors.	[214]
DLP	Soft conductive resin	–	–	Soft actuators	DLP-based printed untethered soft actuators embedded with multiple sensing capabilities are highly promising for intelligent soft robotics applications.	[215]
FDM	TPU	–	23724.82 mm^3	Omni-purpose soft gripper	The proposed 3D-printed soft gripper has a maximum payload to weight ratio of 7.06, a grip force of 31.31 N, and a tip blocked force of 3.72 N and can grasp at least 20 different objects.	[216]
FDM	TPU	Layer-by-layer printing	–	Origami-based soft encapsulating gripper	The direct 3D printing of soft materials on fabric is highly promising for soft actuators with grasping performance are highly delicate and ultra-gentle objects.	[217]
2PP	Propylene glycol methyl ether acetate (PGMEA)	Multi-material laser curable printing	4.9 × 10^{-4} mm^3	Micro-hydraulics soft actuator	The proposed micro printed actuator could transmit forces with relatively large magnitudes (millinewtons) in 3D space for broader applications in micro-robotics and medical.	[218]
DLP	TPU	Layer-by-layer UV-curable	4.5 × 12 × 6 cm^3	Frog-shaped soft robot	DLP-based 3D-printed soft actuators (2.2 g) could exert up to 0.5 Newtons of force that are integrated into a bioinspired untethered soft robot.	[219]
SDM	PU	–	116 cm^3	Soft, atraumatic and deployable surgical grasper	The proposed SDM fingers were used to design a multijointed grasper that relies on geometric trapping to manipulate tissue, which was a highly conformable means of manipulation	[220]
FFF	NinjaFlex (NinjaTek)	–	49.7 × 47.7 × 12.5 mm^3	Monolithic soft gripper with adjustable stiffness	Finite element simulation and experimental results showed that the proposed monolithic 3D-printed soft gripper is fully compliant, low cost and requires an actuation pressure below −100 kPa.	[221]
DLP	Polyurethane acrylate	Multi-material UV-curable printing	500 × 300 µm	Dielectric elastomer actuators for vibrotactile device	The non-prestretch DLP-printed cylindrical actuator demonstrated a remarkable blocked force of 270 mN and maintained 45 % actuation performance at a frequency of 100 Hz.	[222]
SLA	2-hydroxyethyl acrylate, ethylene glycol diacrylate, and phenyl bis (2,4,6-trimethylbenzoyl) phosphine oxide	Multi-material UV-curable printing	500 × 500 × 500 $µm^3$	Multifunctional structured microgel as building blocks for mesoscopic self-assembly	The 3D-printed mesoscopic microgels were assembled and disassembled using respective reduction and oxidation reagents for soft robotic applications.	[223]
FDM	PVC sheets	–	–	Soft prosthetic finger	The reported results showed that the stiffness of the 3D-printed soft finger was increased by 40 % by linearly driving the stiffness augmenting unit.	[224]
Inkjet Printing	Urethane and epoxy	Multi-material UV-curable printing	80 × 5 × 5 mm^3	Tri-legged soft robot with spider mimicry	The developed tri-pedal soft bot demonstrated its power efficiency and controllable locomotion at three input signal frequencies (1, 2, and 5 Hz).	[225]
FDM	Nafion	Layer by layer	5 mm × 10 mm × 0.5 mm	Macro-scale soft robotic systems	The proposed 3D printing of ionic polymer-metal composites exhibited unique actuation and sensing properties for creating electroactive polymer structures for application in soft robotics.	[226]
Polyjet-based 3D printing	–	Multi-material printing	30 µm (layer height)	Unified soft robotic systems comprising a fully integrated fluidic circuit	The fully integrated soft robotic entities consisting of soft actuators, fluidic circuitry, and body features offer a novel way to catalyze new classes of soft robots.	[227]

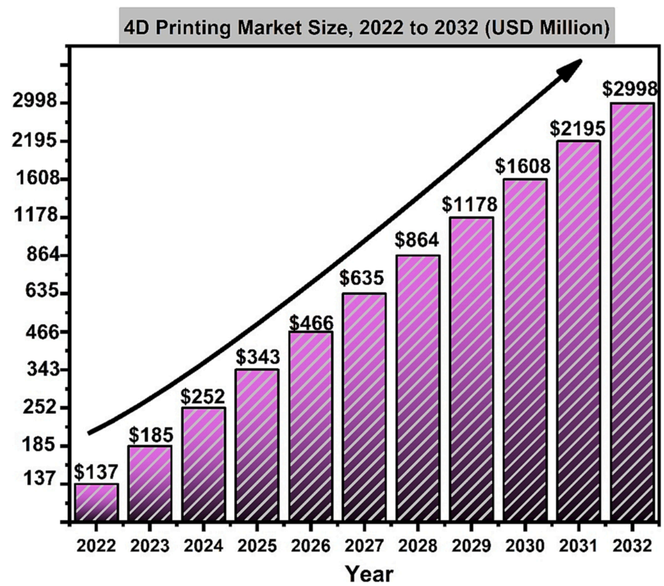

Fig. 4. 4D printing market trends in the upcoming years (Figure drawn based on the information is collected from online available source available at [254]).

stimulant environments such as light, temperature, pH, humidity, magnetic and electric fields [229]. 4D printing enables a higher degree of freedom and flexibility in terms of printable geometry [230–232]. Moreover, 4D printing integrates the product's blueprint into a flexible, and intelligent material [233–235]. The term "4D" refers to alive structures obtained from traditional 3D-printed structures and means the printed structure can change at least one of its key features such as design, color, property, or functionality over a period under a stimulant environment [236]. This opens a new paradigm for new application arenas for their multi-functional behavior including SME, complex rapid deformation requirements [237], reconfigurable structure, actuation, and sensing under stimulant environments for a broad variety of applications such as soft robotics [238], shape-memory structures [239], advanced actuators [240–242], tissue engineering [243], targeted drug delivery [244,245], cell-laden structures [246], self-deployable structures for aerospace applications [247–249], and many more [250–253]. Fig. 4 shows the 4D printing market forecast in the upcoming years.

Smart or stimuli-responsive materials have contributed towards 4D printing by integrating existing 3D printing techniques [255–257]. The smart materials in 4D printing are classified into many sub types such as thermosets and thermoplastic polymers [258,259], various biomaterials [260,261]. Polylactic acid (PLA) [262,263], polyvinyl alcohol (PVA) [264], polycaprolactone (PCL) [265], polyurethane (PU) [266], and hydrogels [267] are mainly considered smart materials for fabricating highly responsive soft actuators at both the macro as well as micro levels [268]. 4D printing further harnesses the fabrication of soft actuators, controllable structures, soft robotics, and many functional devices [269–271].

4D printing brings exciting functionalities to smart sensors including environment self-adaptation, self-sensing, and self-healing [272–275]. Recently, Ren et al. [276] introduced a highly versatile smart tactile sensor through 4D printing using nanocarbon black/PLA composites and shape-memory PU. These sensors demonstrated unique adjustable measuring range and sensitivity by changing the electrode height and spacing produced by the SMP deformation under heat treatment. The shape-changing tactile sensor is regarded as an ideal match for producing self-adjustment and self-adaptation for human-robot cooperation in sensing. To date, various emerging materials such as LCE and different hydrogels are used in 4D printing [277–282]. Fig. 5 depicts the emerging applications of 4D printing for sensors and actuator applications. For example, many hydrogels such as polydimethylsiloxane (PDMS) swelled anisotropically under multiple stimuli in an assembly of bistable elements [283,284]. However, these bistable elements need to be exposed to a mechanical load for their second stable state. Sometimes, mechanical intervention is also imperative for switching the second stable state of these materials to activate the snap-through capacity [285–287]. High-performance printing inks are a key factor for temperature-

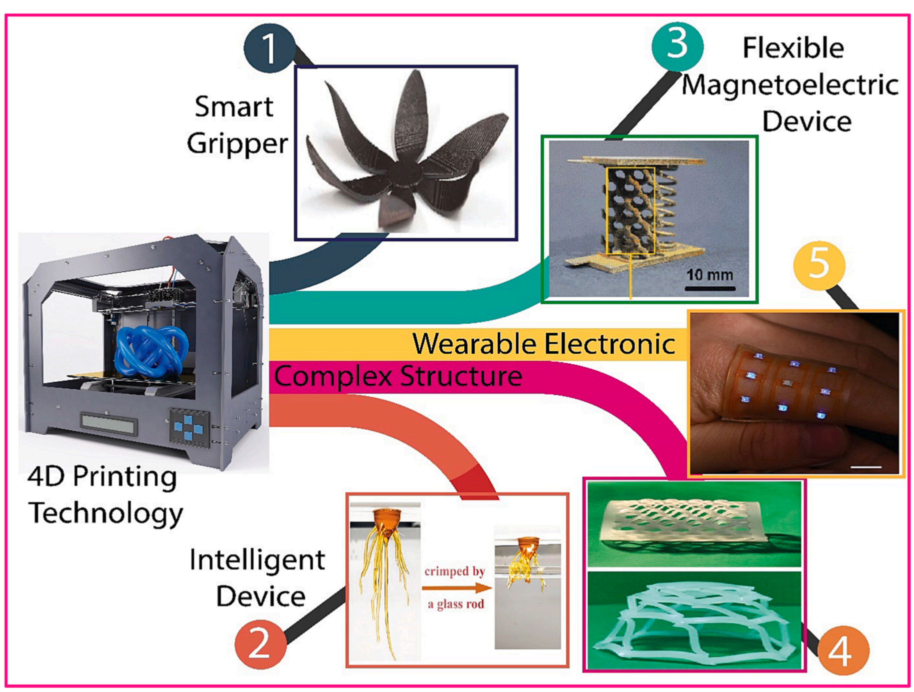

Fig. 5. Recently 4D printing technology was used for various advanced sensors and robotics applications. The figure is drawn based on the various figures collected from (1) Smart grippers by Keneth et al. [293] (Copyright 2023 Elsevier B.V.) (2) Intelligent devices by Lie et al. [294] (Copyright 2022 American Chemical Society), (3) Flexible magnetoelectric devices by Wu et al. [295] (under the terms of the Creative Commons Attribution license 4.0), (4) Complex Kirigami inspired structures by Li et al. [296] (Copyright 2023 American Chemical Society), and (5) Wearable electronics by He et al. [297] (Copyright 2022 American Chemical Society).

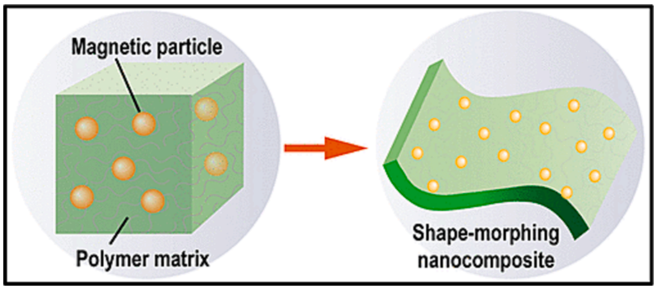

Fig. 6. Shape-morphing soft magnetic materials containing MPs into the polymer matrix (adapted with permission from ref. [310], copyright 2023, American Chemical Society).

sensitive materials, which produce a response aligned with outer temperature change [288–290]. For developing highly flexible electronic devices, temperature-dependent materials are commonly utilized, which generate resistance changes under the temperature change either regular positive or negative responses, for example, the conductivity of typical electronic semiconductors, conductors, and ionic conductors [291,292].

3. Magneto-active soft materials for 3D printing

Magneto-active materials are prominent smart and intelligent materials that can change their mechanical properties like damping, elastic, and shape in the presence of an external magnetic field [298–301]. These materials consist of two major constituents: magnetic fillers and non-magnetic matrix. Based on the host polymer matrices, magneto-active materials are further classified into magneto-active solids and magneto-active fluids [302]. These functional materials offer large deformation, tunable mechanical properties, fast response, and non-contact response [303–305]. Shape-morphing soft magnetic materials are types of smart materials extensively applied for broad applications in soft robotics, sensors, actuators, and other biomedical devices for achieving complicated shape programming [306], as illustrated in Fig. 6. These soft magnetic materials in which soft polymer matrix contain MPs that permit rapid shape transformation reversibly and remotely [307–309]. This section illustrates the different MASMs, which are used to develop soft robots.

Why is magnetic actuation important? Out of all potential stimuli, magnetic triggering and actuation are particularly attractive due to fast complete non-contact interactions [311], wireless nature, and controllable actuation, miniaturization potential and safe interaction with tissues from a biomedical perspective [312–314]. Moreover, magneto-actuated materials show anisotropic stiffness change, even under a relatively small range of stiffness change, while their competitive electro-actuated materials usually work at higher voltage stimulation with higher energy consumption and safety risks [315]. Thus, combining all these advantages offered by magnetic actuation, MASMs through 3D printing are receiving higher attention in novel fields such as soft robotics and flexible electronics [316]. Magnetically driven miniature soft robots demonstrated fast and dexterous responses under the magnetic stimulus [317]. This magnetically induced recovery process is accomplished by inductive heating in an alternating magnetic field [318]. Fe_3O_4-based magnetic microparticles or magnetic nanoparticles (MNPs) are usually incorporated into soft materials to activate the magnetic response [319]. Thus, the fast, reversible actuation and remote manipulation of MASMs are promising for achieving the controlled navigation of soft robots in making the next generation of biomedical devices operating in demanding applications, such as the human body including biosensing, micro-manipulation, and targeted drug delivery [320–323]. Recently, these materials have been proposed for micro-pillar array chips for droplet manipulation applications due to their strong penetrating power [324].

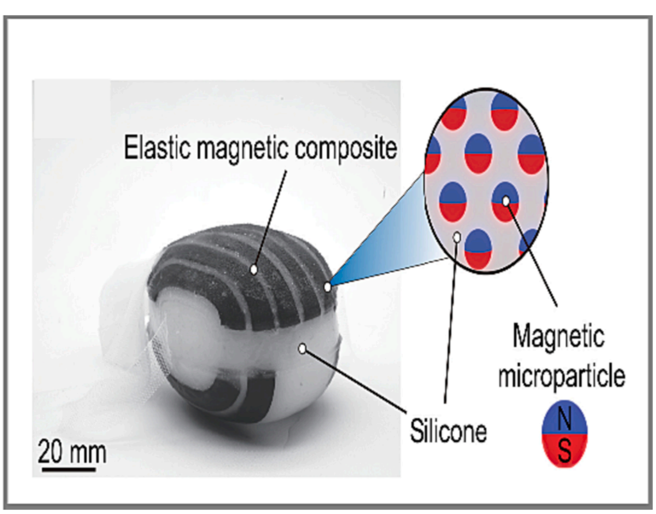

Fig. 7. Magneto-responsive composites composed of MPs and pure silicone are used to develop soft bladder robots for assisting urination (adapted from ref. [344], under the terms of the Creative Commons Attribution license).

Mixing/dispersion of MPs: MPs containing soft material can show isotropic or anisotropic characteristics depending on which fabrication technique is adapted. The fabrication of magneto-active soft composites containing MPs undergoes a curing procedure to stiffen the soft materials [325]. For instance, if the elastomers are cured in the presence of an external magnetic field, the magnetizable particles tend to form chain-like arrangements lending an overall directional anisotropy to the material such materials demonstrated that anisotropic magnetic soft material tend to have stronger coupling with the external magnetic field [326]. It is also crucial to remove gas bubbles as much as possible to prevent cavitation issues. Usually a maximum of 40 % (volume fraction) of MPs, the percolation threshold is achieved in soft polymers [327]. Moreover, along with MPs plasticizers are usually added to enhance mechanical interactions between the dispersed phase and the soft matrix. This is worth mentioning that if an external magnetic field is applied during the curing, the resulting material will be anisotropic because MPs migrate reaching the lowest energy state and therefore more likely to be used in engineering applications. However, if no external magnetic field is applied during the curing process, the resulting material is isotropic. Recently, Garcia-Gonzalez et al. [327] showed that the PDMS-based soft polymer and the platinum catalyst-based crosslinker were put together in such a way that the matrix chains increased their crosslinking degree. Insights of this study showed that a preferred direction of the CIP particles aligned with the field was achieved demonstrating more mechanically stiffer behavior of PDMS/CIP material along a magnetic field direction.

3.1. Magneto-active polymers

Magneto-active polymers (MAPs) usually contain MPs within the soft polymer matrix, which triggers the application of magnetism [328]. These polymers are synthesized by uniform or non-uniform distribution of MPs within the non-magnetic polymer matrix before the curing [329–331]. Additionally, these particles can be aligned in a desirable direction upon the application of a magnetic field during the solidification process. MAPs are also referred to as magneto-sensitive polymers, magneto-active elastomers, magneto-sensitive elastomers, or magneto-rheological elastomers. Based on the hysteresis loop of MPs and their coercivity, MAPs are further classified into hard MAPs and soft MAPs [332–334].

The MPs of soft MAPs have a low magnetic coercivity and these particles do not adequately reserve the magnetization under a null external magnetic field [335]. Some common examples of these MPs

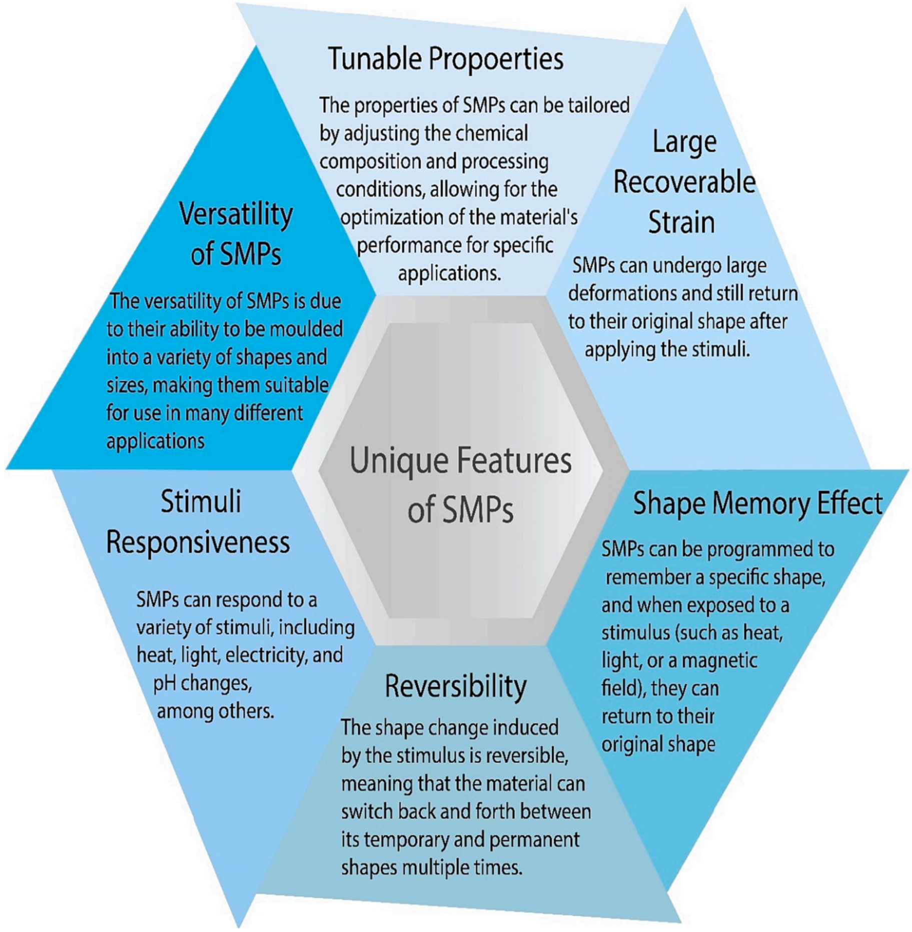

Fig. 8. Prominent features of SMP enabling 3D printing of smart materials (Figure drawn with the help of ref. [353]).

include a Si-Fe alloy and Fe-Al series of alloys. In these polymers, MPs move due to dipole–dipole interactions between particles in the presence of a magnetic field [336]. Such movements and rearrangements of MPs introduce some internal stresses that induce deformations and change the mechanical properties. Soft MAPs can only help in achieving simple and limited actuation for soft robotics applications [337]. On the other hand, the MPs of hard MAPs featuring high coercivity like neodymium–iron–boron (NdFeB) can sustain magnetism even after the removal of an external magnetic field. Consequently, upon applying a further magnetic field, these particles tend to align themselves in the field direction, introducing internal torques within these responsive polymers [338]. Therefore, hard MAPs are preferred for soft robotics applications, as the relatively stable magnetism of these polymers permits directly amendable magnetic fields to generate specific programmable responses [339–341].

Magneto-active composites are soft and flexible composites which are fabricated by embedding a certain ratio of hard or soft MPs into a soft elastomeric matrix such as polyurethane rubber, silicone or gels, as illustrated in Fig. 7. These composites offer dynamic control of mechanical properties through the magnetic field stimulus [342]. These composites are either isotropic with random orientations of MPs cured without an external magnetic field or anisotropic with properly aligned MPs under the applied magnetic field to ensure higher magnetic attraction forces. These composites can quickly deform and transform their shapes, upon the application of varying magnetic fields for achieving bending, twisting, and expansion in a controlled and untherered way [343].

3D printing of magneto-active soft composites can be useful for producing soft structures with good mechanical properties. Nowadays, magnetorheological elastomer (MREs) composites which are filled with MNPs such as CIP, and Fe_3O_4 exhibit tunable rheological and viscoelastic properties for meeting the demand of novel applications such as soft robotics, self-deployable structures, actuating damping devices, vibration isolators, medical inserts, and flexible electronics [345–347].

3.1.1. Shape morphing magneto-active composites

Shape morphing magneto-active composites contain both shape memory and magneto-active properties and can be fabricated using 3D printing technology [348–350]. These composites demonstrate excellent shape programming behavior upon the application of an external magnetic field [351]. Magnetic filled SMPs can be both spatially and temporally activated and allow external noninvasive control of movement [352]. Fig. 8 shows some prominent features of SMP enabling its smart behavior and promising feedstock of 3D printing.

SMP-based composites are highly tunable for controlling many shape memory properties [310]. For instance, the addition of various 2D materials such as graphene, CNTs, manganese dioxide (MnO_2), iron oxide and silver nanowires etc, multifunctional features such as robust self-adhesion, feasible 3D printability, rapid self-healing ability, and electrical conductivity of composites can be improved for developing novel wearable devices [354–358]. Moreover, various SMPCs such as citric acid-based SMPC, polyester urethane (PEU), acrylamide, N,N'-dimethyl acrylamide (DMAA), ethylene glycol, dimethacrylate, and silicone: Ecoflex and silicon elastomer are commonly employed in combination with each other and some other materials as a potential SMPC [359,360]. The interest in 3D printing of SMPC is steadily growing in many fields covering soft robotics biomedical devices, and flexible electronics [361]. Most of the SMPCs are based on the magnetic stimulus

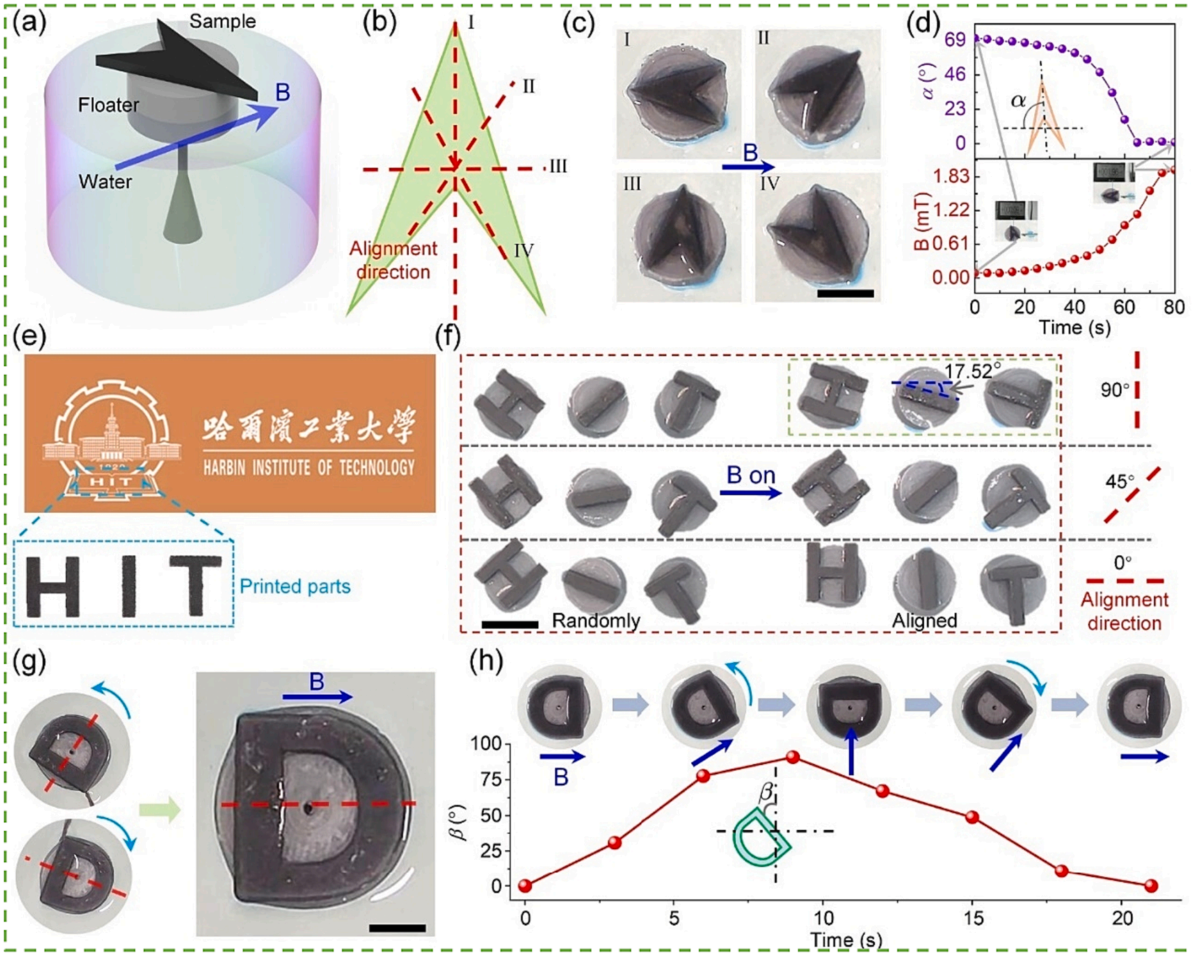

Fig. 9. FASMC rotating actuation (a) the compass with arrows rotates freely rested on fluid. (b) Chain directions of CIP inside the FASMC, (c) The 5 wt% CIP arrow samples driven by a magnet. (d) The angle difference under magnetic field, (e) Three 3D-printed letters 'H', 'I' and 'T' producing an array in (f), at 90°, 45° and 0°, when CIP alignment direction in 1st, 2nd and 3rd row respectively, (f) letter arrays randomly oriented under magnetic field, (g, h) Rotation of samples under testing (adapted from ref. [363], under the terms of the Creative Commons CC BY license).

by embedding MNPs into the polymer matrices, usually ferrite and soft magnetic materials. The shape of SMPCs can be conveniently adjusted by applying an external magnetic field to achieve various characteristics including facile controllability, rapid response time, and reversible behavior for broad application prospects [362]. Recently, Wu et al. [363] prepared a flexible anisotropic soft-magnetic composite (FASMC) through DLP-based printing using flexible long-chin acrylic resin monomer and soft CIP-based MNPs. Insights of this study showed that multiple complex structures of FASMC with strong anisotropic magnetic properties exhibited large deformation, controlled motion, anti-deflection, variable stiffness metamaterial, and array assembly, as depicted in Fig. 9. These behaviors of FASMC are particularly attractive when targeting next-generation sensors and actuators with superior magnetic properties in one or more specified directions.

Soft magnetic composites have been orderly deposited using an advanced 4D printing technique to build deformable actuators under low-strength magnetic field [364]. Reisinger et al. [365] introduced a novel technique for controlling the temperature of dynamic bond exchanged in covalently crosslinked polymer networks. Later, light-mediated curing was used for printing various functional objects, as presented in Fig. 10(a_1), through DLP-based 3D printing, with spatially controlled reshaping capabilities. Furthermore, fiber-reinforced, and highly filled magneto-active thiol–ene polymer composites were effectively used for on-demand activation of dynamic transesterification with various reshaping capabilities (referring to Fig. 10(a_2)), which gives rise to the potential use of 3D-printed magneto-active materials in various active and soft devices.

In another novel study, encoding of various shapes and forms by magneto-/electro-active SMPC structures was explored using carbon black-filled conductive PLA and iron-filled magnetic PLA through FDM [366]. The shape recovery technique was exploited under temperature and the magnetic field for a unique composite actuator was investigated. Results proved that the 4D-printed composite actuator achieved a maximum bending angle of 59° under a low external magnetic field and was fast enough to revert to its original shape when powered by a power supply, as presented in Fig. 10(b_1)-Fig. 10(b_4). This research proved that the 4D-printed composite actuator strategy has broad application prospects in the field of soft robotics by keeping in line with sustainability rules.

3.2. Magneto-active multifunctional composites

The world is continuously exploring novel smart materials with more versatile functionalities [367]. As a result, it is a promising initiative to integrate the advantages of multi-active ingredients into a single material or structure, through monolithic [368] or layered forms [369]. Compared to conventional MAPs, magneto-active multifunctional composites can developed by integrating the advantages of LCEs and MREs [370–372]. For instance, LCEs exhibit high work density and large strains (up to 400 %) to multiple environmental stimuli like heat, light, and electric field [373,374]. Valiant efforts were made by researchers to combine the distinct features of LCEs and MREs for developing soft

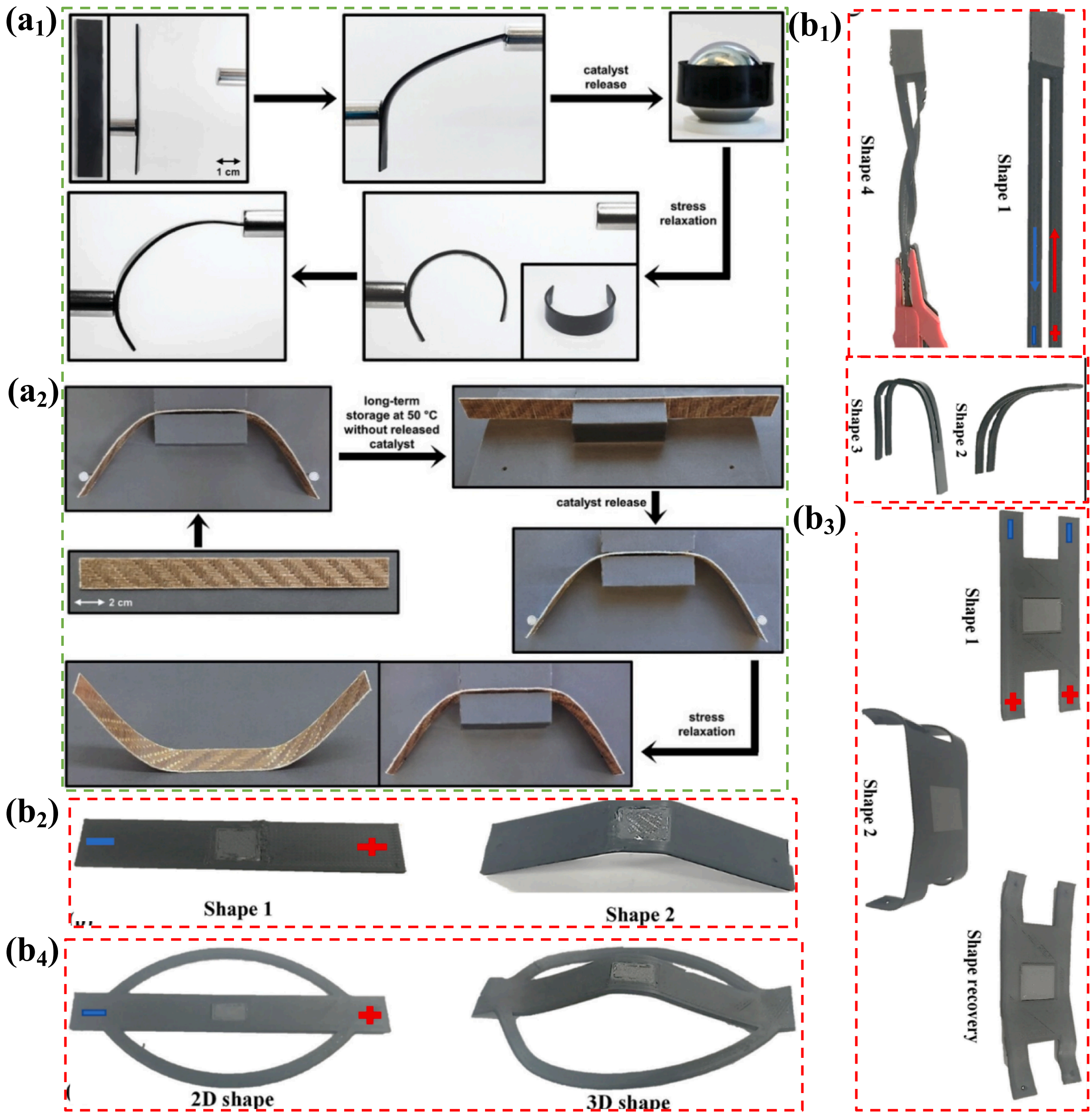

Fig. 10. (a_1-a_2) Permanent reshaping of composite structures, (a_1) Magnetically assisted reshaping of a Fe_3O_4 particulate composite, (a_2) Reshaping of a fiber-reinforced composite (adapted with permission from ref. [365], copyright 2023, Wiley-VCH GmbH); (b_1) Different shapes of a 2D U-shape materials, (b_2) Transformation of 1D beam shape to 2D shape under 60 V power supply and the permanent magnet, (b_3) Conversion of a 2D rectangular shape into a 3D structure (93 % shape recovery), (b_4) Programming a 2D pyramid into a 3D structure (adapted from ref. [366], under a Creative Commons Attribution 4.0).

materials with enhanced and unparalleled functionalities [375–379]. For instance, Zhang et al. [377] developed an untethered miniature 12-legged robot, via a facile fabrication process (casting and soft lithography) by integrating three distinct configurations of LCEs and MREs, as illustrated in Fig. 11(a). The results revealed that this robot responded to wireless stimuli of a controlled magnetic field and surrounding temperature. Thus, complex shape morphing behaviors with anisotropic material properties can be achieved by using the multi-responsiveness of these soft composites. Similarly, Zhang et al. [378] developed a multi-responsive actuator with accurately controlled deformation through the integration of MREs and PDA-coated LCEs. This facile material-structural synergetic design triggered complex and multimode programmable deformation including shrinkage/bending, bidirectional bending, twisting/bending, and rolling/bending. Additionally, this shape-morphing behavior could also be manipulated locally and sequentially, thanks to its photo-sensitive feature.

These soft composites can also be used to develop multifunctional structures with synchronous color-changing and shape-morphing properties such as biomimetic camouflage devices. For instance, Li et al. [379] reported a versatile and facile strategy to develop reconfigurable thermochromic biomimetic structures, such as chameleon and butterfly, as illustrated in Fig. 11(b). The single biomimetic structure contained a combination of LCEs, and MREs embedded with multiple color-changing dyes, which enabled the thermo-magnetic dual response of an octopus structure along with a camouflage feature. This response helped it to achieve adaptive and diverse biomimetic motions (rotating, rolling, swimming, and crawling), accompanied by a color camouflage. Thus, multifunctional magneto-active soft composites are highly suitable to fabricate bilayer multi-stimuli actuators capable of complex and accurately controlled deformations, and these actuators can be used in versatile fields including biomedical, camouflage, and soft robotics.

Nowadays, multifunctional magneto-active bilayer structures can

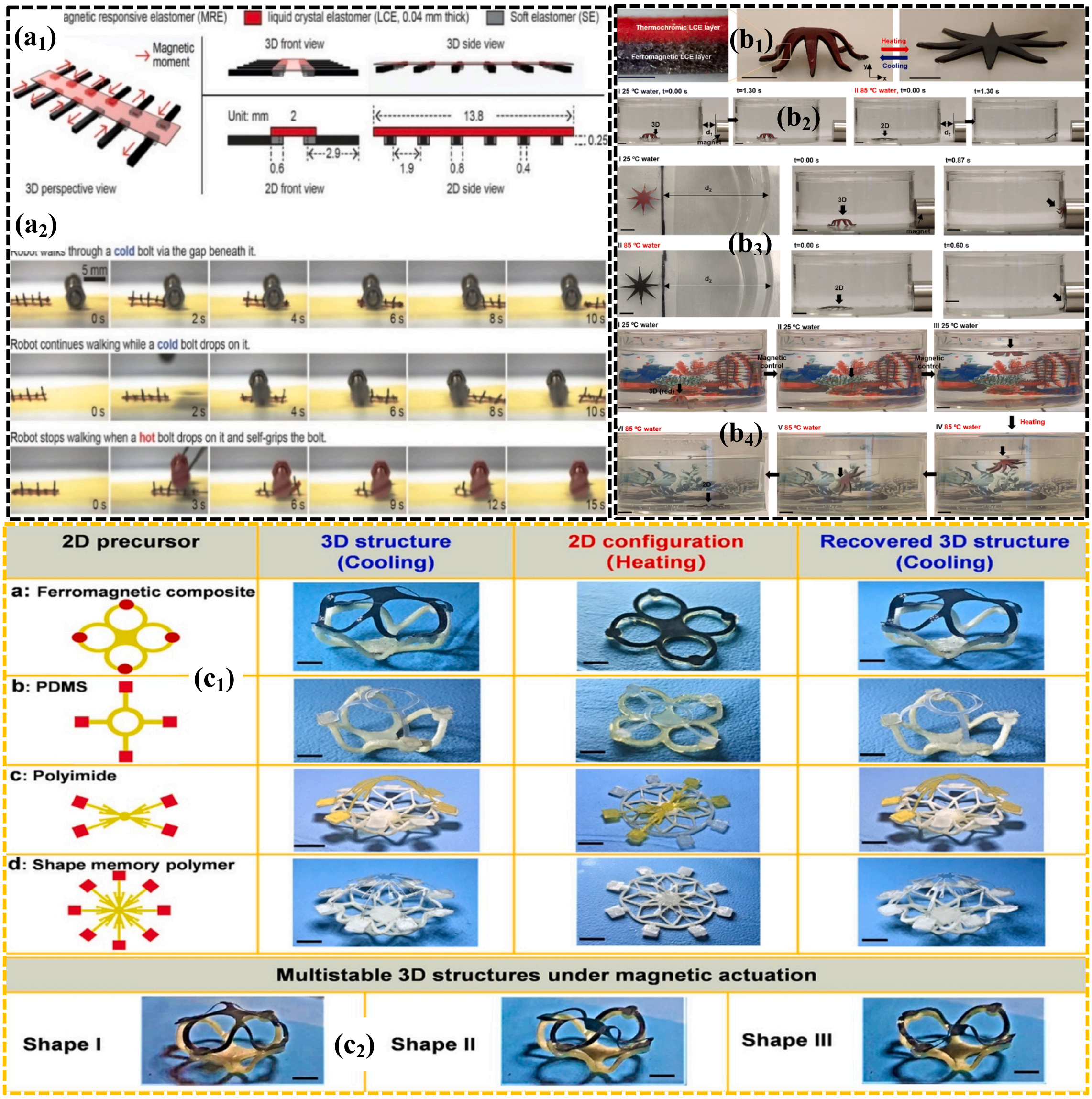

Fig. 11. (a_1) Schematics demonstrating the design untethered miniature 12-legged robot; (a_2) Robot movement and self-gripping of the hot bolt (adapted from [377], under the terms of the Creative Commons CC BY license); (b_1) Bilayer structure consists of ferromagnetic and thermochromic layers; (b_2) Magnetic actuation of octopus structure at different water temperatures; (b_3) Adaptive motion of octopus structure, when water temperature changed from 25 °C to 85 °C, under the same magnetic stimulus; (b_4) Different motion and camouflage behaviors of octopus structure through thermo-magnetic dual responsiveness (adapted with permission from ref. [379], copyright 2022, Royal Society of Chemistry); (c_1) Diverse assembled 3D mesostructures and their configurations under heat stimulus; (c_2) Multistable 3D mesostructure under magnetic stimulation (adapted with permission from ref. [381], copyright 2021, American Chemical Society).

also be manufactured by integrating programmable SMPs with non-programmable LCEs, to achieve remote and on-demand actuations. These multi-actuated composites are highly suitable for remote actuation in biomedical devices and soft robotics, where deployment and automated shape programming in a delicate or closed environment are required [380–382]. For instance, Li et al. [381] devised a facile approach to develop a multi-responsive (magnetic + heat) shape morphing 3D mesostructures, as illustrated in Fig. 11(c). The study demonstrated that these mesostructures exhibited versatile geometries and reconfigurations under heat and magnetic stimuli.

3.3. Magneto-active hydrogels

The development of magneto-active hydrogels (MAHs) is considered a panacea for developing more complex parts with excellent biodegradability and crack-healing properties [383–385]. Recently, 3D-printed hydrogels have gained significant attention due to their simple, accurate, and repeatable manufacturing. In this regard, polydopamine (PDA) hydrogel, poly(3,4 ethylenedioxythiophene):polystyrene sulfonate (PEDOT:PSS) and polyacrylamide (PAAM) are widely used for achieving toughness, and biocompatibility and validating the 3D printability of such a hydrogel into customized architectures [386–388]. Moreover, hydrogel products with excellent multiscale architectures and improved binding affinity at the interface of other polymer chains [389]. Mostly two networks of hydrogels and polymers termed static, and dynamic are extensively used to develop smart structures. Static dealing structural integrity of materials or dynamic coping mostly with self-recovery and self-healing properties [390].

Different natural and synthetic polymers or their combinations are used to develop hydrogel chains through different cross linking ways [391–393]. MAH was first proposed in 1996 and has been extensively researched ever since. Magnetic hydrogels with unique and distant magnetic manipulation are captivating, particularly for hydrogel-based flexible and soft actuators [394–396]. These hydrogels contain hydrogel

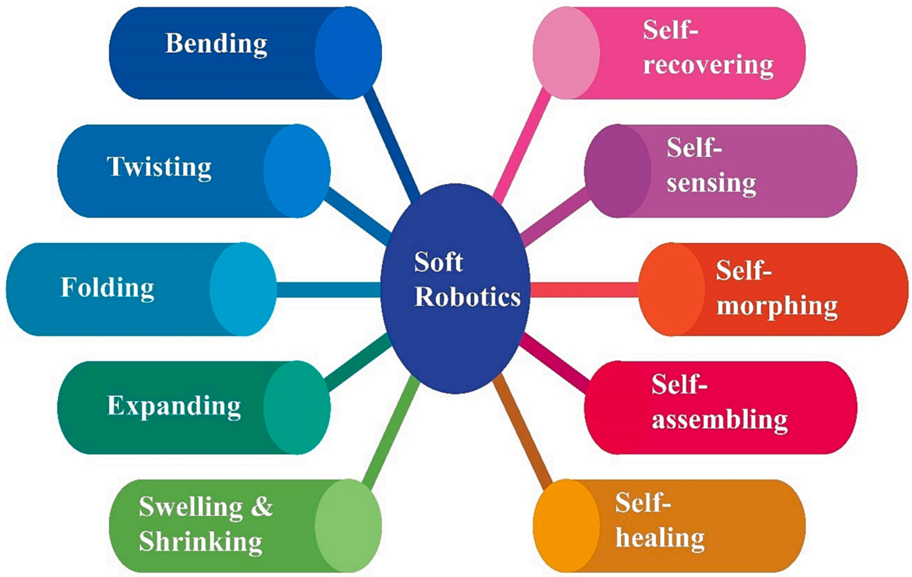

Fig. 12. Prominent feature changes developed (on the left side) and necessary functions (on the right side) of 3D-printed soft robotics.

chains embedded with nano-/micro-scaled ferromagnetic or paramagnetic fillers that permit rapid actuation in response to an external magnetic field. These hydrogels easily entrap MPs and exhibit excellent stability and processability [397–400]. Magnetic response appears in MAHs due to the addition of MPs [401]. These hydrogels have distinct advantages such as wireless actuation, facile operation, complete biosafety and biodegradability, self-adaptability, intelligence, highly controllable magnetic responsiveness, fully reversible response, and compatibility with miniaturization and integration [267,402–404]. Thus, 3D printing of MAHs has an enormous prospect in remote-controlled and untethered soft actuators, bionics, soft robotics, flexible electronics, hyperthermia cancer therapy, deployable micro-devices, and minimally invasive surgery [405–409].

4. Applications

MASMs with sophisticated functionalities are particularly attractive for various fields [410] including actuators [411], soft robotics [412] and responsive medical devices [413], sensors for drug delivery agents [414], artificial muscles [415] and implants [416]**.** This section covers the recent developments in terms of shape-morphing behavior such as self-assembly, self-healing, and changes in various smart material properties which are responsible for their advanced applications in various sectors [417]. Advances in magneto-active composites have led to the development of magnetic soft machines as building blocks for small-scale robotic devices [418]. Likewise, electromagnetic actuators are particularly appealing in numerous fields, especially in the micro-size realm [419].

4.1. Soft and intelligent robots

Soft actuators in robotics have gained tremendous attention all over the world due to their unique advantages such as being capable of performing a multi tasks across different domains, high deformability, dexterity, high controllability, safety, noncontact features, and robustness for various purposes [420–422]. Compared with traditional rigid robots, soft robots have numerous advantages such as motorless driven mechanisms, simple structures, good flexibility, silent operation, and biocompatibility [423–425].

Intelligent magnetic soft robots can change their structure in programmable and multifunctional modalities depending on material architectures and methods for controlling magnetization profiles [426]. Particularly, pneumatic soft actuators [427], and pneumatic origami actuators were explored due to their unique attributes for producing a large deformation of patterns with highly energy-efficient devices and safe tissue interaction [428]. However, there is a price to pay for the universal soft gripper, as its vulnerability limits its lifespan (50,000 grips), particularly when sharp objects are present (5000 grips) [429]. However, soft magnetic actuators offer versatile locomotion modes including walking, crawling swimming, rolling and jumping motions have shown great potential for emerging applications [187,430,431].

Soft robotics are usually constructed of inherently flexible materials which improve their ability to adapt to complicated situations and cooperate interactions with humans and soft actuators [432–434]. Fig. 12 shows the key features and their dynamic behavior of soft robotics under a stimulant environment. Traditionally, MPs are incorporated in soft robotics for introducing anisotropy in two ways First, after the fabrication of the soft robot and second while fabricating the soft robot. However, the starting material such as the magnetic composite of a soft resin and MPs remains the same for both methods.

Recently, a pneumatic origami structure using liquid silicone rubber was printed through an industrial 3D printer. The proposed industrial printer directly printed the 3D folded structure (origami-inspired structure) to maximize the design freedom for grasping various objects [435]. Urs et al. [436] studied unique two quasi-direct-drive actuators weighing 8–15 kg robots made from 3D-printed components for an overall cost of less than USD 200 each. These thermal actuated actuators were subjected to 420 k strides of gait data which nearly doubles the thermally driven torque and is useful in high-speed legged robots while matching the performance of traditional metallic actuators. These 3D-printed designs are regarded as highly customizable and reproducible soft actuators [437], for potential applications in robot legs. Recently, Wan et al. [438] studied three kinds of pneumatic soft actuators for fabricating an out-pipe crawling soft robot. Results revealed that the pipe robot realized omnidirectional turning and could adapt to diverse shapes and sizes of pipes with a movement speed of 2.85 mm/s. Moreover, the small in size, low in mass and has a higher degree of freedom the soft robotic arm achieved omnidirectional bending and a specific range of grasping work, for potential applications in underwater pipe soft robots. Li et al. [439] studied multilayer DLP-based printing for patterning MNPs including micro-structure through 2PP using gelatin methacryloyl (GelMA)-based hydrogel with neodymium-iron-boron (NdFeB) or iron particles in the ultraviolet (UV)-curable PDMS-based polymer matrix. Results showed that magnetic torque actuation

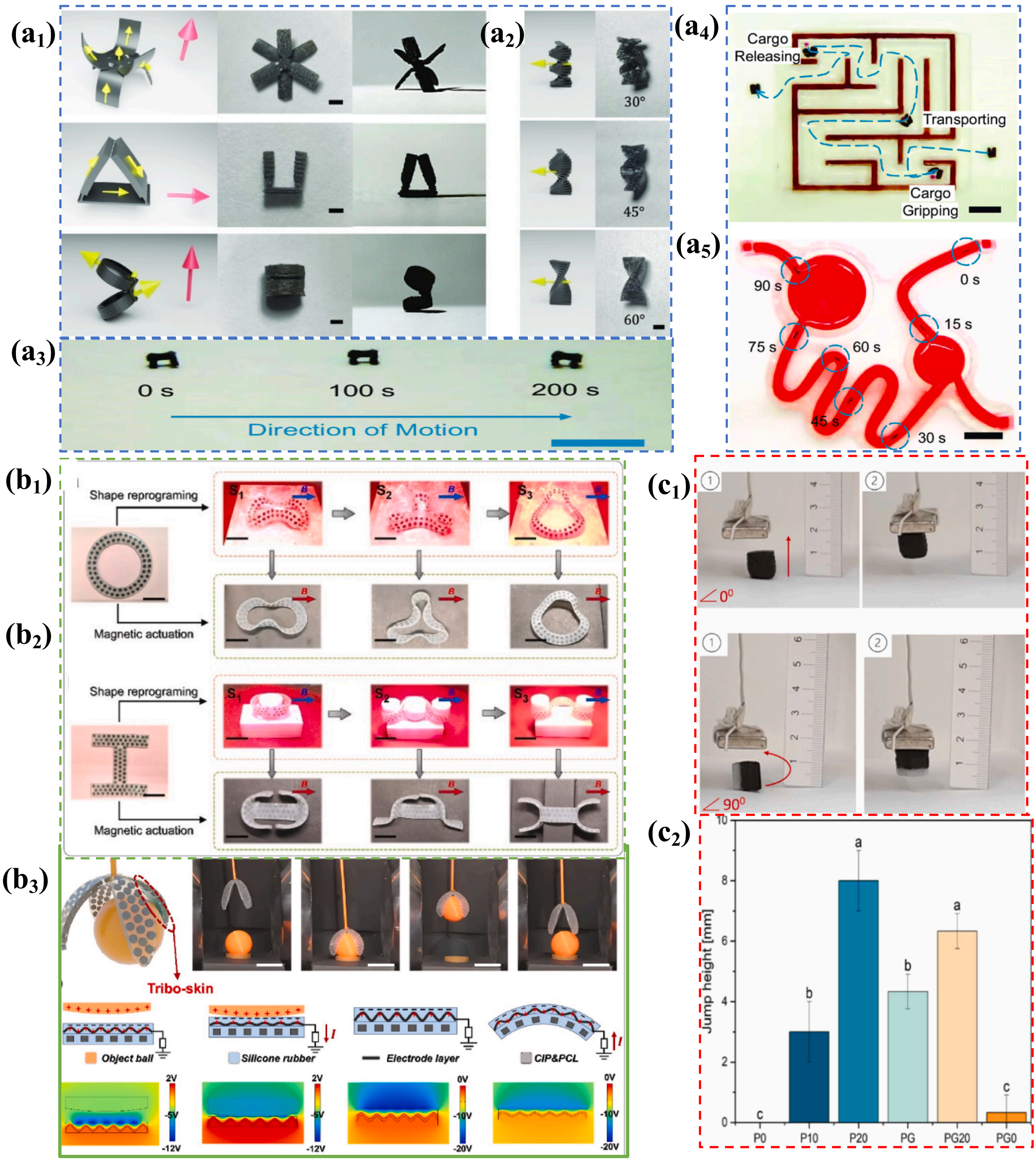

Fig. 13. (a_1) Images of the 3D-printed robots under magnetic actuation (actuation field highlighted with red arrow) with encoded magnetization profiles (magnetization direction highlighted with the yellow arrow at each segment), (a_2) Helical robots with various helix angles, (a_3) Motion of the robot with the oscillatory frequency of 2 Hz under actuation field, (a_4) Navigation of capsule-like robot in a maze map for cargo manipulation including gripping, transporting, and releasing, (a_5) Navigation of helical robot in a vascular model (adapted from ref. [439], under the terms of the Creative Commons Attribution License,); (b_1-b_2) Reprogramming and magnetically actuated shape morphing behavior of 3D-printed various characters (b_1) "O", (b_2) "H" under magnetic field 400 and 300 mT, respectively, (b_3) Snatching function of four leaves-based soft gripper under 300 mT (adapted with permission from ref. [441], copyright 2022, Elsevier Ltd.); (c_1) Various jumping behavior of magnetic hydrogel under a magnetic field, (c_2) Difference of jumping heights for various 3D-printed cubes (adapted from ref. [442], under the Creative Commons CC-BY-NC-ND license).

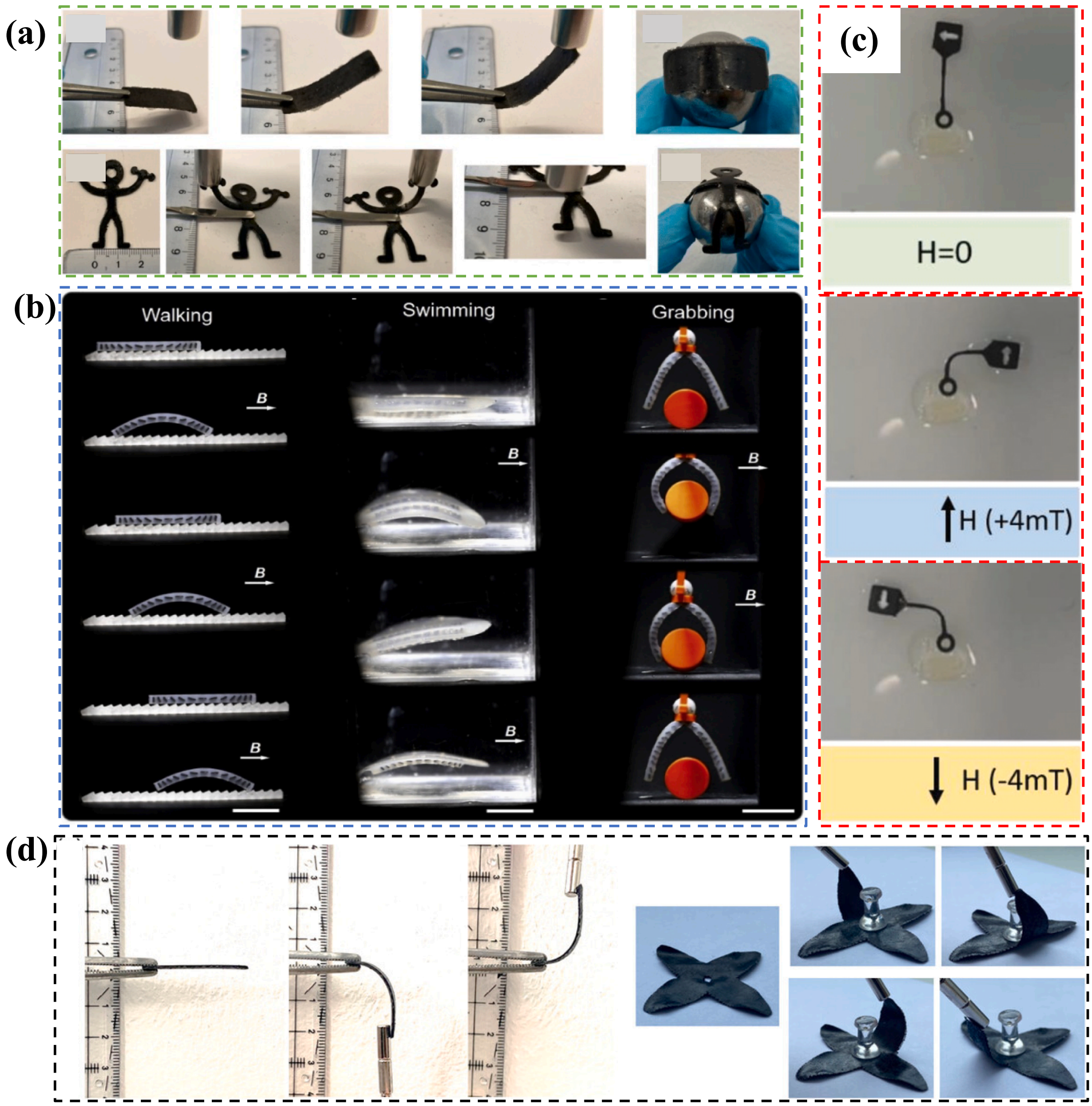

Fig. 14. (a) Activation of DLP 3D-printed-based objects with resin-2 containing 4 wt% of Fe_3O_4 nanoparticles (adapted from ref. [445], under the Creative Commons Attribution license); (b) Various shape-programmable behaviors magnetic actuated soft materials, such as Inchworm-like soft robot walking motion of the on serration plate, swimming of the manta ray-like soft robot under water, and grabbing and releasing of the soft gripper with a weight of the cylindrical object is 15.3 g (adapted with permission from ref. [448], copyright 2020, Elsevier Ltd.); (c) Shape morphing behavior of a magneto-responsive soft hammer such as bending for two opposite directions of the applied magnetic field adapted from ref. [449], under the terms of the Creative Commons CC BY license); (d) Shape memory behavior of 3D-printed structures under magnetic field (1.24 T) with AlNiCo magnets stripe and flower (adapted with permission from ref. [450], copyright 2022, Wiley-VCH GmbH).

produced various shape changes such as gripping, swimming, rolling, and walking, as depicted in Fig. 13(a_1)-Fig. 13(a_3) are induced by programming heterogeneous magnetization within discrete multilayer robot segments. Moreover, the opening angle of a capsule-like robot under magnetic actuation, as depicted in Fig. 13(a_4)-Fig. 13(a_5) was useful for drug delivery. Thus, the proposed facile approach is feasible for the creation of versatile 3D multi-material actuators for broader applications.

MASMs are reconsidered as fast, untethered, and reversible shape reconfiguration attractive for novel soft robotics [440]. For instance, Qi et al. [441] investigated a heat-assisted magnetic reprogramming approach for developing 3D-printed magneto-active soft matter using CIP as a soft-magnetic reinforcing filler with the elastic matrix silicone rubber. The magnetic reprogramming approach relied on heating PCL-based thermoplastic matrix above its melting point and applying magnetic fields during cooling for reorienting soft MP chains for achieving multiple deformation modes with unique shape-morphing features, as presented in Fig. 13(b_1)-(b_2). Moreover, the proposed approach was successfully employed for multiscale and reprogrammable soft machines such as adaptive grasping of a soft gripper with the tunable actuation response, as presented in Fig. 13(b_3). Lastly, the unique sensing performance of triboelectric skin (due to the use of CNT as a conductive filler) was also demonstrated by using electrical signals to identify the deformation and contact behaviors. Thus, the magnetic reprogramming approach provides a new concept for designing new active materials for broader applications in soft robotics.

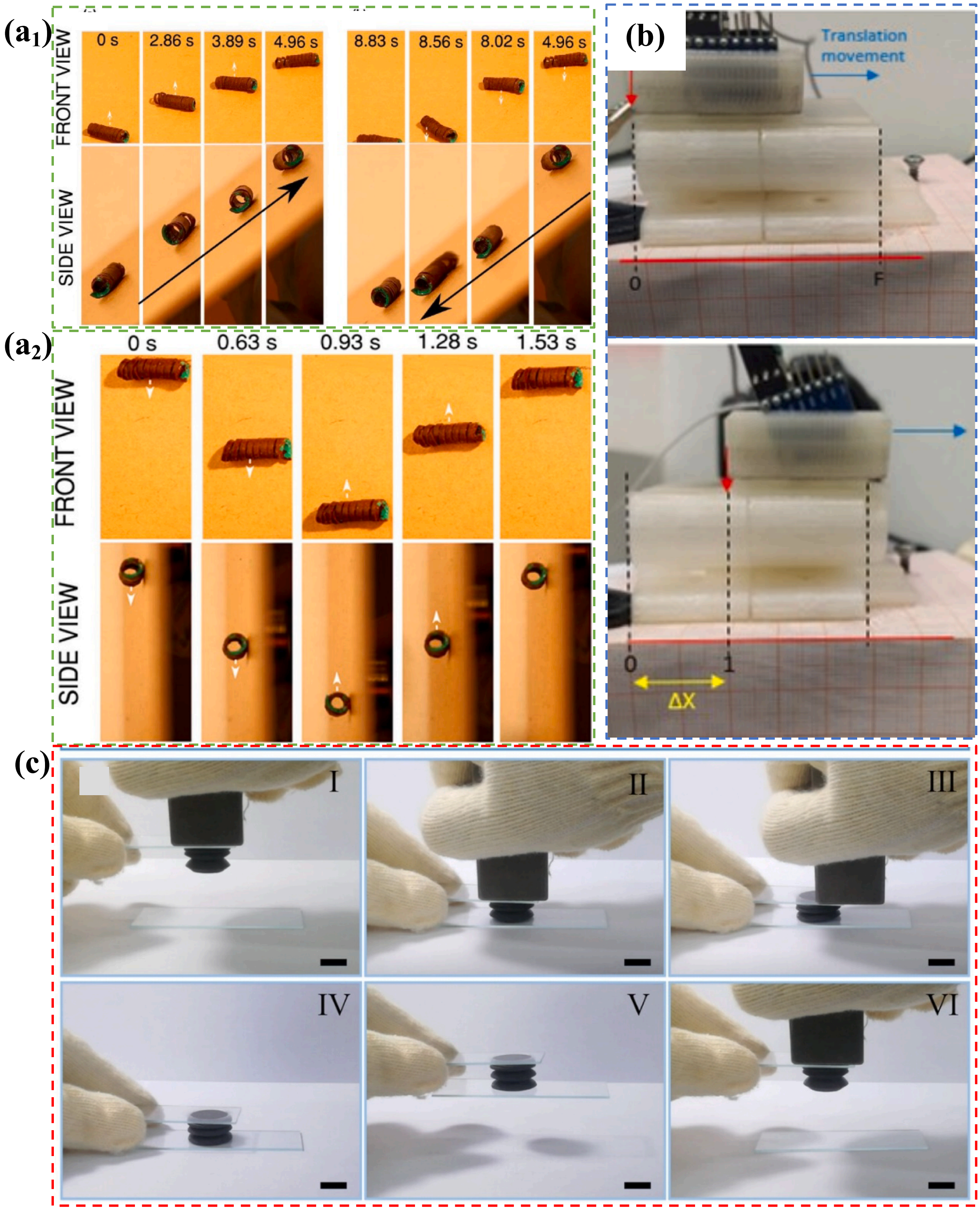

Fig. 15. (a_1-a_2) Images of untethered locomotion of helical coil on a 45° incline (a_1) upward, downward, and (a_2) on the 90° vertical wall, front view and side view (adapted with permission from ref. [452], copyright 2020, Elsevier B.V.); (b) Translation movement of the actuator with and without current supply: initial position, and final position with current supply (adapted from ref. [453], Under a Creative Commons license); (c) Images of sequential grasping and releasing the glass slide with sucker actuator (adapted from ref. [456], under the terms of the Creative Commons CC BY license).

In another novel study by Simińska-Stanny et al. [442] soft actuators were fabricated using printable magnetic hydrogel ink through multi material DIW. Results showed that magnetic hydrogels had good mechanical stability, unique magnetic responsiveness, highly porous as well as noncytotoxic towards fibroblasts. Moreover, 3D-printed magnetic actuators demonstrated excellent actuation behavior, as depicted in Fig. 13(c_1)-(c_2) by magnetically induced jumping rolling and bending. The proposed 4D printing of magnetically responsive hydrogel strategy would provide an efficient way to fully capitalize on the role of biocompatible materials for developing a wide range of soft actuators.

Soft robotics always suffer permanent damage from irregular external stimuli and repetitive motions during their long service life [443], thus the self-healing ability of smart material is highly desirable for overcoming these issues [444]. Cazin et al. [445] explored the

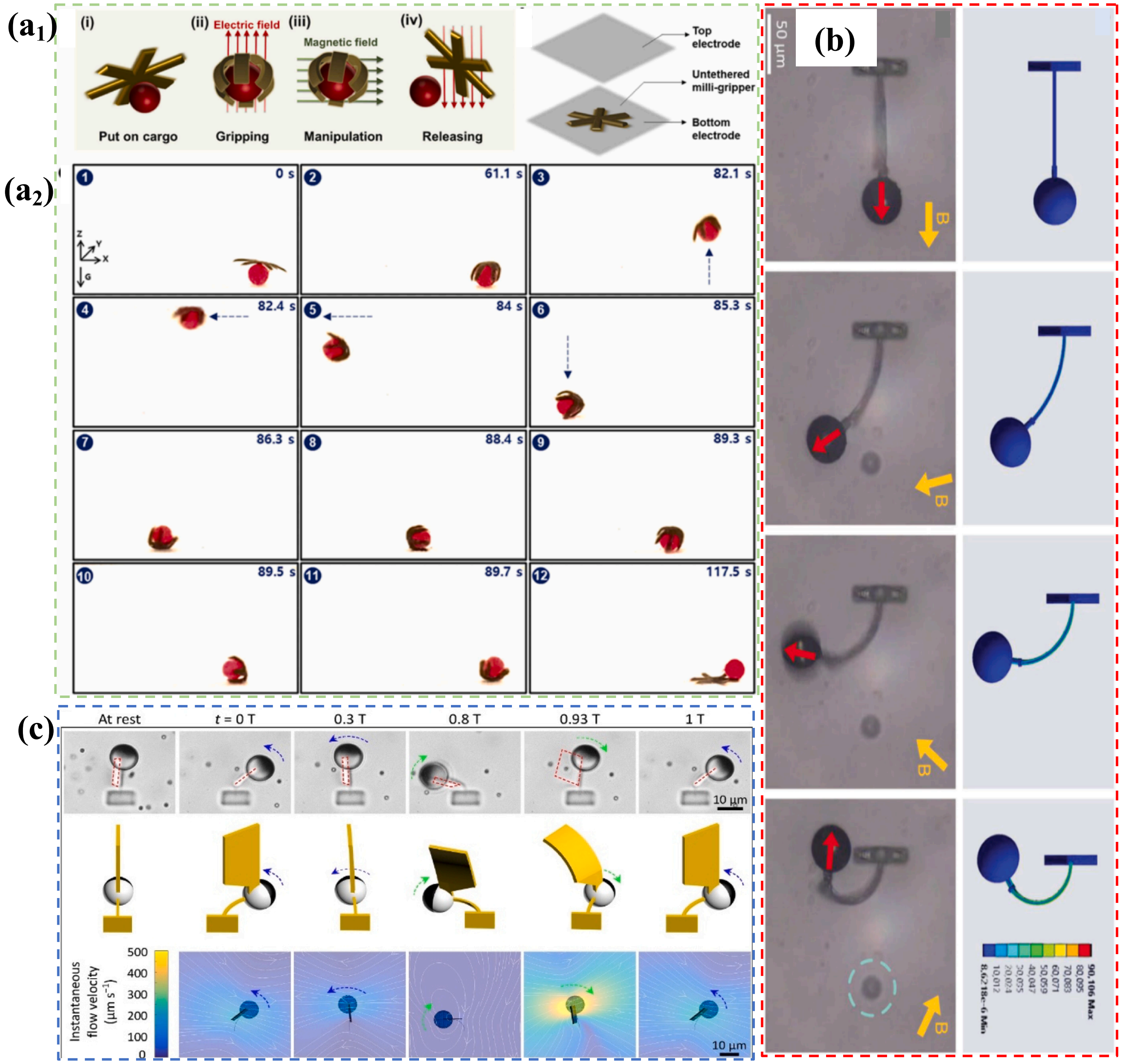

Fig. 16. (a_1-a_2) Untethered milli-gripper used for cargo delivery test: (a_1) Schematic diagram illustrating the untethered milli-gripper on cargo stimulated by a magnetic field and releasing of the cargo induced by an electric field, and schematic diagram showing the electrode system used in the cargo delivery test, (a_2) Explanation of sphere-shaped cargo during delivery text (adapted with permission from ref. [466], copyright 2023, Elsevier B.V.); (b) Magnetic actuation of the skeleton experimental and simulation results (adapted from ref. [467], under the terms of the Creative Commons CC BY license); (c) the deflection of the flag-shaped structure during the magnetic (blue arrows) and elastic (green arrows) strokes and the induced instantaneous flow (white lines in the modeling results. Photo credit: Shuaizhong Zhang and Rongjing Zhang, Max Planck Institute for Intelligent Systems (adapted from ref. [469], under a Creative Commons Attribution License 4.0 (CC BY).

magnetic response with thermo-activated healability using Fe_3O_4 nanoparticles in a dynamic photopolymer network (thiol-acrylate resins containing magneto-active fillers) through DLP-based 3D printing. Results demonstrated that the healing performance of 3D-printed structures was observed due to the recovery of magnetic and mechanical properties under temperature-triggered mending. As a proof of concept, the 3D-printed magneto-responsive structures were thermally healed, reshaped, and activated under magnetic field stimulus, as presented in Fig. 14(a).

MASMs embedded with hard MPs are regarded as robust materials for achieving fast-transforming actuation [400,446,447]. For instance, Qi et al. [448] proposed a unique technique for fast and reversible shape-programming of magnetoactive soft materials with stable shape transformation properties. The high-performance deformation of soft material was achieved using a flexible matrix and soft-magnetic 3D printing filament. These 3D-printed soft materials are used for numerous biomimetic structures such as inchworms, manta ray, and soft grippers with multiple capabilities including walking, swimming, and snatching, as illustrated in Fig. 14(b). This work enabled potential applications such as medical care, soft robotics, and bionics applications.

Lantean et al. [449] investigated complex macroscopic gear-based devices through DLP using MAPs containing Fe_3O_4. Insights of this

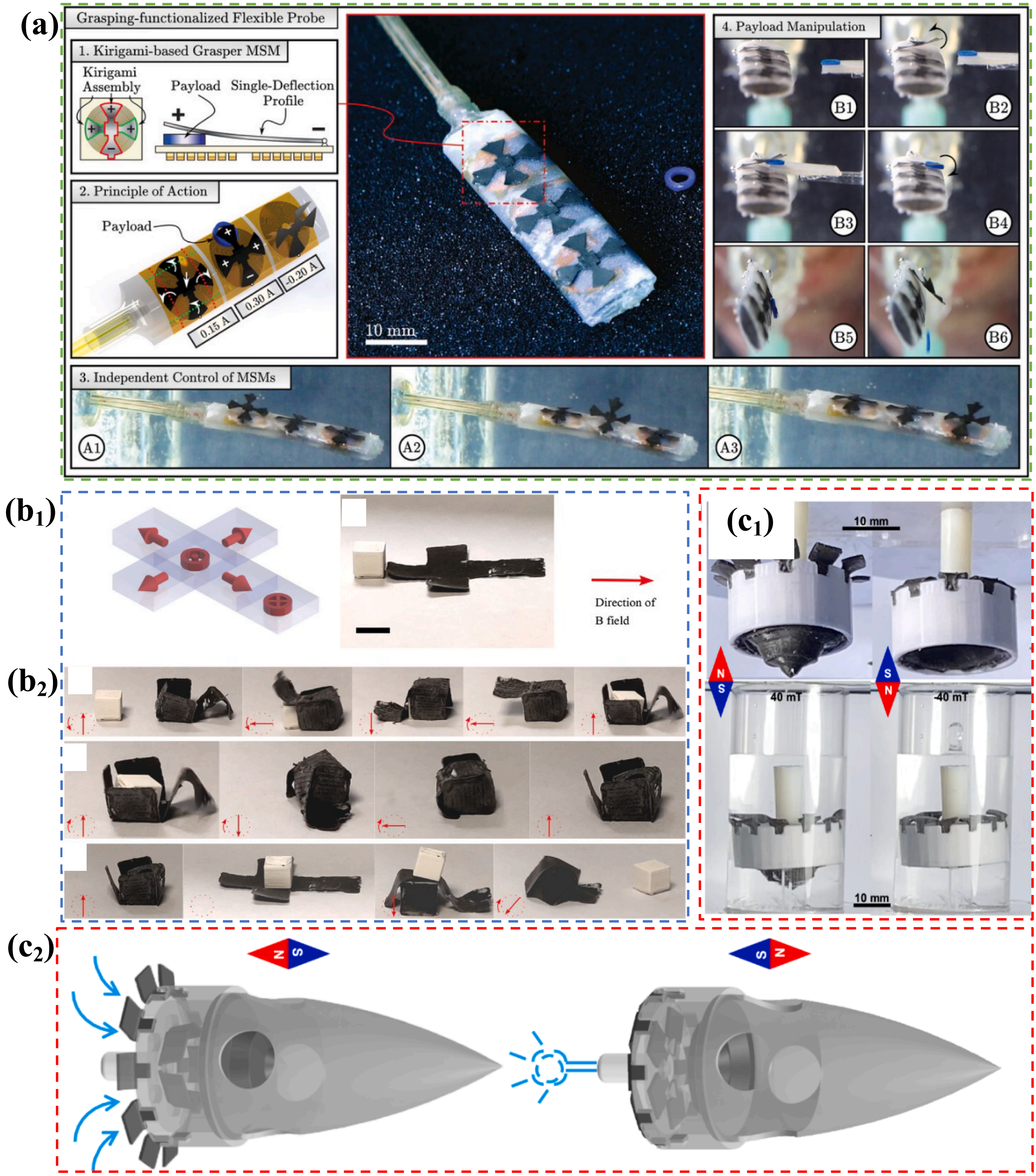

Fig. 17. (a) A flexible tip functionalized for payload (20 mg) grasping and release using near-field magnetic soft machines (adapted from ref. [470], under the terms of the Creative Commons CC BY license); (b_1) Schematic diagrams of the printed sheet with six various magnetization directions on the six faces of the cube, and the printed sheet with said magnetizations placed alongside a cubic object of 0.5 g (b_2) Load carrying ability, the cube folding rolling over the object to pick it up under a magnetic field, the cube carrying the object to the desired location where and dropping of object under unfolding at the target location before rolling back to a desired point (adapted from ref. [471], under the Creative Commons CC-BY-NC license); (c_1) Image of cavity water filling water of bionic squid swimming robot (40 mT), and image of water jet of bionic squid swimming robot (–40 mT), (c_2) Sucking and schematic diagram showing swimming robot driven bt the harmonic magnetic field (adapted from ref. [472], under a Creative Commons Attribution 4.0 International License).

study revealed magneto-responsive hammer-shape actuators, as presented in Fig. 14(c) with different stiffnesses demonstrating various motions including rotation and bending. Thus, magneto-responsive gears made from MASMs have advantages in broader applications including linear actuators, gear-trains, and micro grippers. Rossegger et al. [450] explored magnetic-driven actuators through DLP-based using magneto-responsive thiol-click photopolymers containing Fe_3O_4. The thiol crosslinker further imparts softness and flexibility to magnetic actuators. Moreover, as proof of concept, various 3D prints such as strips and flowers, as depicted in Fig. 14(d) showed magnetically driven movement for their promising role in soft robotics and other fields.

3D-printed magnetic actuated soft robotics offers an unprecedented geometric configuration with more degree of freedom due to the programmable magnetization profile [377,451]. For instance, Bayaniahangar et al. [452] fabricated 3D-printed soft magnetic helical coil actuators using PDMS embedded with iron oxide particles. The developed complex helical coil structures were supported with Pluronic f-127 hydrogel and had 30 % iron oxide particles. This allowed linear magnetic actuation with 360 % device's linear actuation and 80° bending actuator in helical coils. Insights of this study also revealed that the 3D-printed helical coils under magnetic field stimulus demonstrated untethered soft robot locomotion as presented in Fig. 15(a_1)- Fig. 15(a_2) on 45- and 90-degree inclines. Pavone et al. [453] printed support-free actuators to exploit the Lorentz Force: permanent magnets and Gallium for effective movement of the actuator. The insights of this study revealed that 3D-printed actuator has a wide range in numerous fields such as limb prosthesis wearable devices, and human motion. Moreover, at a maximum current of 6.10 various actuator movement (displacement of 20 mm and acceleration of 1.10 m/s^2) was observed as presented in Fig. 15(b).

Soft actuators are made of flexible or compliant materials and give large deformation and high stability for many applications [454,455]. Recently, Cao et al. [456] developed ultra-flexible magnetic actuators through a facile FDM-based 3D printing technique using thermoplastic rubber (TPR) pellets/CIPs. Also, the 3D-printed magnetic actuator exhibited highly functionalized manipulations and controllable deformation of the sucker and pump actuator for sticking objects and pumping liquid as presented in Fig. 15(c). Thus, multifunctional, and ultra-flexible magnetic actuator offers a promising strategy for fabricating highly complex and controlled deformable structures for soft robotics applications.

4.2. Untethered microrobots

Microrobots are robots whose dimension reaches in micron-sized realm for performing necessary tasks at a micron scale including sensing, object manipulation, and improved navigation under external stimuli or environmental sources [186,457–459]. The science of robotics is accelerating towards the conception of microrobots with new functionalities, especially under magnetic properties to control the motion of microrobots [460]. In this regard, 3D printing techniques are captivating for making perfect microrobots ensuring their satisfactory performance. Among them, 2PP is regarded as the best technology for producing microbots due to its highest resolution at the nanometric scale, and the creation of monolithically 3D complex structures using diverse materials including inorganic and organic, passive, and active [461,462]. Untethered microrobots due to their small size and mobility have enormous prospects for localized diagnosis, in minimally invasive surgery, targeted delivery of agents, tracking, imaging, and sensing, micromanipulation, cell delivery, and biopsies [463]. Among them, magnetic actuation exerts magnetic force and torque on magnetic materials in microrobots to actuate and control them, which has the advantages of fuel-free, simple direction, speed control, and harmless penetration through living tissues [464]. Microrobots are now considered the pioneer in the development of advanced healthcare systems in personalized medicine [465]. For instance, Jang and Park [466] developed an untethered milli-gripper fabricated from 3D-printed biodegradable chitosan hydrogel ink coated with citric acid superparamagnetic iron oxide nanoparticles (SPIONs). Results showed that a 3D-printed gripper was promising for gripping and releasing cargo under an applied electromagnetic field, as presented in Fig. 16(a_1)-(a_2). Moreover, the untethered milli-gripper demonstrated a precise position control due to the high magnetization of the citric acid-coated SPIONs. Thus, the proposed work proved that the biomimetic untethered milli-gripper also be employed as a minimally invasive small soft robot in vivo for numerous biomedical applications including targeted drug delivery. Pétrot et al. [467] fabricated remotely actuatable NdFeB-based MNPs. Reported results demonstrated that magnetically deformable 3D culture substrate actuated under a magnetic field and bends back and forth along its longest axis, as presented in Fig. 16(b). Also, these structures had soft, curved, and dynamic properties of tissues in vivo for potential applications in micro-actuator field.

Soft robotics driven from AM of naturally available materials have proved to be more effective in achieving complex structures in a more deterministic manner [468]. For instance, Zhang et al. [469] exploited wirelessly actuated programmable microfluidic cilia using naturally available materials FePt Janus microparticles/silk fibroin (SF) hydrogels. Insights of this study showed that high tunable actuation performance of proposed material for various arrangements (antiplectic, symplectic, and diaplectic metachrony) and 2D arrangements (circular and triangular) was achieved, as presented in Fig. 16(c), under less than 10 mT external magnetic field. Such robust integration of the multimaterial including FePt and SF rendered cilia system allows researchers to use them for future applications in biomedical and health care devices.

Miniature robots can be deployable on the water surface for achieving high controllability for various applications. Richter et al. [470] proposed novel microscale magnetic soft actuators. Insights of this study showed that ultrathin (80 μm) and lightweight (100 gm^{-2}) magneto-responsive actuators could lift, tilt, pull, or grasp near each other under electromagnetic near-field, as presented in Fig. 17(a) at low energy consumption (0.5 W). It was envisioned that such soft micro magneto-active robot would serve as a pioneer for next-generation soft robots in various prevailing applications in both biomedical and engineering sectors.

Ansari et al. [471] printed anisotropic soft structures using magnetic ink containing a UV-curable resin and MNPs using an extrusion bioprinter. A custom electromagnetic coil system was used during extrusion for orienting the magnetic moment of the particles in the ink. Results exhibited that with 1:1 particle-to-resin ratio in the magnetic ink under a 20 mT field for orientation for printed structure demonstrated a preferential magnetization index up to 0.99. It was shown experimentally that soft structures have tremendous promise in shape morphing capabilities for an object using a folding cube robot through loading, carrying, and dropping, as presented in Fig. 17(b_1)-Fig. 17(b_2). Lin et al. [472] studied a novel magnetic-driven folded diaphragm inspired by the locomotion of earthworms having various radial magnetization properties for controlling the contraction and stretching between body segments. Experimental results showed that the developed folded diaphragm exhibited distinctive features for producing different shapes including untethered soft robotic systems as soft drivers (actuators) for their practical applications such as soft biomimetic robots and diaphragm pumps under a magnetic field, as illustrated in Fig. 17(c_1)-Fig. 17(c_2). This approach unravels many opportunities to fabricate multifunctional robots including the swimming robot inspired by squid and bio-earthworm crawling robot.

4.3. Biomimetic devices

Biomimetic is a type of human-made actuation material or device that can initiate motions under force [473]. Different bioinspired designs of scale shapes and arrangements result in various types of

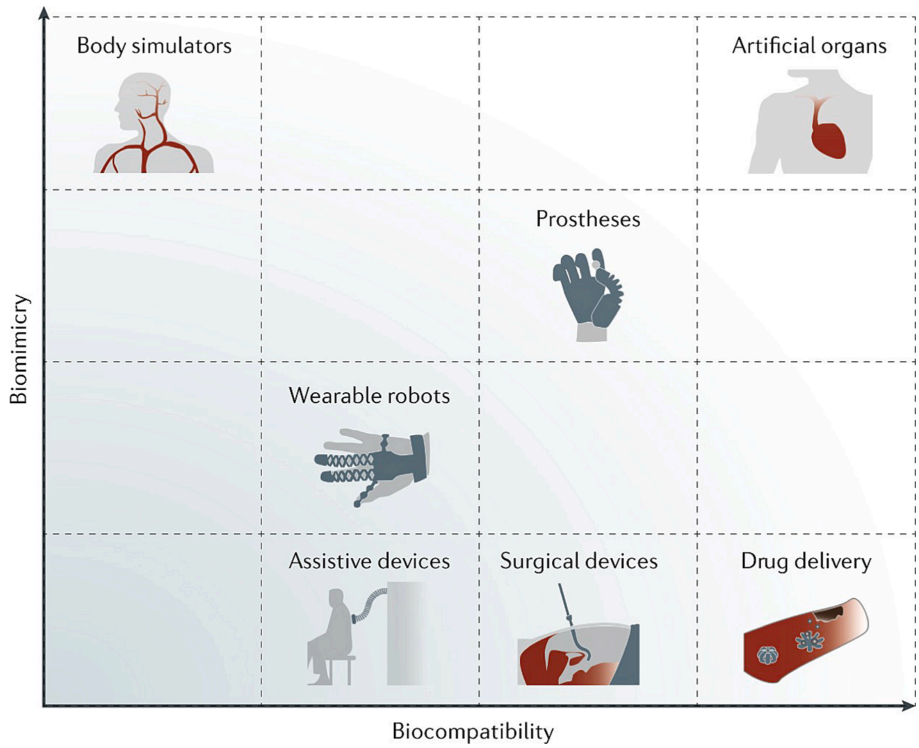

Fig. 18. Biomedical soft robots from a materials perspective comparing levels of biomimicry and biocompatibility (adapted with permission from ref. [487], copyright 2018, Springer Nature).

anisotropic friction, providing a means of switching the robot's locomotion for desired conditions [474]. Moreover, due to the huge demand for recreating human skin with the functions of the epidermis and dermis for interactions with the physical world [475], soft actuators have attracted considerable interest in the biomimetic field for many biomedical applications [476]. Magnetic robots actuated wirelessly and rapidly under an external magnetic field for non-invasively access and navigation in difficult-to-reach areas inside the human body. This is because of deformation 3D-printed smart structures which have unique implemented actions such as gripping and lifting as well as self-healing ability [477]. Using this facile strategy, other smart biomaterials could be designed which is in great demand and used for a variety of applications, such as bionic grippers [478], open-channel microfluidic chip for controllable liquid transport [479], tissue engineering [480], and drug delivery [481]. These soft robots can be precisely actuated at target sites for intelligent cargo release under a magnetic field [482] and applications related to neurological disorders such as motor and sensory deficits [483]. Thanks to their intelligent responsiveness, researchers have rationally designed magnetic actuated soft robots that can encapsulate therapeutic agents for biomedical applications [484]. Now, 3D-printed biomimetic-based devices especially those made from biodegradable materials have captivating adaptivity, complex designability and stimuli responsiveness [485] and have brought significant advancements for various biomedical applications [486], as highlighted in Fig. 18.

Biomimetic devices are usually flexible, reconfigurable, compliant, and adaptable to switch between various states (flexible to stiff) for demanding applications such as targeted drug delivery [488]. For instance, Choi et al. [489] proposed the idea of a soft carrier using through fabricating the lid, border, and hemisphere using a thermo-responsive poly(N-isopropylacrylamide) (PNIPAM)/polyethylene glycol (PEG) hydrogel and SPIONs using 3D printing. Results showed that the hemisphere allowed the successful storage and transport of cargo (soft carrier) under dual stimuli such as near-infrared (NIR) light and magnetic field with different shapes and numbers of cargo, as presented in Fig. 19(a_1)-(a_2).

Cao et al. [490] studied biomimetic magnetic actuators through an FDM-based 3D printing technique using TPR particles and CIP. Insights of the study showed that various shape transformations of magnetic actuators such as the predation behavior of octopus tentacles, the flower blooming behavior of the plant and the flying behavior of the butterfly, as presented in Fig. 19(b_1)-(b_2). It was anticipated that the 3D-printed MASMs could open new avenues for the fabrication of a diverse range of soft robotics with multiple functions.

The integration of functionalities offered by smart materials with free structures under potential stimulus renders an enriched design platform for producing artificial human organs such as a bioengineered robotic heart with beating–transporting functions [491], and many more for bionic fields. One such study explored by Gao et al. [492] through a novel composite printing powder for the preparation of asymmetric magnetic actuators using TPR and NdFeB. The experimental results demonstrated that the folding deformation amount of multi-dimensional asymmetric magnetic actuators was five times that of bending deformation. Furthermore, these actuators produced rich deformation shapes such as butterfly wing bionics and trapper, as depicted in Fig. 20 making them ideal for soft robotics and bionics fields.

Wang et al. [493] printed a millimeter-scale magnetic soft robot (referring to Fig. 21(a-b)) using NdFeB/PDMS, multiwalled carbon nanotubes (MWCNTs)/PDMS and reduced graphene oxide (rGO)/PDMS integrated with temperature, tactile and electrochemical sensing functions. Furthermore, the shape morphing behavior (Fig. 21c) of the robot showed remarkable sensing performance such as linearity of 3.383 kΩ/°C, and electrochemical stimuli with a low detection limit of 0.036 mM for NaOH solutions. Thus, the proposed study anticipated the performance of such a robust soft robot for next-generation targeted drug delivery, as presented in Fig. 21(d-e).

Intelligent tactile sensing is critical for soft robotics so that they can interact safely with unstructured environments and produce desired motions [494] under many shapes such as bionic flowers, and bionic worm robots [495]. Wang et al. [496] used a highly viscous magnetic

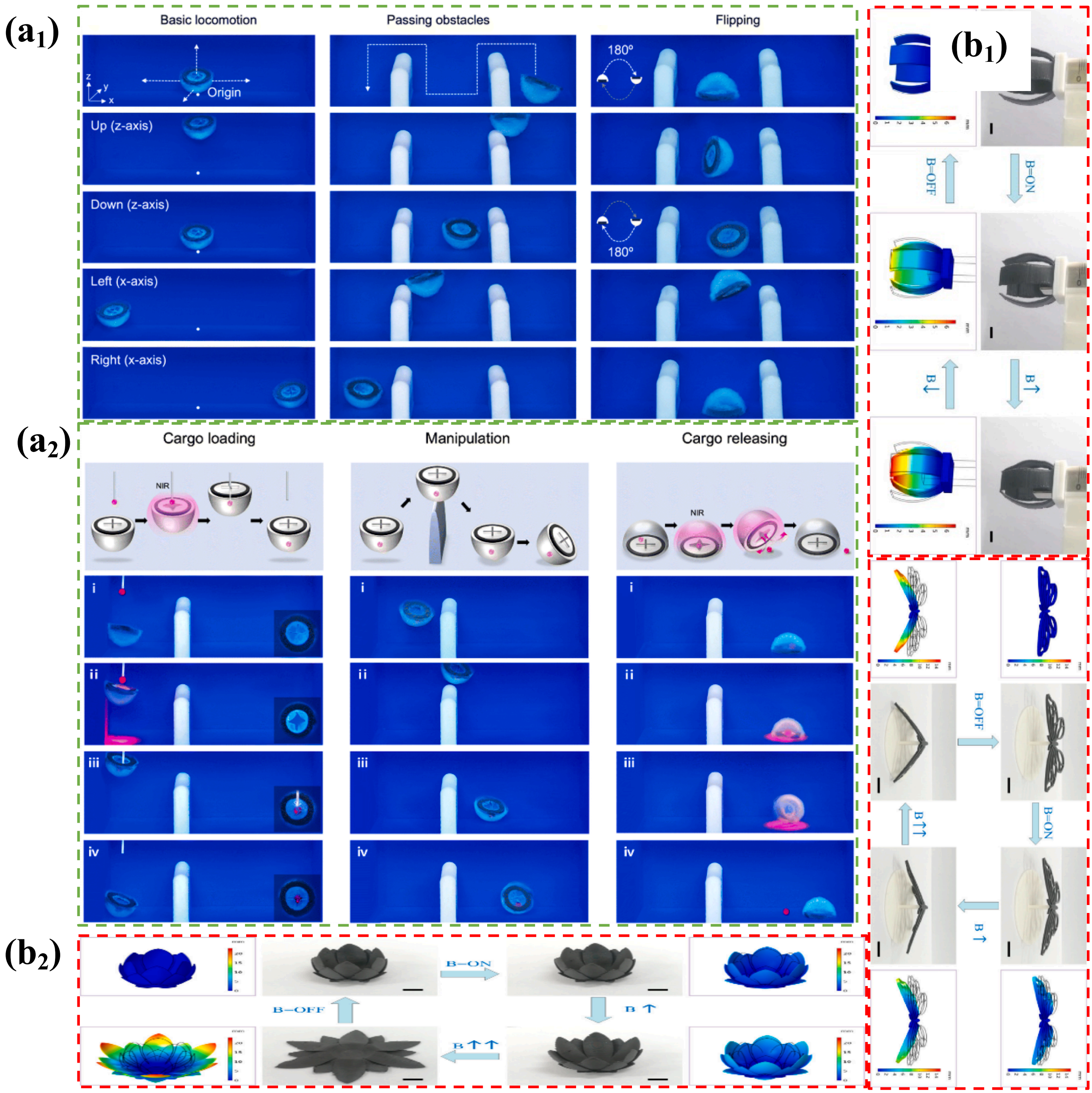

Fig. 19. (a_1) Various images of basic locomotion, passing obstacles, flipping the smart soft carrier flips by 180° using a magnet without relying on an external wall such that the downward-facing lid faces upward, (a_2) Cargo delivery test of the smart soft carrier, cargo loading, schematic of manipulation, and cargo releasing (adapted with permission from ref. [489], copyright 2023, Royal Society of Chemistry); (b_1-b_2) Magnetic field-induced deformation and finite element simulation of various biomimetic magnetic actuator: (b_1) tentacle and butterfly, (b_2) flower (adapted with permission from ref. [490], copyright 2021, American Chemical Society).

composite ink for designing various bionic soft robots. Various actuator prototypes with various magnetization orientations and profiles have been fabricated such as bionic soft robots and magnetically powered electrical switches to successfully perform different operations including dragonflies and inchworms as presented in Fig. 22(a_1)-(a_3). Thus, the proposed study confirmed that the magnetic responsive materials with programmable patterning fulfil the future of soft robotics in functional and practical applications.

Wang et al. [497] explored an insect-scale magnetoelastic robot using PDMS embedded with NdFeB-based MPs having improved controllability designed. The robot produced a controllable jumping motion by tuning magnetic and elastic strain energy. Results showed that on-demand actuation was applied for precisely controlling the pose and motion of the robot during the flight phase for effectively performing numerous tasks with integrated functional modules, as depicted in Fig. 22(b_1)-(b_4).

Yao et al. [498] studied the diversification of actuation modes of magnetic-active actuators using blending matrix of PCL and thermoplastic polyurethane (TPU) and soft CIP-based MNPs as fillers through 3D printing. The results showed that 3D-printed magneto-active

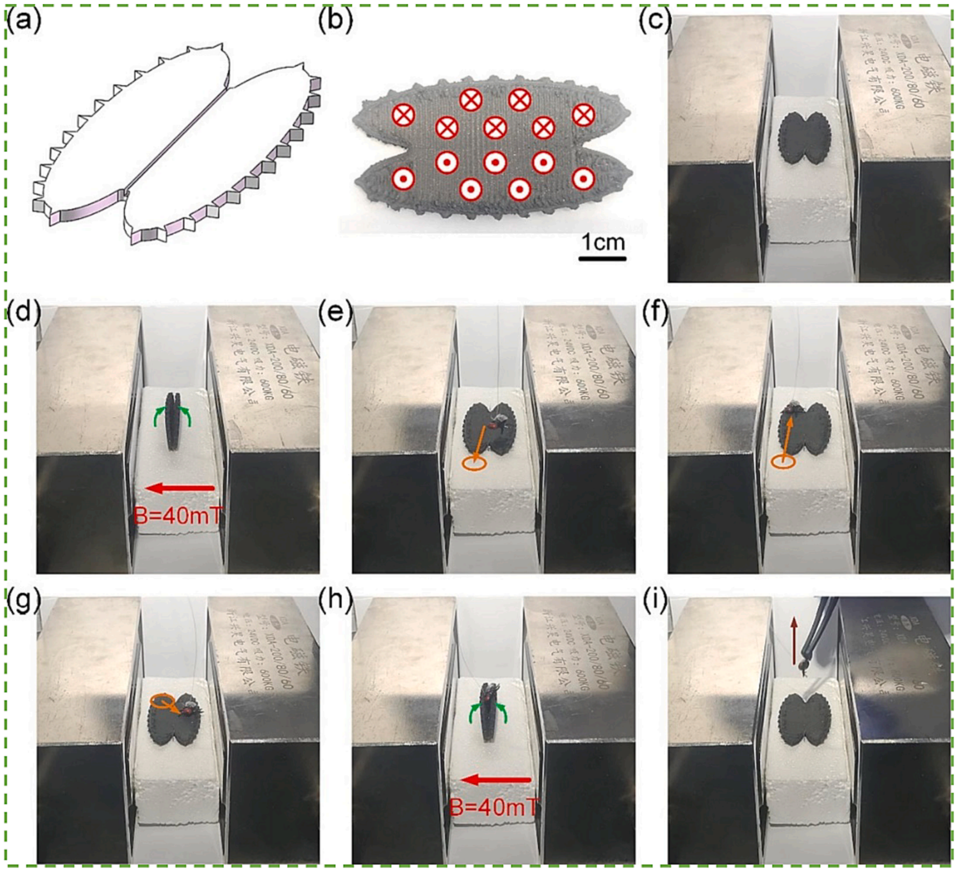

Fig. 20. Demonstration of a trapper. (a) 3D-printed model structure. (b) Direction of magnetized for the trapper. (c-d) The trapper is in a deformed state. (e-i) The trapper grasping process (adapted with permission from ref. [492], copyright 2023, Elsevier Ltd.).

structures have excellent shape fixation, shape recovery rates, exceptional flexibility, and magnetorheological effects, as presented in Fig. 22 (c). The shape-morphing behavior was an excellent match with the simulation results and has an ideal role in numerous fields such as intelligent flexible robotics and biomedicine.

Magneto-active metamaterials or field-responsive novel origami structures whose shapes or properties modulated under a magnetic field hold great promise for many applications [499]. For instance, Moonesi et al. [500] reported novel 3D-printed origami-inspired scaffolds using Fe_3O_4 and cellulose acetate (CA). Results demonstrated the cells' favourable surface morphology, superparamagnetic behavior, wettability, and appropriate compressive stiffness for cell proliferation, prominently decreased degradation, and acceptably low iron ion release of the printed scaffolds. Moreover, an optimized foldability with varying scaffold architecture was observed under magnetic field stimulus due to the presence of Fe_3O_4 magnetic particles which further allowed the scaffold folding, as presented in Fig. 23(a). Guan et al. [501] developed a magnetically assisted DIW using alumina micro-platelets and fumed silica for printing various structures. The printed structures had the ability to be turned into ceramics with anisotropic properties, including their magnetic response, high electrical conductivity, and self-shaping ability, as depicted in Fig. 23(b_1)-(b_2). This work showed that multilaterals with their magnetic response can be employed for multifunctional devices with tailored and improved properties.

Luo et al. [502] prepared various magnetic-controlled liquid block structures with the ability to program and reconfigure precisely under an external magnetic field. Liquid biomaterial inks were prepared by gelatin methacryloyl (GelMA)/alginate (ALG) and carboxyl modified Fe_3O_4 MPs. Results showed that various liquid blocks including H-type and the spinal column-like scaffolds demonstrated biomimetic morphologies and various functions, as presented in Fig. 23(c_1)-(c_3). Thus, considering the outstanding biomimetic functions from natural materials the mentioned liquid blocks above together with the essence of the magnetically controllable show great application potential for tissue engineering.

Zhao et al. [503] prepared personalized 3D printing of a bio-designed tracheal scaffold using shape memory PLA/Fe_3O_4 composites filament under the magnetic stimulus. Results showed that a 3D-printed tracheal scaffold with glass sponge microstructure exhibited higher strength, and shape fixation for its unique ability to adapt the complex environmental conditions in the soft tissue of patients. Moreover, 3D-printed scaffolds changed to a temporarily deformed configuration and deployed back into a conformed shape under magnetic field stimulus, as presented in Fig. 23(d_1)-(d_2).

Shao et al. [504] reported a facile technique for magnetically-driven triple-finger micro-gripper through 3D printing with robust micromanipulation in both water as well air. Also, the 3D-printed gripper was attached to a robotic arm to exhibit its ability to manipulate micro-objects both air and water, as depicted in Fig. 24. This work proved that when the magnetic field is removed the low remanent magnetization permits the actuator to recover to its original status by elastic energy while improving magnetic response under the magnetic field. Consequently, the developed 3D printing micro-gripper has broad biomedical application prospects such as the operation of live cells and soft tissues.

4.4. Advanced sensors and flexible electronics

In addition to performing many intelligent functions, stimuli-responsive smart sensors can perform many tasks such as self-validating, self-testing, self-identifying and self-adapting as part of their task or responsibility [505]. As opposed to conventional sensors, smart sensors can manage their functions by being stimulated by

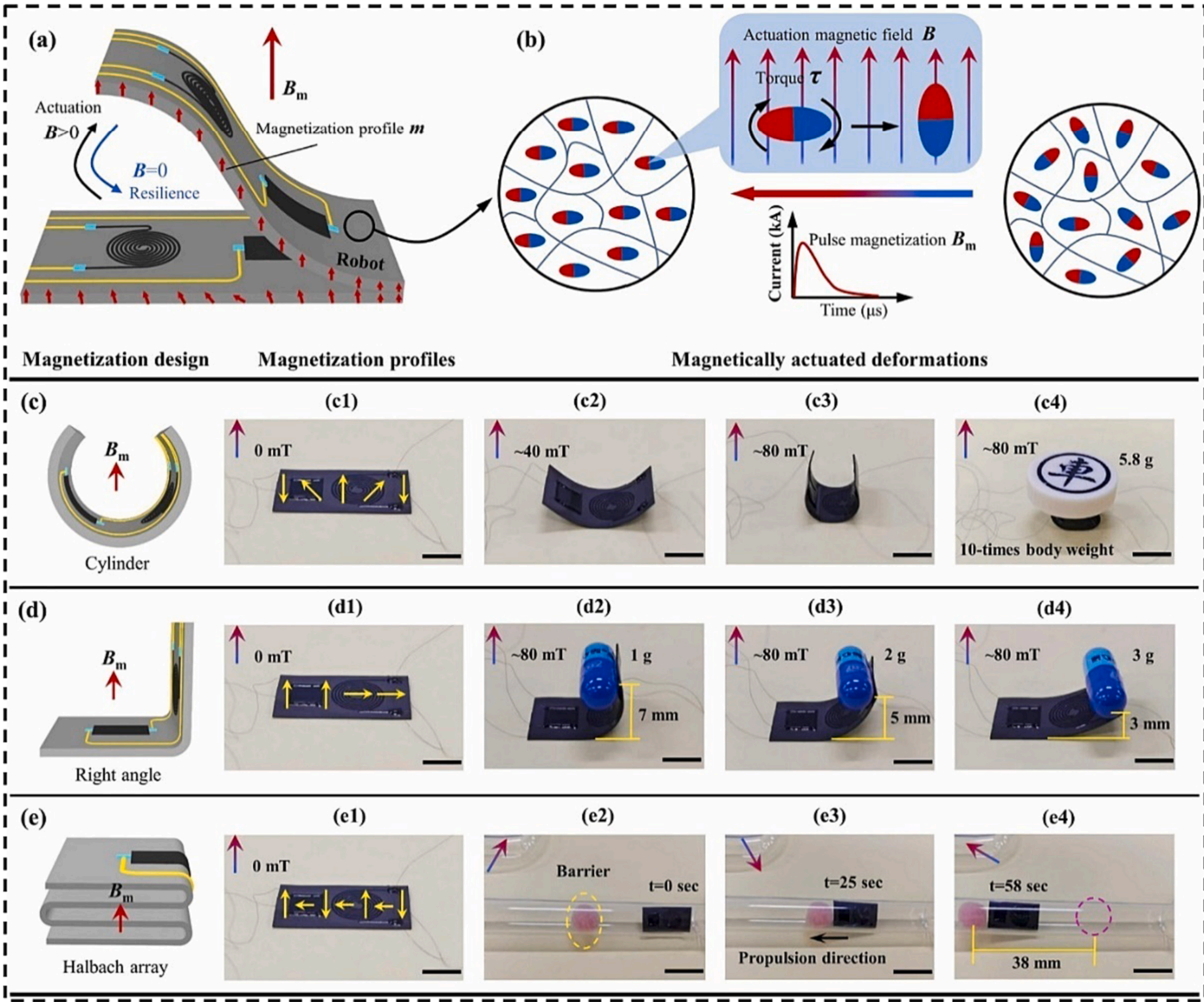

Fig. 21. Magnetic soft robot multi-dimensional deformations and actuation mechanism. (a) The folded robot was magnetized under a magnetic field having a unidirectional pulse Bm and implanted into its body with the magnetic profile. (b) Illustration of magnetization and actuation mechanism. The robot performed multi-dimensional deformations driven by an external actuation field using origami-based reconfigurable magnetization: (c) cylinder, (d) right angle, (e) Halbach array (adapted with permission from ref. [493], copyright 2022, Elsevier B.V.).

external factors (external environment) in which they are located and thus manage a variety of conditions. These features of smart sensors are particularly attractive for achieving self-adaptation, advanced learning, and signal processing architecture, within a single integrated circuit. Smart sensors are crucial for designing stretchable electronics such as wearable monitoring systems, skin electronics, invasive electrophysiological recordings, and prosthetics [506].

Flexible electronics-based devices are an emerging area and have extreme importance in both engineering and biomedical sectors [507,508]. Not limited to this, smart grippers, flexible sensors, intelligent devices stretchable ionotropic devices and many more which have not discovered yet are often required similar processing mechanisms for their operation [509,510]. There are various difficulties in these devices' fabrication through traditional 3D printing techniques such as in unbalancing printability, shape fidelity, static nature, ionic conductivity, stretchability, and other functionalities [511–513]. Such devices from 3D printing of smart materials (4D printing) can greatly benefit from the remarkable patterning capability, complex design, and shape-changing behaviors. More importantly, many smart materials in 3D printing such as LCE demonstrate excellent recoverable shape-morphing organisms which are best suited for applications such as grippers, valves, sensors, soft robotics, etc. [514]. Recently, Han et al. [515] investigated novel magnetic microfibers, using NdFeB and PLA through filament extrusion-based printing. The printed ferromagnetic microfibers were magnetized to achieve various deformations of microfibers under magnetic fields. Moreover, the thickness, mixing ratio, and length of the magnetized microfibers provided unique and customized deformation of the microfiber for numerous applications in smart sensors and actuators.

Zhang et al. [516] developed a fully flexible soft robot through a light-cured 3D printing technique using a tentacle-integrated liquid metal spiral wire with Nd2Fe14B magnetic powders/Ecoflex (liquid silicone) composites. The various fabrication parameters were optimized for achieving good energy transmission efficiency between the two tentacles of soft robots. Moreover, printed soft robots demonstrated unique motion under an external magnetic field as depicted in Fig. 25 (a). Also using electromagnetic induction these soft robots can transmit electric signals to the oscilloscope.

Another novel study by Dezaki et al. [517] explored 4D-printed MRE composite actuators using silicone resins loaded with strontium ferrite-based MNPs and a thin conductive carbon black PLA. The developed composite actuator with programmable magnetic patterns showed excellent shape memory behavior such as electroactive under Joule

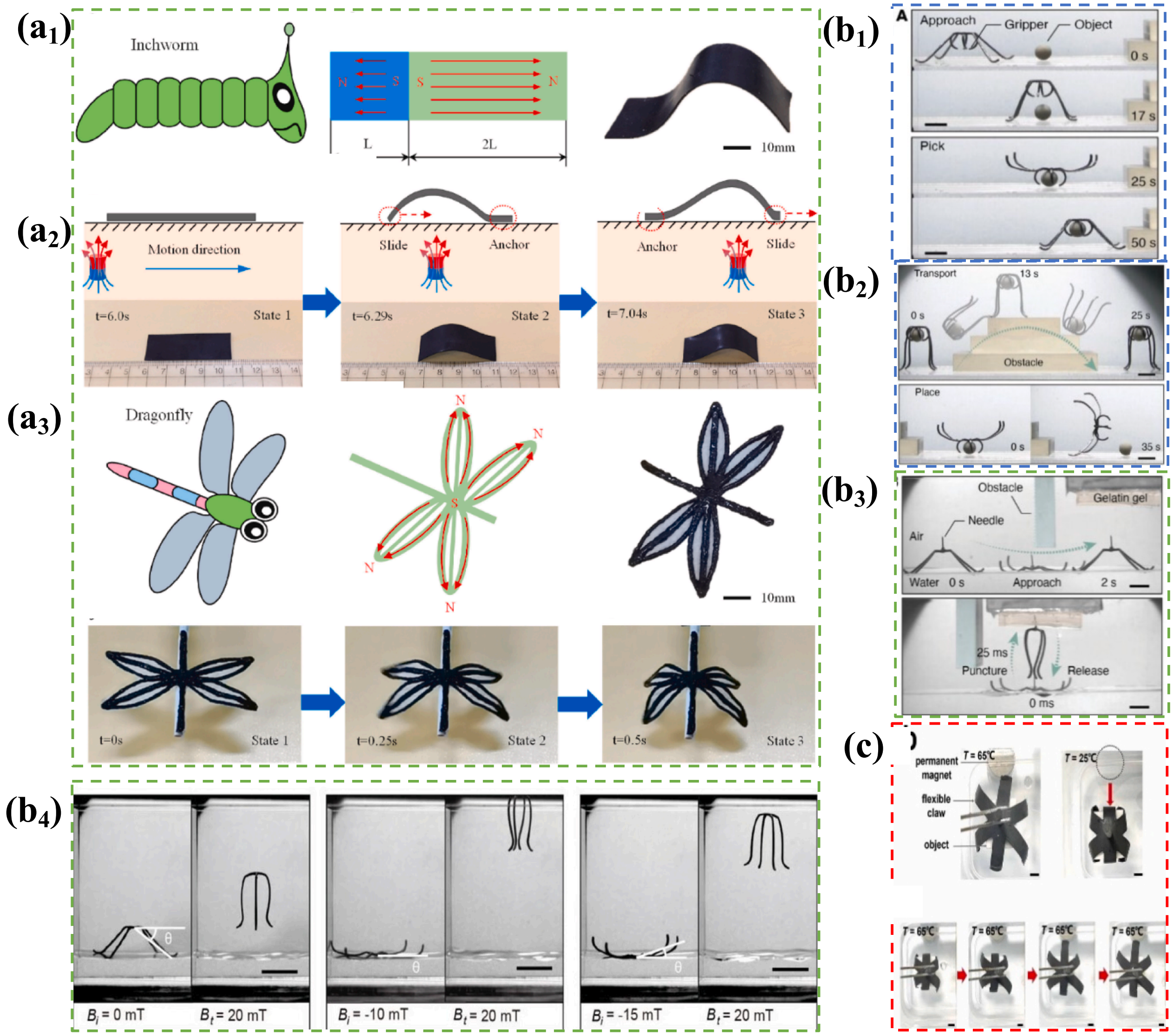

Fig. 22. (a_1) Inchworm bionic soft robot schematic diagram, a plate-shape magnetic actuator magnetization domains of the bionic inchworm robot, and the bionic inchworm robot (prototype) drive by the magnetic actuation, (a_2) The bionic inchworm robot three-step motion stages with corresponding experimental results, (a_3) Schematic and actual results of the bionic dragonfly robot under 0–140 mT magnetic fields (adapted with permission from ref. [496], copyright 2021, Elsevier Ltd.); (b_1-b_3) A robot with a soft gripper picks, transports, and places a tiny object in water, (b_2) A robot with a needle overhead performs adaptive locomotion and targeted puncturing, (b_4) In-flight maneuver of the jumping robot under magnetic stimulus (adapted from ref. [497], under the terms of the Creative Commons CC BY license); (c) The snatching and grabbing function of a flexible were activated by a permanent magnet and the release behavior of the flexible claws occurred in the absence of magnetic field and at 65 °C (adapted with permission from ref. [498], copyright 2023, IOP Publishing Ltd).

heating and magnetic fields. Moreover, the printed actuator (1.47 g) can lift weights to 200 g. As such, the developed printing process provided highly remotely controlled shape-memory features of 3D-printed composite actuators. Also, Sundaram et al. [518] fabricated complex actuators (>106 design dimensions) through multi-material drop-on-demand 3D printing using both soft and rigid polymers with MNPs. Results showed that developed multi-material 3D printing with optimized topology allowed complex actuators to use them in liquid interfaces as highlighted in Fig. 25(b). Table 4 summarizes the state-of-the-art 4D printing technologies which are recently been studied for various smart sensors and actuator-based applications. Likewise, Huang et al. [519] used an interesting approach for fabricating Fe_3O_4 driven fiber-Tip multimaterial microcantilever-based magnetic field sensor using an advanced femtosecond laser-induced 2PP technique. Insights of this study showed proposed sensor exhibited a minuscule size and a high magnetic sensitivity of 119 pm mT–1 in the range of 0–90 mT. Moreover, these sensors showed the false-color scanning electron microscopy (SEM) images of the polymeric magnetic microcantilever from the top view and the side view as presented in Fig. 25(c_1)-Fig. 25(c_2). Thus, this new facile approach can be employed for different stimulus-responsive microsensors and micro-actuators on the fiber tip. Saiz et al. [520] showed that magneto-responsive PCL/Fe_3O_4 inks containing up to 10 wt % Fe_3O_4 can be employed for high level microstructures with fiber diameters of 9.2 ± 0.6 μm using novel melt electrowriting-based 3D printing technique. Reported results demonstrated that printed samples exhibited tunable magnetic responses under various MNP concentrations and multi-material designs, as presented in Fig. 25(d). This methodology can bridge the wide-open gap for designing various complex structures at the microscale level using different active fillers combined for many mysterious applications.

5. Contemporary challenges and prospects

When the shape of a 3D-printed structure is designed to morph over

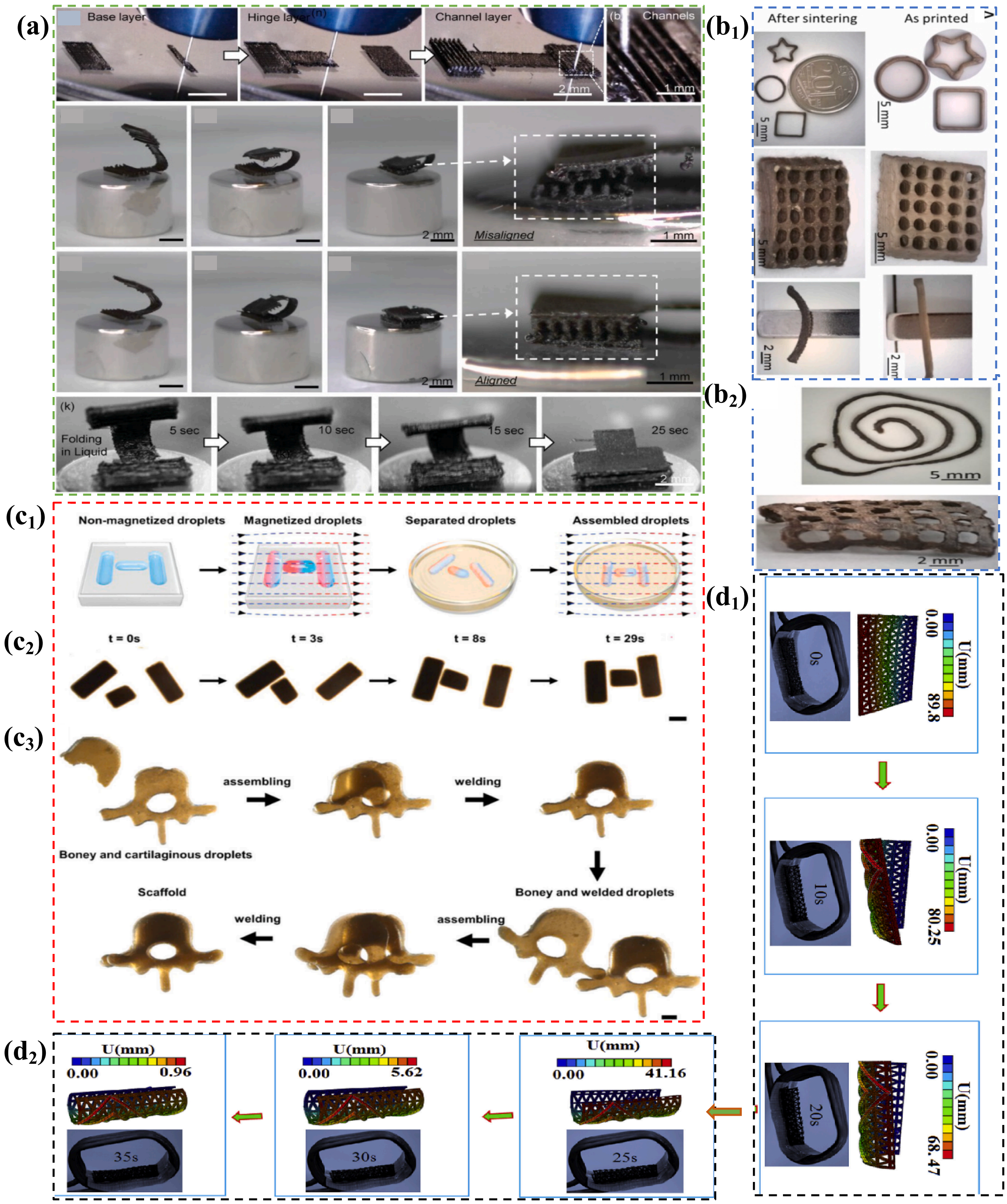

Fig. 23. (a) Scaffold printing and foldability: on a Petri dish via solvent casting DIW and folding as a time lapse are shown with a Fe_3O_4-MNPs with 7 mm long hinge and 15-Fe_3O_4-MNPs base layers (adapted with permission from ref. [500], copyright 2023, Wiley-VCH GmbH); (b_1) Photos of 3D-printed samples as printed and after sintering, (b_2) Complex 3D structures obtained after self-shaping during the sintering process (adapted with permission from ref. [501], copyright 2022, Elsevier B. V.); (c_1-c_2) Fabrication of “H”-shaped liquid blocks through all-liquid molding, magnetizing and patterning, (c_3) Bone-like and cartilaginous liquid blocks were suspended in the oil, and manipulated by external magnetic field to assemble into a spinal column-shaped structure (adapted from ref. [502], under the terms of the Creative Commons CC BY license); (d_1-d_2) Function verification of bioinspired tracheal scaffold in vitro actuated under magnetic field (adapted with permission from ref. [503], copyright 2019, Elsevier Ltd.).

time, it’s referred to as 4D printing. These geometry shifts can be induced in any number of ways, with some of the most common being electrical stimulation, heat, and moisture [551–553]. Mostly DIW and DLP-based 4D printing methods are currently available and studied. However, novel 3D printing techniques such as 2PP and micro-printing may provide a breakthrough in multi-responsive tactics for complex shapes and efficient control over their shape-morphing behaviors [554–556]. In a bid to emulate the movement mechanism of the printed structures, the researchers employed computational design techniques that used selectively printing ‘bend lines’ into the geometry of the multilayer structures [557–560]. Material choice was also crucial in 3D printing as the actuation of the smart material would only be possible

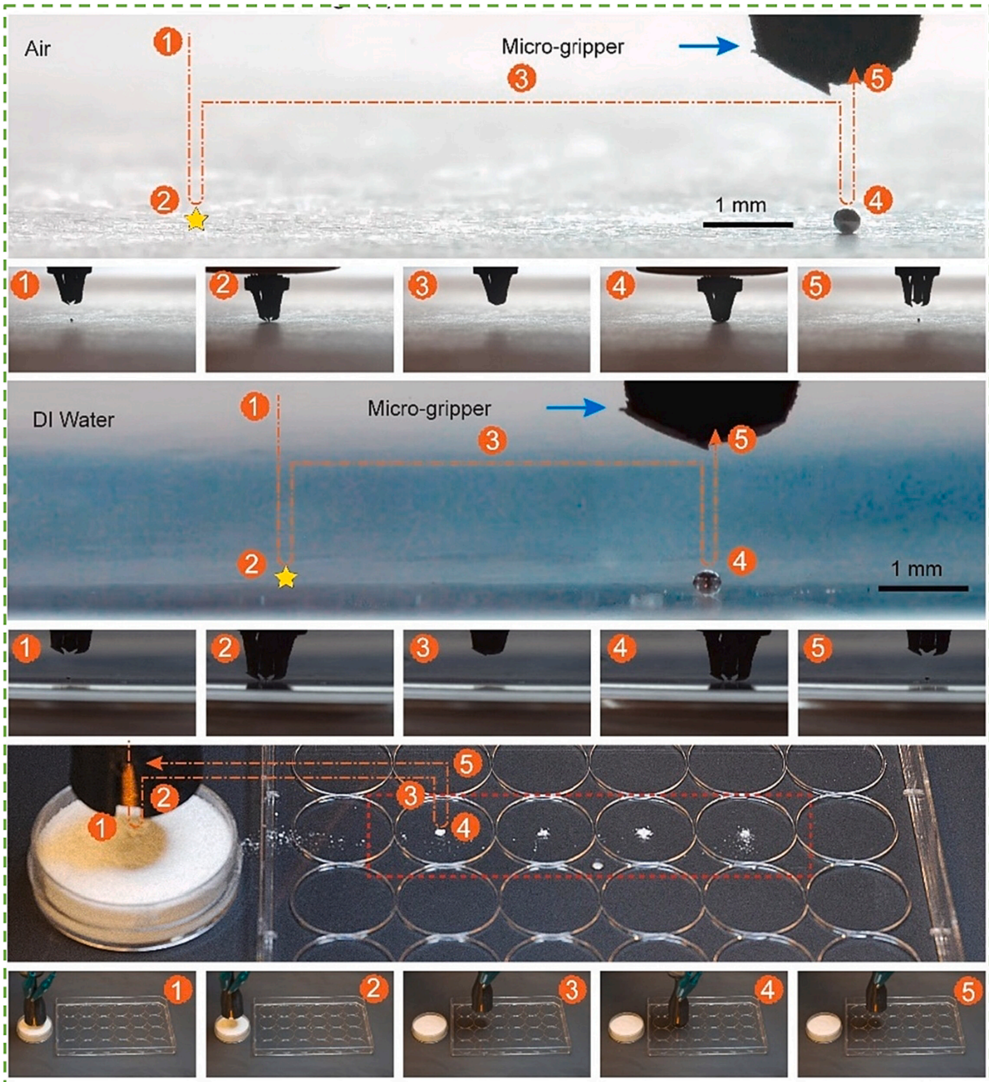

Fig. 24. Manipulating and transporting tin microsphere in various mediums such as in air, in DI water and lastly for salt powders (adapted with permission from ref. [504], copyright 2021, Elsevier B.V.).

with a material responsive to any stimuli. Many 3D-printed objects are pre-programmed to morph using intelligently placed layers and folds, which can contract and expand to give the desired effect [561–563].

Most of the studies discuss only single material-based printing techniques while multi-materials have huge potential in actuators for soft robotics, kirigami/origami and complex structures, and controlled sequential folding [564,565]. Furthermore, 3D printing at the micro-scale has excellent potential to demonstrate various shape-morphing behaviors for the possibility of releasing and trapping micro-objects. Various micro-shapes such as smart box-like 3D microstructures, and microspheres can be useful for high-tech applications such as on-demand drug delivery [566–568]. Also, soft devices are promising candidates in extreme environments where human interaction is not possible. To date, their mechanical properties are not up to the mark and thus 3D-printed soft robotics have limited use [569,570]. The time-dependent thermomechanical properties of soft actuators are also a promising field. Furthermore, the soft actuators support heavy loads only at low temperatures but the load-carrying capability at high temperatures is quite limited [571–573].

Despite their high control precision and robustness, soft magnetic structures make it difficult to design uniform magnetization profiles. Thus, magneto-deformation modes and types are significantly limited. Moreover, it remains challenging to realize complex and diverse magneto-deformations, particularly in hard magnetic materials. Furthermore, the diffusion of particles within the polymer matrix is controlled by external fields applied during printing. Thus, it is very crucial to control particle concentrations spatially and to displace particle accumulations freely during the crosslinking process. Consequently, MPs susceptible to magnetic fields are shifted into previously free regions, offering more degrees of freedom in printed structure [574].

FDM although widely available for producing smart structures has its limitations in nozzle caliber and printer precision particularly for fabricating micro-scale parts [575]. Existing magnetic miniature soft robots are usually fabricated from SLA or 2PP for aching high-shape transformations and locomotive behaviors. However, in the case of DLP various effects such as isotropic magnetization of soft actuators are observed which prevents selective actuation of one portion of the robot, articulated actuation, limits the number of possible degrees of freedom, and shape profiles. Generally, magnetic actuation portfolios are achieved by rationally imputing “logic switch” sequences. However, their performance can be further improved by considering stepwise magnetic controllability, self-healing, multi-responsiveness, and remolding ability [576].

Soft materials such as polymers are prone to structural damage under external factors that affect cracking, embrittlement, external loading,

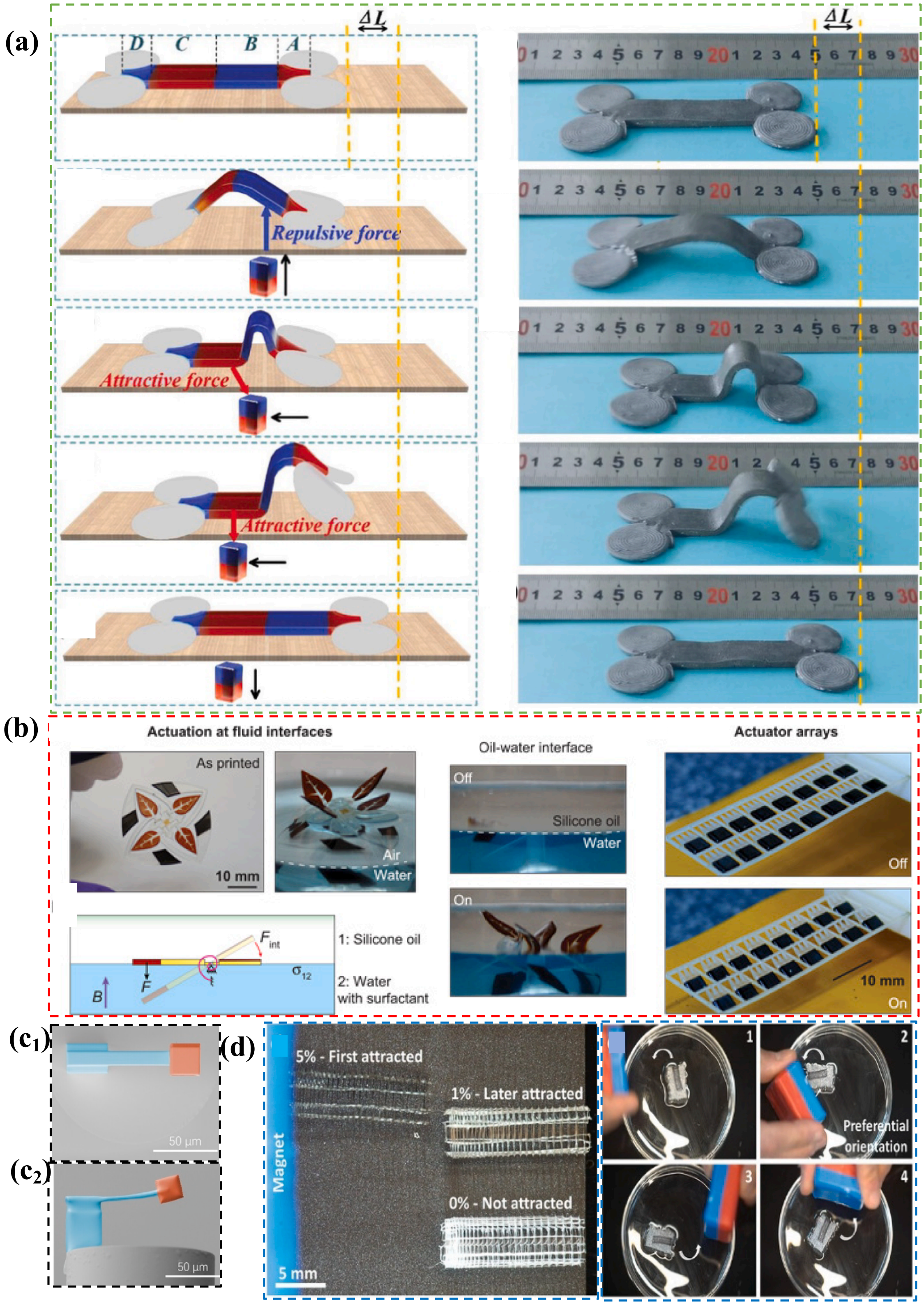

Fig. 25. (a) Schematic and real-time crawling motion of the soft robot at various moments in a cycle (adapted with permission from ref. [516], copyright 2023, Wiley-VCH GmbH); (b) Magnetic actuator arrays capable of deforming under applied field use in liquid interfaces, some panels return to their flat position easily when the water is disturbed. With and without an applied magnetic field, experimental results of actuation at the silicone oil–water interface and an array of 16 identical actuators with serrated edges are presented (adapted from ref. [518], under the terms of the Creative Commons Attribution license); (c_1-c_2) Magnetic microcantilever morphological characterization including false-color SEM images of the magnetic cube (orange)-modified fiber-tip microcantilever (blue) in different views (adapted from [519], copyright 2023, American Chemical Society); (d) Different response with distance and wt% (on the left side) and constant rotation of the 5 wt% Fe_3O_4 toward a preferential orientation facing the magnet from the side with higher mass accumulation (on the right side) (adapted from [520], under the terms of the Creative Commons Attribution license 4.0).

Table 4
Summary of some recent works from 2020 to now on 3D printing of MASMs for soft robotics and novel actuators-related applications.

Year	AM	MASMs	Stimulus	Actuator motion(s)	Targeted application	Ref.
2023	SLA	NdFeB/PEGDA	Magnetic field	Bending	Soft Robotics	[521]
2023	Multi-material extrusion	Conductive PLA/TPU	Magnetic field	Bending and jumping	Soft frog-shaped robot	[522]
2023	3D direct laser printing	FePt/PETA/PNIPAM-AAc	Magnetic and pH	Swelling	Microrobots for on-demand cargo delivery	[523]
2023	Extrusion-based printing	PDMS/$BaTiO_3$/Fe_2O_3	Magnetic field	Bending	Flexible electronic devices	[524]
2023	FDM	Shape memory PU foam composite	Magnetic field	Bending	Soft actuators for grasping the objects	[525]
2023	FFF	Cu-PLA	Magnetic field + temperature + electric field	Grasping objects (bending, twisting, and folding)	Flexible gripper	[526]
2023	SLA	FLGPCL04 polymer/Fe_3O_4	Electric and magnetic field	Stretching	Micropumps	[527]
2023	FFF	PLA/PDMS/NdFeB	Magnetic field	Bending	Superhydrophobic surfaces for droplet control	[528]
2023	DIW	NdFeB/PDMS/MWCNT/rGO	Magnetic field	Curling, bending, folding, and twisting	Targeted drug delivery	[493]
2023	FFF	PEU/PLA/MWCNTs	Electric current	Bending	Soft robotics	[529]
2023	SLA	Water, acrylamide and PEGDA	Magnetic	Swelling	Soft robotics for minimally invasive interventional microsurgery	[530]
2023	SLA/DLP	NdFeB/PDMS	Magnetic field	Twisting and bending	Diagnosis and treatment of occlusions in various circulatory systems.	[531]
2023	DLP	PEGDA	Magnetic field	–	Swimming microrobot	[532]
2023	FDM	Iron particles/PLA	Magnetic field	Gripping and bending	Smart grippers	[533]
2023	FDM	PLA/TPU/Fe_3O_4	Magnetic field	Folding and gripping	Smart actuators	[534]
2023	Extrusion	Iron particles/PEGDA	Magnetic field	Folding and bending	Actuators and soft robotics	[530]
2023	Extrusion	PVA/NdFeB	Magnetic field	Flipping of bilayers (curving of structures)	Tunable mechanical metamaterials	[535]
2023	Extrusion-based printing	Epoxy (EPON 8111) resin and curing agent (EPIKURE 3271)	Magnetic field	Bending	Medical devices such as oxygen masks	[536]
2022	DIW	Carbon/Fe/PDMS	Magnetic field	Rolling and bending	Soft robots for underwater applications	[537]
2022	LAM	Silicone: Ecoflex	Magnetic field	Complex shape morphing structures	Soft robotics	[538]
2022	DIW	PLMC/ PTMC/Fe_3O_4	Magnetic field and heat	Bending	Soft robots	[539]
2022	FDM	PEEK/Fe_3O_4	Magnetic field	Folding and bending	Electrical motors for space-compliant	[540]
2022	DIW	TPU/PCL/Fe_3O_4	Heat and magnetic field	Bending and grasping	Flexible robotics	[541]
2022	SLA	PCL/Fe_3O_4	Electromagnetic field	Deflection of membrane	Micro-actuators	[542]
2022	DIW	CIP/ natural rubber	Magnetic field	Gripping and bending	Soft robotics	[543]
2022	DIW	ALG/MC/PAA/Fe_3O_4	Magnetic field	Rolling, jumping, and bending.	Soft robotics	[442]
2021	FDM	PHB/PCL/CNFs/Fe_3O_4	Magnetic field	Bending	Smart actuators	[544]
2021	FDM	PLA/Fe_3O_4	Magnetic field	Expansion and stretching	Treatment of left atrial appendage occlude	[545]
2020	DLP	Ferrofluid/PDMS	Magnetic field	Bending	Soft gripper	[546]
2020	2PP	GelMA/$CoFe_2O_4$/ $BiFeO_3$	Magnetic field	–	Micro-swimmers for differentiation of neuron-Like cells	[547]-
2020	SLS	PA-12/γ-Fe_2O_3	Magnetic field	Grasping and bending	Smart grippers	[548]
2019	DIW	NdFeB/PDMS	Magnetic field	Gripping and bending	Soft robots for medical applications	[549]
2018	DIW	Iron particles/PDMS	External magnetic field	Bending	Bionic robots	[550]

and eventual functional degradation. This lowers their overall lifespan. This can be avoided through recovering functional performance such as "self-healing" after incurring (minor) structural damage. One way to achieve this is "self-heal ability" using polymer chemistries involving reversible primary and/or secondary bond networks or embedded monomer reservoirs that use bio-inspired features [577]. Focused research is needed on sustainable soft actuators for achieving high performance and mitigating environmental issues in terms of their waste at their end life [578].

There is a huge need for high-end simulation and control platforms to strengthen the real-time application of adaptive 4D-printed systems in various environmental interactions, which is still in demand. Development of sensor-less adaptive 4D printers can be developed in future using reversible multi-stable compliant mechanisms. Moreover, rising artificial intelligence and machine learning techniques can also play a pivotal role in improving the functionality of smart devices by optimising the 3D printing theoretical design parameters for the efficient designing of application-specific devices [579].

With the need to manipulate smaller objects in confined spaces, robotic grippers are increasingly becoming miniaturized. With increasingly smaller grippers, it faces challenges in microfabricating, assembling, and actuating them. Although flexible actuators provide excellent performance, some of them require external wires to connect to a power source or require higher ambient temperatures, limiting their application [580]. Actuators for modern-day robots are evolving for improved power efficiency, topology, and size, optimizing for weight and other performance metrics [581]. 3D printing has revolutionized many industries [582], but its integration with sensors and robotics is still at an embryonic stage. It needs emerging printing techniques for proper embedding sensors and actuators into 3D-printed objects.

Recently, the emergence of 2D materials allows us to achieve high mechanical properties of 3D complex structures by mixing 3D printing and 2D materials such as graphene montmorillonites, carbon nanotubes, cellulose nanocrystals, carbon nanofibers, and so on, thereby forming shape memory polymeric nanoarchitectures [583] generally through DIW [584]. These novel 2D materials even at low concentrations such as 0.1 wt% graphene nanoplatelets improved significantly shape recovery behavior [585].

A great deal of progress has already been made with stimulus-responsive magnetic actuators. For further improvements in their functions and to broaden their practical applications, there is still much to be done, as summarized in Fig. 26**.** First, the 4D stage of soft actuators

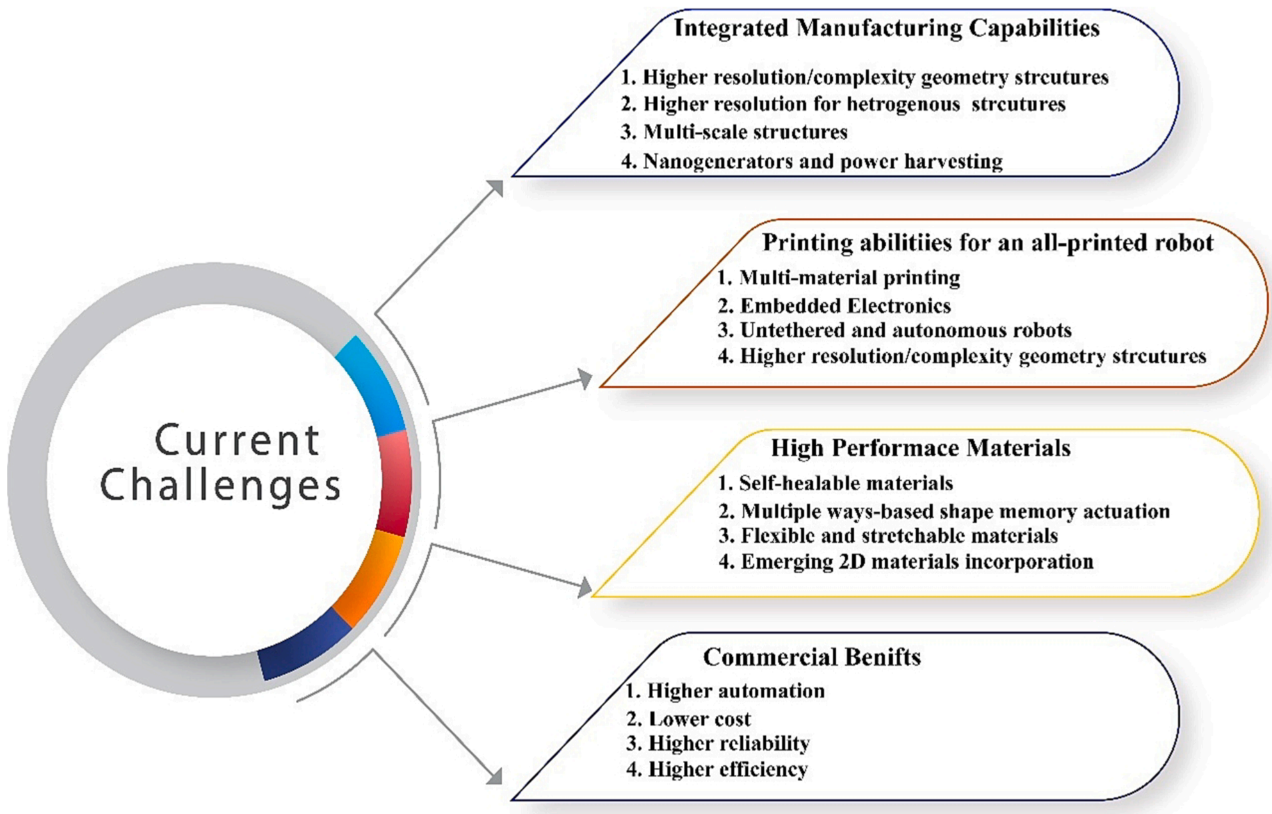

Fig. 26. Future roadmap for advanced sensors and soft robotics application in 4D printing self-healing.

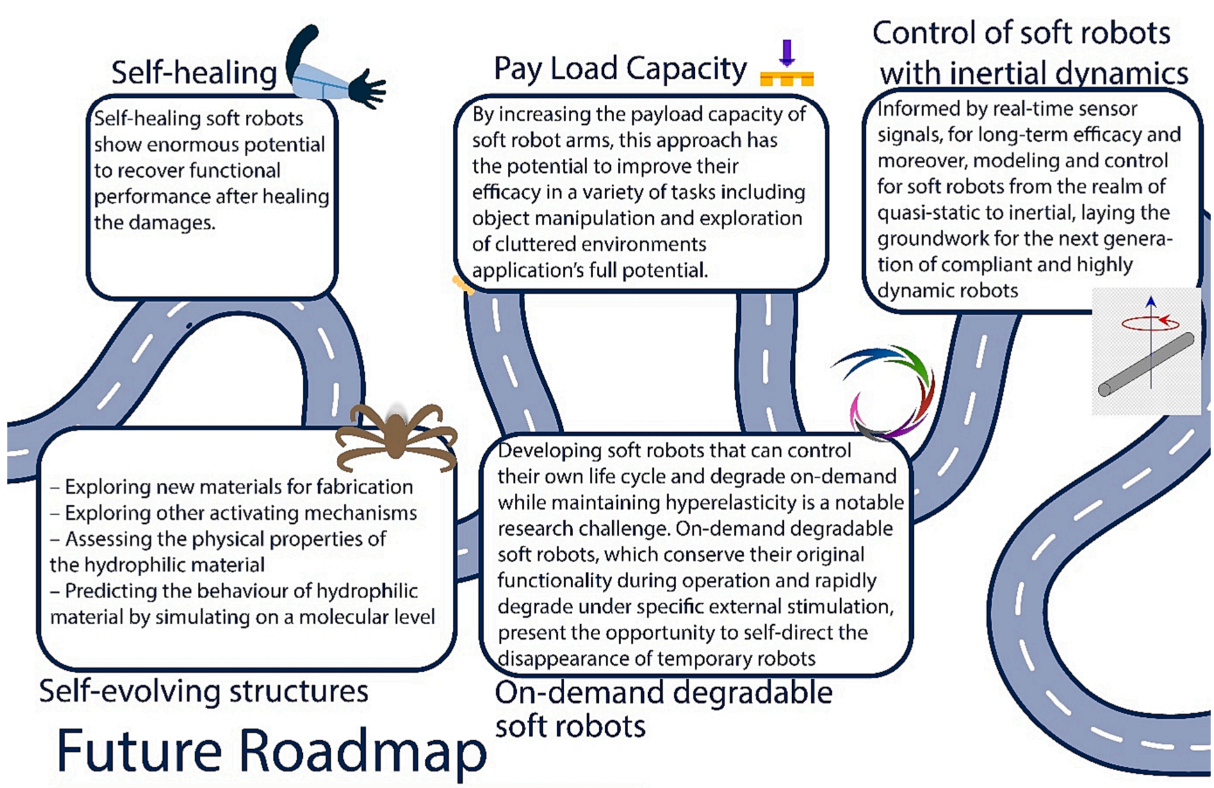

Fig. 27. Future roadmap for advanced sensors and soft robotics application in 4D printing self-healing [600], payload capacity of soft robot arms [601], rapid modeling and control [602,603], and degradable soft robot [604,605].

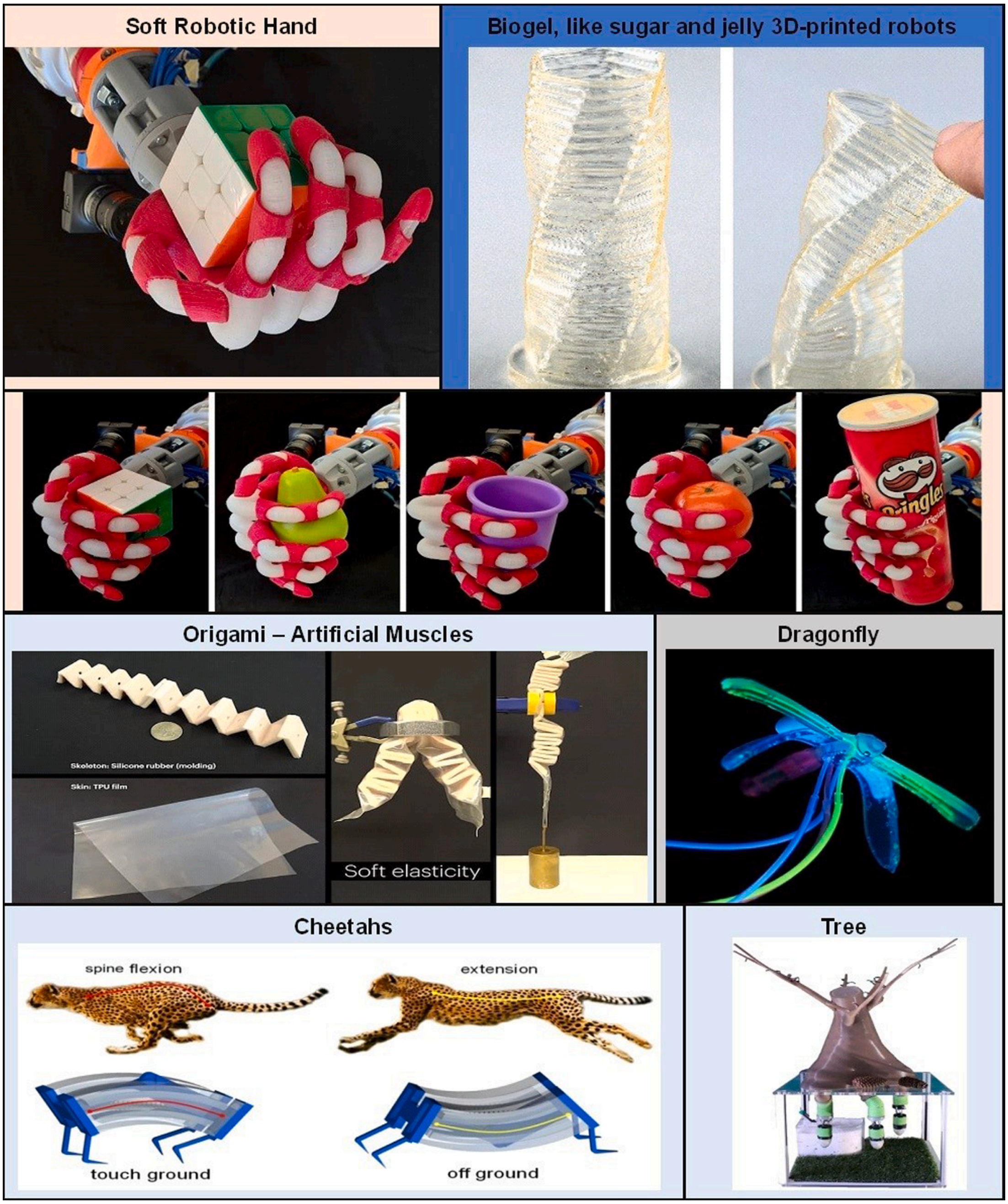

Fig. 28. Emerging soft robotics in various shapes including soft robotic hand (adapted from [606] (Pic credit Elvis et al.), Biogel, like sugar and jelly 3D-printed robots (Credits: A. Heiden et. at the Johannes Kepler University), Origami inspired Artificial Muscles and Origami Gripper, Dragonfly (Pic Credit: DraBot of Duke University), Cheetahs (Pic credit: North Carolina State University), Tree (Pic Credit: Plantoid IIT Italian Institute of Technology) (Various figures are adapted from [607]).

is not mature enough to realize practical applications. However, overcoming the main bottleneck including the fabrication of large parts and, mainly, the regulated transformations and movement of actuator parts under external stimuli can pave the foundation for more practical applications [586]. Second, it is still a challenge to produce more complex deformations for precisely controlling their local stimuli response, particularly material handling. It is expected that mag-bots used in remote, confined spaces with more complex designs for various purposes such as material handling [587]. Third, besides macroscopic deformation, changes in their other macroscopic properties such as color change could also be useful for opening many avenues [588]. Fourth, commercialization of the printed actuators involves the synthesis of novel SMP characterized by various types of response and advanced printing skills, all of which are a major part of 4D printing [589–591]. Due to the lack of soft materials, their commercial introduction is still at an early stage. Thus, significant attention needs to be paid to the variety of 3D printers and the availability of smart materials for 4D printing perspective.

From laboratory evaluation to clinical application, safety aspects and regulatory pathways should be considered. Due to the complexity of the human body, future research should increasingly focus on the clinical use of microrobots as well as nanorobots [592] for alleviating various challenges related to them such as detoxification, biocompatibility [593], biological barriers, biosensing, biodegradation propensity and functioning in complex biological fluids [594–596]. Biomedical applications often require magnetic soft robots to navigate in unstructured aquatic-terrestrial environments [597,598]. Furthermore, for precise positioning and efficient operation, the miniature magnetic robot needs to be enhanced both in terms of controllability and agility. Recently, a 4D printed shape-programmable soft robot with near-infrared light and magnetic stimulation was effectively employed for remote manipulation of placing drugs, particularly in the application of hazardous chemical operations [599]. For future research, we anticipate that several challenges related to the following areas need to be addressed, as summarized in Fig. 27**.** This will improve the functionality as well as the performance of today's state-of-the-art soft robotics (referring to Fig. 28) for many unknown applications.

6. Summary

Interestingly, we can learn a lot about shape morphing behavior of smart materials by drawing inspiration from nature. In this review, we have highlighted various 3D printing methods; new MASMs, and fabrications of various functional structures including sensors, and soft actuators, for broad applications in flexible electronics and biomedical. Particularly, this review study focuses on the justification of 3D printing of smart materials under magnetic stimulus for developing the state-of-the-art in soft robotics and providing recent breakthroughs in the proposed field. The 3D printing technology is replacing many traditional manufacturing techniques in the development of unthinkable, complex shapes and multifunction advanced sensors and actuator applications. It has been observed that the potential of 3D printing in the development of soft robotics has been significantly expanded due to emerging materials such as LCEs, polymers and their composites, and hydrogels for producing advanced intelligent devices. Furthermore, explications of the shape morphing mechanisms such as bending, twisting, and folding are easily achievable under the magnetic stimulus, which permits the printed actuators to gain control of their various soft robotics functions. Lastly, we provided some of the current 3D printing challenges such as low mechanical properties, response under multi-programming and stimuli that need to be addressed in future studies. Finally, we provide future perspectives, for the designing of the next generation of 3D-printed biodegradable and sustainable soft robots with much higher payload capacity. Thus, there is significant improvement required in the arena of 3D printing of MASMs, with more focused research towards its practical applications.

CRediT authorship contribution statement

Muhammad Yasir Khalid: Writing – original draft. **Zia Ullah Arif:** Writing – original draft, Writing – review & editing. **Ali Tariq:** Writing – original draft. **Mokarram Hossain:** Supervision, Writing – review & editing. **Kamran Ahmed Khan:** Supervision, Writing – review & editing. **Rehan Umer:** Supervision.

Declaration of Competing Interest

The authors declare that they have no known competing financial interests or personal relationships that could have appeared to influence the work reported in this paper.

Data availability

No data was used for the research described in the article.

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