

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.

Keywords: 3D printing, 4D Printing, magneto-active materials, soft robotics, smart actuators

Highlights

1. Magneto-active soft materials (MASMs) are novel smart materials for multifunctional robotics applications.
2. Highlighting the contemporary trends of 3D-printed MASM-based soft robotics.
3. Incorporating the future research directions of 3D-printed MASMs.

List of Symbols

2D	Two-dimensional
2PP	Two photon polymerization
3D	Three-dimensional

47	4D	Four-dimensional
48	AAc	Acrylic acid
49	ABS	Acrylonitrile butadiene styrene
50	ALG	Alginate
51	AM	Additive manufacturing
52	BJ	Binder jetting
53	CA	Cellulose acetate
54	CIP	Carbonyl iron particles
55	CNF	Cellulose nanofiber
56	CNT	Carbon nanotube
57	DMAA	N,N'-dimethyl acrylamide
58	DLP	Digital light processing
59	DIW	Direct ink writing
60	DLW	Direct laser writing
61	FDM	Fused deposition modelling
62	FFF	Fused filament fabrication
63	FePt	Iron platinum
64	FASMC	Flexible anisotropic soft-magnetic composite
65	GelMA	Gelatin methacryloyl
66	LCE	Liquid crystal elastomer
67	MAHs	Magneto-active hydrogels
68	MAPs	Magneto-active polymers
69	MASMs	Magneto-active soft materials
70	MC	Methylcellulose
71	MJ	Material Jetting
72	MPs	Magnetic particles
73	MRE	Magnetorheological elastomers
74	MNPs	Magnetic nanoparticles
75	MWCNTs	Multiwalled carbon nanotubes
76	NdFeB	Neodymium-iron-boron
77	NIR	Near-infrared
78	PA	Polyamide
79	PAA	Poly(acrylic acid)
80	PAAM	Polyacrylamide
81	PBF	Powder bed fusion
82	PCL	Polycaprolactone
83	PDA	Polydopamine
84	PDMS	Poly(dimethylsiloxane)
85	PEEK	Polyether ether ketone
86	PEG	Polyethylene glycol
87	PEGDA	Polyethylene glycol diacrylate
88	PETA	Pentaerythritol triacrylate
89	PEU	Polyester urethane
90	PHB	Poly-hydroxybutyrate
91	PLA	Poly(lactic acid)
92	PLMC	Poly(D,L-lactide-co-trimethylene carbonate)
93	PNIPAM	Poly(N-isopropylacrylamide)
94	PP	Polypropylene
95	PTMC	Poly(trimethylene carbonate)
96	PU	Polyurethane
97	PVA	Poly(vinyl alcohol)
98	PVC	Poly(vinyl chloride)
99	SDM	Shape deposition manufacturing
100	SEM	Scanning electron microscopy
101	SF	Silk fibroin

102	SME	Shape memory effect
103	SMP	Shape memory polymer
104	SMPC	Shape memory polymer composite
105	SLA	Stereolithography
106	SL	Sheet lamination
107	SLM	Selective laser melting
108	SLS	Selective laser sintering
109	SPIONs	Superparamagnetic iron oxide nanoparticle
110	rGO	reduced graphene oxide
111	TPR	Thermoplastic rubber
112	TPU	Thermoplastic polyurethane
113	UV	Ultraviolet
114	VP	Vat Photopolymerization

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143 1 Introduction

144 Under constant evolution, the ambition to drive and pursue modern technologies has
145 significantly improved today's living standards. This has happened because of scientific
146 progress, leading towards transformation in many areas including materials, their synthesis
147 techniques, and properties characterization, thus, opening a new paradigm for many novel
148 applications [1]. Three-dimensional (3D) printing or additive manufacturing (AM) is regarded
149 as a novel and emerging manufacturing technique for many materials and it is now being
150 imposed in scale-up on an industrial scale [2]–[6]. 3D printing is also drawing attention from
151 researchers due to its ability to produce complex parts with higher accuracy, adaptability and

152 availability all over the world [7]–[10]. Various 3D printing techniques such as ink-based, light-
153 based and laser-based are introduced [11]–[13] and performed significantly for various
154 materials such as polymers [14], elastomers [15], metals [16], and polymer composites [17].
155 Ink availability, balancing printing quality including layer thickness, and layer height are some
156 of the important design criteria in 3D printing [18]–[20]. From a sustainability perspective, 3D
157 printing has so much to offer, for instance, various natural biomaterials [21]–[23] can be used
158 as a potential ink source for exciting applications without creating any waste [24]–[26].
159 Moreover, 3D printing of composite materials has improved mechanical properties than
160 traditional composites [27]–[29]. This technology has provided the opportunity for multi-
161 material printing which includes two or more different materials as well as solid material into a
162 medium, creating a suspension for desired ink for any geometry [30]–[32]. Many complex
163 structures such as helical coils, origami, and kirigami-inspired structures, and functionalized
164 micro-architectures can be printed with extreme accuracy [33]–[38].

165 3D printing has opened up many interesting avenues for real or practical applications as well
166 as continuously thriving for new platforms for incorporating many emerging materials including
167 nanomaterials for achieving wide goals for a broader community perspective [39]–[42].
168 Recently, during the coronavirus disease 2019 (COVID-19) pandemic, 3D printing also played
169 its part by fabricating personal protective equipment [43]–[45]. Other biomedical applications
170 of 3D printing include patient-specific models that can be used to train medical staff and
171 improve patient consent and understanding, wearable devices such as orthotics and
172 prosthetics [46]–[48], tissue engineering [49]–[52], drug delivery systems [53]–[55] as well as
173 gadgets to make life easier [56]–[58].

174 The use of 3D printing is growing in almost every field including analytical chemistry [59],
175 microfluidic devices [60], and detection of analytes for medical diagnosis [61], electrochemical
176 sensors [62], and system health monitoring [63], [64]. However, the cost, limited print
177 materials, the need for post-processing of devices [65], and the need for higher resolution still
178 limit the broader application of this technology. One of the significant drawbacks is that printed
179 functions remain static after 3D printing which limits its applications in some of the novel areas
180 where many printed functions, such as self-healing ability, elastic conductivity, and shape-
181 morphing mechanism in many devices (e.g., wearable electronics, soft robots, and flexible
182 biosensors) performances are required [66]–[69]. Among all these 3D-printed drawbacks, the
183 shape-morphing behaviors of printed materials have paramount importance in advanced
184 engineering applications [70]–[72]. Lately, an improved form of AM relatively inspired by
185 shape-morphing behaviors in nature, the four-dimensional (4D) printing technique has been
186 introduced [73]–[75]. 4D printing can also be defined as using smart materials for adopting
187 external stimuli in the 3D printing research division [76]–[80]. Researchers have developed a
188 4D printing technique for gaining more accurate control of the shapes of printed parts such as
189 shrinking, swelling, folding, bending, rolling, origami, twisting, or locomotion under various
190 stimuli [81]–[86].

191 Recently soft actuators and robotics have been studied extensively [87]. Soft robotics have
192 some unique capabilities in comparison to traditional robots such as constantly changing
193 stiffness and shape morphing ability for performing specific tasks such as grasping and lifting
194 toxic or hazardous objects under extreme environmental conditions [88]–[91]. In fact, shape
195 compliance of soft actuators provides a viable avenue to address many unsolved problems of
196 today [92]–[96]. The fabrication of soft robotics through the conventional synthesis route is
197 tedious and time-consuming, and more importantly, its shape-morphing behavior is not
198 satisfactory. To date, various synthesis routes have been used for fabricating soft robotics,
199 including solvent casting [97], lithography [98], roll-to-roll technology [99], laser heating [100],
200 spraying with spin technique [101], magnetized modules assembly with dynamic covalent
201 bonds [102], electron beam lithography of nanomagnets [103] and bonding agent [104], [105].
202 Among them adding magnetic particles (MPs) to 3D-printed smart material is a promising and

203 innovative way to achieve highly functionalized soft actuators [106]–[108]. To date, various
 204 magnetic materials such as electrical steel (FeSi), iron oxide (Fe₃O₄) and carbonyl iron
 205 particles (CIP) have been added to many shape memory materials. In the presence of
 206 magnetic field strength, the 3D-printed magnetic actuated soft robotics exhibited unique
 207 phenomena for changing their shape, structures as well and properties which are beneficial to
 208 many applications [109]. This unique 3D-printed magnetic actuation attribute with desirable
 209 performances is an ideal choice for practical application in the healthcare sector [110] like
 210 targeted drug delivery and tissue engineering [111]–[114]. Moreover, magnetically actuated
 211 actuators with remote magnetic steering capabilities have also proven their potential in
 212 minimally invasive medical procedures [115]–[118]. **Figure 1** summarizes the key features
 213 found today in soft robotics and their exciting role in many diverse applications.

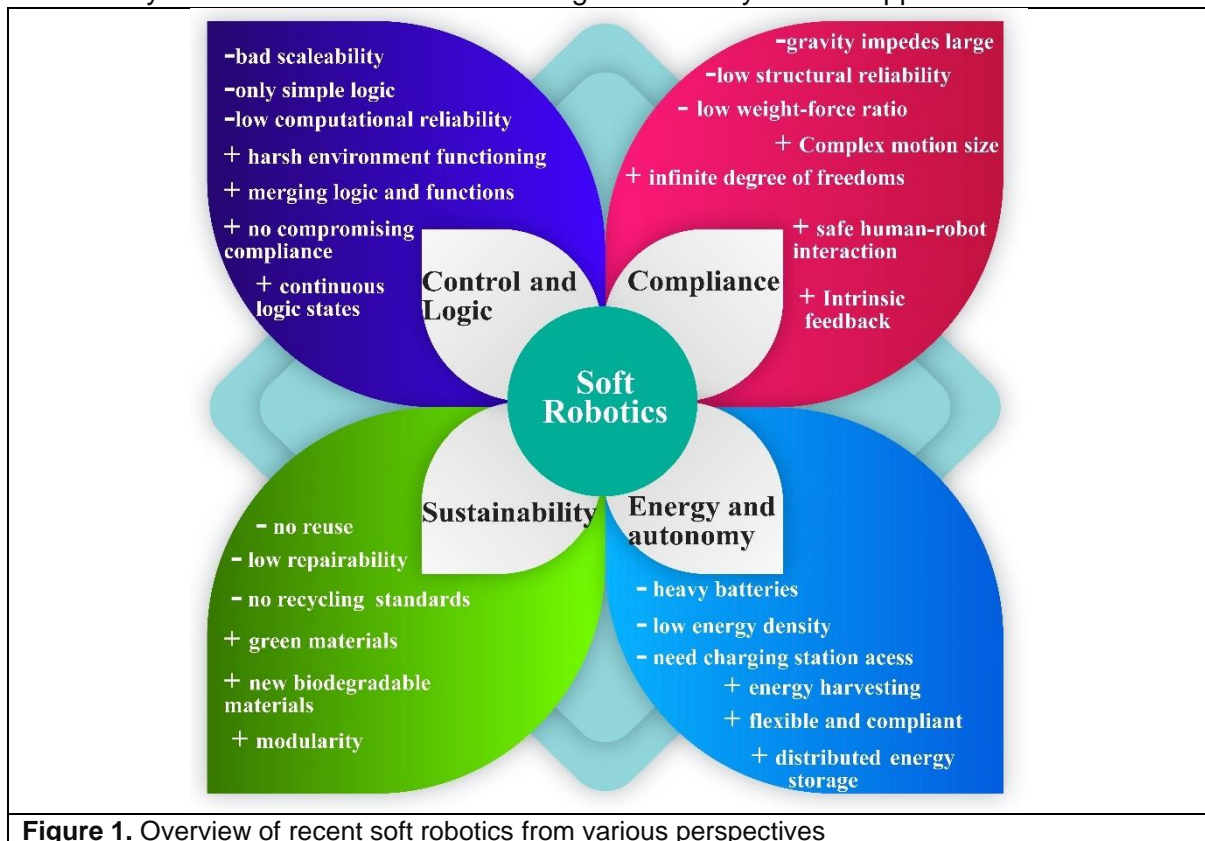
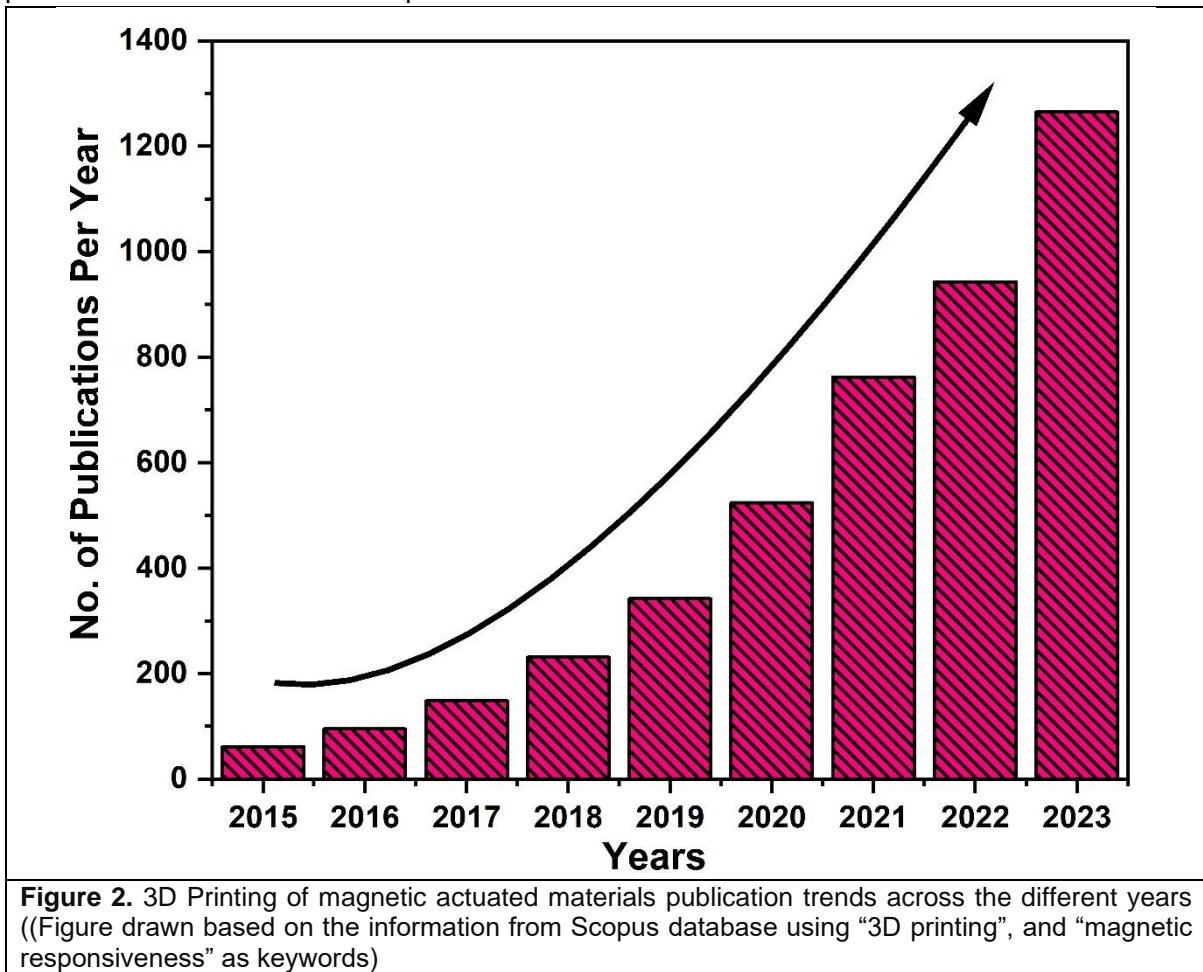


Figure 1. Overview of recent soft robotics from various perspectives

214 1.1 Scope of Review

215 Considerable progress has been made in the design of high-performance soft actuators.
 216 Herein we have provided some guidelines based on the latest research studies on how to use
 217 the power of 3D printing of smart materials in making high-performance novel devices such
 218 as smart grippers, wearable electronics, stretchable ionotropic devices, and many intelligent
 219 devices from AM techniques, and smart materials point of views. The broad aim of this review
 220 is to i) stipulate an exhaustive overview of 3D printing of magneto-active polymers, ii) identify
 221 key smart materials employed for magnetic actuation and their key mechanisms for exciting
 222 applications, iii) propose a series of guidelines for tackling future challenges and highlighting
 223 existing scientific and technological gaps in the field, and iv) discuss potential opportunities for
 224 fabricating high-performance soft robotics towards practical applications. **Figure 2** shows the
 225 publication trends of 3D printing of smart materials under magnetic stimulus across the
 226 different years and significantly publication trends proving that there is a need for a systematic
 227 review to summarize the novel studies. Furthermore, we develop this review by highlighting
 228 the key aspects of various published studies related to this emerging field and adapting a
 229 systematic approach for balancing between the 3D printing technology and the performance

230 of printed devices. **Table 1** provides a brief comparison between a current review and recently
 231 published reviews on similar topics.



232 **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	-	-	-	✓	-	-

233 **1.2 Smart materials for 3D printing**

234 Smart materials can perceive and respond under normal conditions related to their
 235 surrounding environment; however, these materials are unable to improve or optimize their
 236 response when sudden change has happened in their surrounding environment [124].
 237 Whereas intelligent materials can adapt to those changes, and can respond well accordingly
 238 and purposefully, for improving and optimizing their response [125]. Smart and intelligent
 239 materials are under constant evolution for their applications in various artificial actuators. The

240 motions of these actuators are inspired by nature such as life-like motions for bioinspired
241 robotics [126]–[128]. Moreover, these materials can offer functionalities beyond traditional
242 ones particularly for developing unique actuators due to their ability to adapt easily and deform
243 according to the environment. Smart materials also include self-healing materials, self-
244 transforming materials, their auxetic behavior, softening and hardening behaviors under
245 compression and tension, action-at-a-distance phenomena and respond overtime to assemble
246 into new compositions via bending, spreading, twisting, shrinking, and folding [129]–[131].
247 These dynamic functions of smart materials are teamed with the 3D-printed complex
248 geometries of parts for soft robotics, advanced actuators, biomimetic devices, and self-
249 deployable structures applications [132]–[134].
250 Shape-memory materials are the type of smart materials which trigger their response under
251 the environmental stimulus, without relying on the application of an external force [135]–[138].
252 Different shape memory polymers (SMPs), liquid crystal elastomers (LCEs), hydrogels, and
253 shape memory polymer composites (SMPCs) are effectively used for the fabrication of flexible
254 devices through 4D printing [139]–[141]. It is worth mentioning that among all the SMP, SMPC,
255 and the role of multifunctional hydrogels are highly effective in the development of novel smart
256 structures [142]–[144]. Various two-dimensional (2D) materials such as graphene, and carbon
257 nanotubes (CNTs) can further improve the shape memory effect (SME) of these smart
258 materials [145].

259 **2 3D Printing**

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

267 3D printing techniques are characterized by contact-based and contactless methods. Fused
268 deposition modelling (FDM), material jetting (MJ), and direct ink writing (DIW) come under
269 contact-based methods [158], whereas the photopolymerization process, powder bed fusion
270 (PBF), and direct energy deposition, are common contactless technologies for 3D printing
271 [159]–[161]. Of all these techniques, stereolithographic (SLA) and FDM are the most employed
272 processes. FDM includes high-temperature nozzles for feeding the filament, and later
273 depositing layer-by-layer sheets of a melted layer with high fabrication speed [162]. FDM also
274 has significant advantages such as versatility and affordability for all types of structures (small
275 to large) and less expensive 3D printing techniques [163]–[166]. Moreover, a wide variety of
276 inks in DIW can be deposited onto arbitrary substrates with random or even complex
277 geometries. Thus, sometimes it is interpreted as a powerful technique for fabricating advanced
278 and sophisticated electronic equipment with high resolution [167]–[169]. However, the
279 possibility of needle clogging during the low speed and high shear forces are some major
280 drawbacks of FDM [170]. Fused filament fabrication (FFF) is also considered as simplest and
281 most widely used 3D printing technology for a large variety of thermoplastic materials at low
282 cost for multi-material 3D printing for various applications [171]. Another popular 3D printing
283 commonly employed is SLA. It has customizability and the ability to print complex geometries
284 through the step method of photo polymerization, scanning the liquid UV-curable matter with
285 a laser [172]–[175]. This permits high print resolution and excellent speed that may be greater
286 than FDM. Furthermore, SLA is extremely suitable for the fabrication of customized soft
287 robotics for wearable applications [176]–[178]. **Figure 3** illustrates the working principles of
288 various AM technologies, which are used to print MASMs. Moreover, increasing

289 miniaturization and higher demand for microfabrication scale has diverted the attention of
 290 researchers towards micro and nano-printing techniques [179] such as two-photon
 291 polymerization (2PP) also referred to as direct laser writing (DLW) [180]. In this technique, a
 292 photo-reactive resin is exposed to high-energy femtosecond laser beams and provides
 293 excellent spatial resolutions in the range of 100 nm [181]–[183]. **Table 2** highlights the key
 294 aspects of current AM technologies. **Table 3** summarizes the key benefits achieved by soft
 295 robotics using AM technologies.

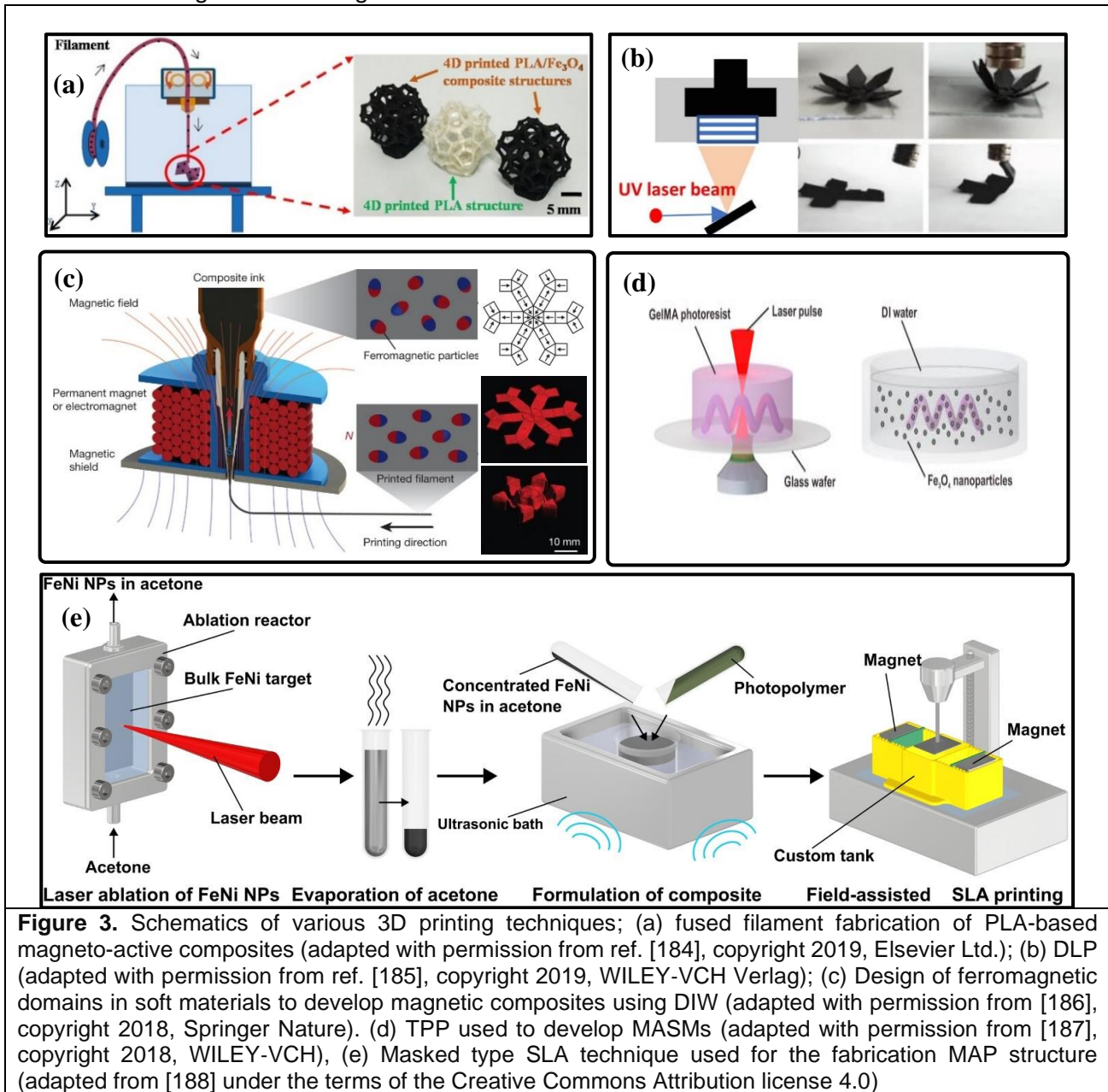


Figure 3. Schematics of various 3D printing techniques; (a) fused filament fabrication of PLA-based magneto-active composites (adapted with permission from ref. [184], copyright 2019, Elsevier Ltd.); (b) DLP (adapted with permission from ref. [185], copyright 2019, WILEY-VCH Verlag); (c) Design of ferromagnetic 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)

296 **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	Plastic in melt form is extruded	Thermoplastics, hydrogel, liquid polymer, and	0.050–0.400	100-600	Dependent on geometry, materials and	(\$300) low cost for home use and high	[189]

FDM	through a nozzle	colloidal suspension		100-150	dissolvable supports can be used	for professional use (\$2000–\$8000)	[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	\$100k-\$250k	[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]

297

298 **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	[215]

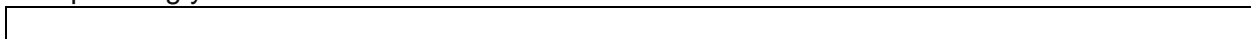
					embedded with multiple sensing capabilities are highly promising for intelligent soft robotics applications.	
FDM	TPU	-	23724.8 2 mm ³	Omni-purpose soft gripper	The proposed 3D-printed soft gripper has a maximum payload to weight ratio of 7.06, a grip force of 31.31 N, and a tip blocked force of 3.72 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]

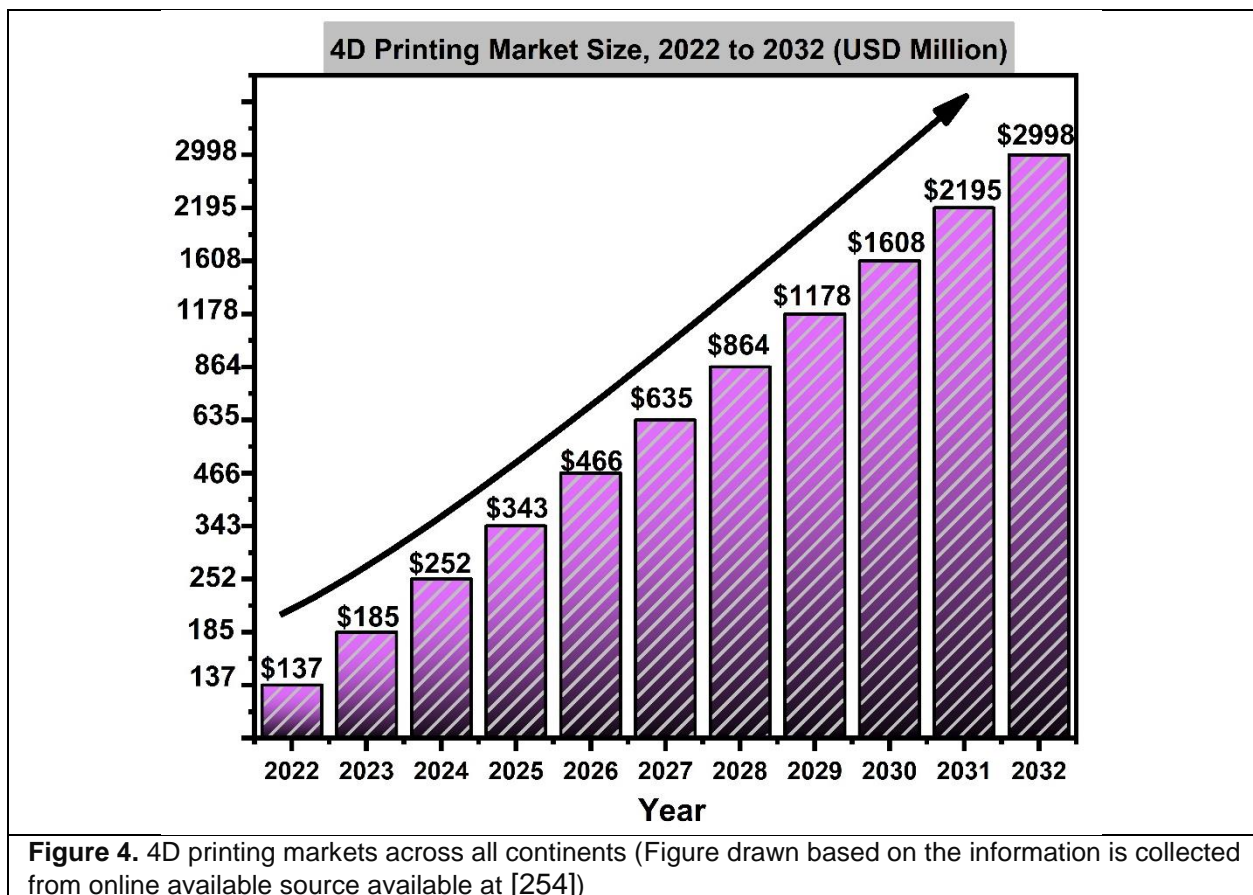
FDM	PVC sheets	-	-	Soft prosthetic finger	The reported results showed that the stiffness of the 3D-printed soft finger was increased by 40 % by linearly driving the stiffness augmenting unit.	[224]
Inkjet Printing	Urethane and epoxy	Multi-material UV-curable printing	80 × 5 × 5 mm ³	Tri-legged soft robot 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]

299

300 2.1 4D Printing

301 Considerable progress in 3D printing technology was achieved by MIT researchers in 2013 by
302 introducing a shape-morphing capability into 3D-printed objects termed 4D printing [228]. It
303 was made possible by the rapid expansion of smart materials, commercial 3D printers, and
304 stimulant environments such as light, temperature, pH, humidity, magnetic and electric fields
305 [229]. 4D printing enables a higher degree of freedom and flexibility in terms of printable
306 geometry [230]–[232]. Moreover, 4D printing integrates the product’s blueprint into a flexible,
307 and intelligent material [233]–[235]. The term “4D” refers to alive structures obtained from
308 traditional 3D-printed structures and means the printed structure can change at least one of
309 its key features such as design, color, property, or functionality over a period under a stimulant
310 environment [236]. This opens a new paradigm for new application arenas for their multi-
311 functional behavior including SME, complex rapid deformation requirements [237],
312 reconfigurable structure, actuation, and sensing under stimulant environments for a broad
313 variety of applications such as soft robotics [238], shape-memory structures [239], advanced
314 actuators [240]–[242], tissue engineering [243], targeted drug delivery [244], [245], cell-laden
315 structures [246], self-deployable structures for aerospace applications [247]–[249], and many
316 more [250]–[253]. **Figure 4** shows the 4D printing market forecast across all the continents in
317 the upcoming years.





318 Smart or stimuli-responsive materials have contributed towards 4D printing by integrating
 319 existing 3D printing techniques [255]–[257]. The smart materials in 4D printing are classified
 320 into many sub types such as thermosets and thermoplastic polymers [258], [259], various
 321 biomaterials [260], [261]. Polylactic acid (PLA) [262], [263], polyvinyl alcohol (PVA) [264],
 322 polycaprolactone (PCL) [265], polyurethane (PU) [266], and hydrogels [267] are mainly
 323 considered smart materials for fabricating highly responsive soft actuators at both the macro
 324 as well as micro levels [268]. 4D printing further harnesses the fabrication of soft actuators,
 325 controllable structures, soft robotics, and many functional devices [269]–[271].
 326 4D printing brings exciting functionalities to smart sensors including environment self-
 327 adaptation, self-sensing, and self-healing [272]–[275]. Recently, Ren et al. [276] introduced a
 328 highly versatile smart tactile sensor through 4D printing using nanocarbon black/PLA
 329 composites and shape-memory PU. These sensors demonstrated unique adjustable
 330 measuring range and sensitivity by changing the electrode height and spacing produced by
 331 the SMP deformation under heat treatment. The shape-changing tactile sensor is regarded as
 332 an ideal match for producing self-adjustment and self-adaptation for human-robot cooperation
 333 in sensing. To date, various emerging materials such as LCE and different hydrogels are used
 334 in 4D printing [277]–[282]. **Figure 5** depicts the emerging applications of 4D printing for
 335 sensors and actuator applications. For example, many hydrogels such as
 336 polydimethylsiloxane (PDMS) swelled anisotropically under multiple stimuli in an assembly of
 337 bistable elements [283], [284]. However, these bistable elements need to be exposed to a
 338 mechanical load for their second stable state. Sometimes, mechanical intervention is also
 339 imperative for switching the second stable state of these materials to activate the snap-through
 340 capacity [285]–[287]. High-performance printing inks are a key factor for temperature-sensitive
 341 materials, which produce a response aligned with outer temperature change [288]–[290]. For
 342 developing highly flexible electronic devices, temperature-dependent materials are commonly
 343 utilized, which generate resistance changes under the temperature change either regular

344 positive or negative responses, for example, the conductivity of typical electronic
 345 semiconductors, conductors, and ionic conductors [291], [292].

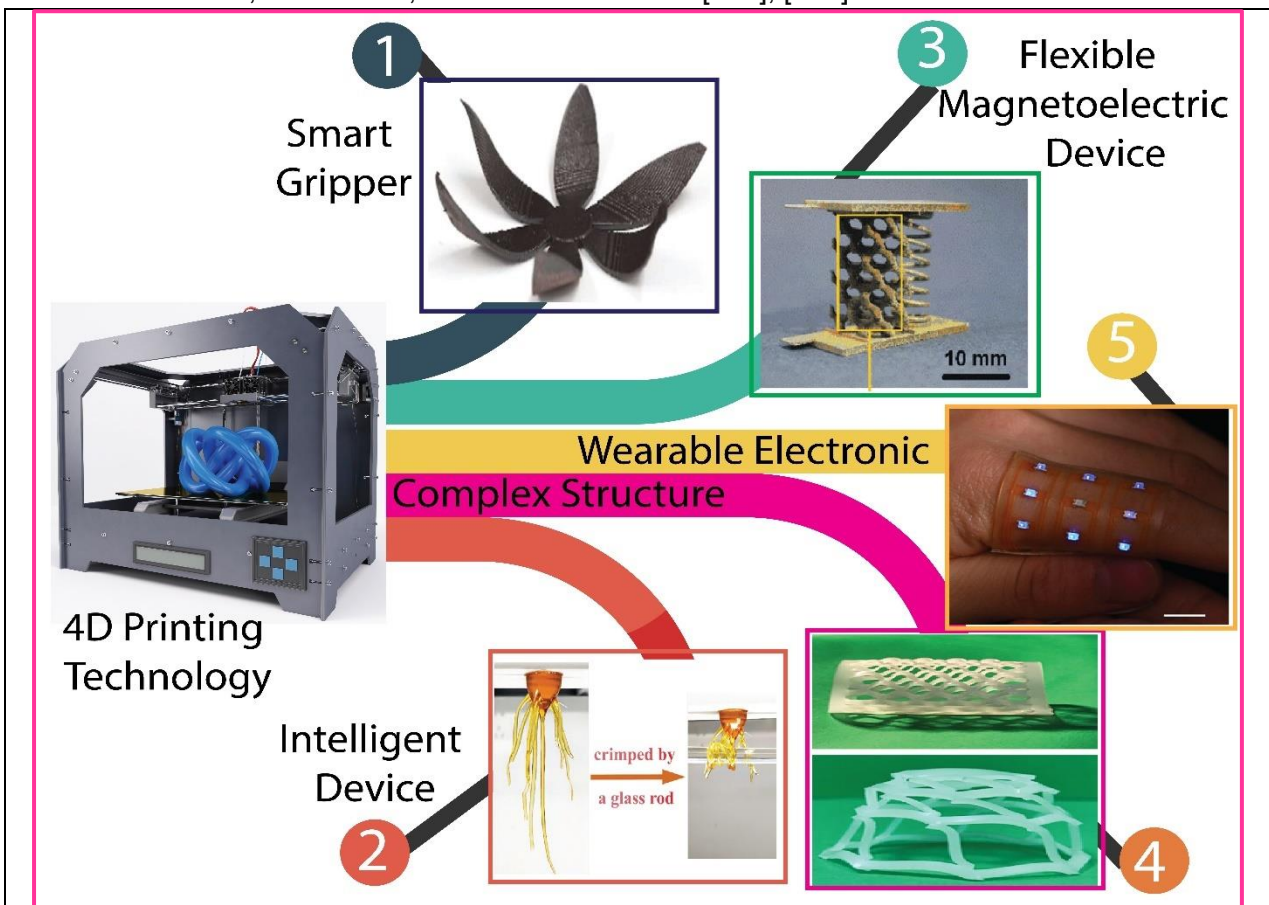
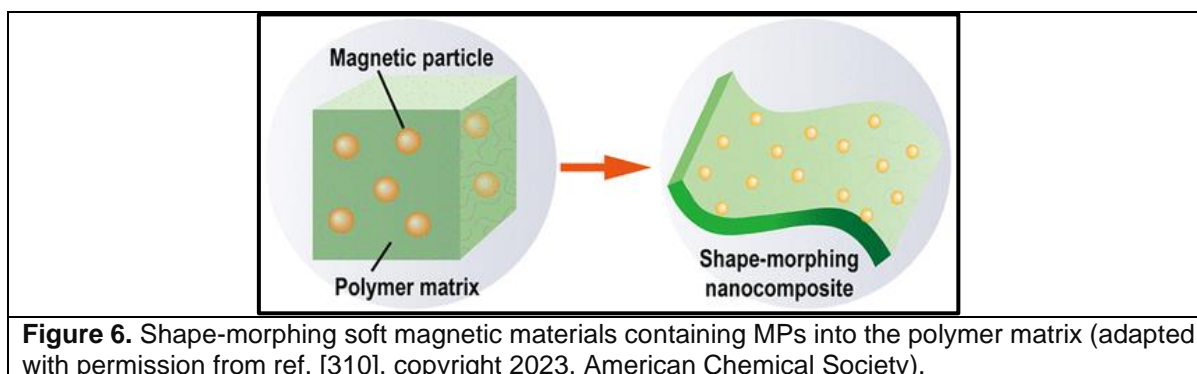


Figure 5. Recently 4D printing technology was used for various advanced sensors and robotics applications. The figure is drawn based on the various figures collected from (1) Smart grippers by Keneth et al. [293] (Copyright 2023 Elsevier B.V.) (2) Intelligent devices by Lie et al. [294] (Copyright 2022 American Chemical Society), (3) Flexible magnetolectric 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).

346 **3 Magneto-active soft Materials for 3D Printing**

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



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

378 **Mixing/dispersion of MPs:** MPs containing soft material can show isotropic or anisotropic
 379 characteristics depending on which fabrication technique is adapted. The fabrication of
 380 magneto-active soft composites containing MPs undergoes a curing procedure to stiffen the
 381 soft materials [325]. For instance, if the elastomers are cured in the presence of an external
 382 magnetic field, the magnetizable particles tend to form chain-like arrangements lending an
 383 overall directional anisotropy to the material such materials demonstrated that anisotropic
 384 magnetic soft material tend to have stronger coupling with the external magnetic field [326]. It
 385 is also crucial to remove gas bubbles as much as possible to prevent cavitation issues. Usually
 386 a maximum of 40 % (volume fraction) of MPs, the percolation threshold is achieved in soft
 387 polymers [327]. Moreover, along with MPs plasticizers are usually added to enhance
 388 mechanical interactions between the dispersed phase and the soft matrix. This is worth
 389 mentioning that if an external magnetic field is applied during the curing, the resulting material
 390 will be anisotropic because MPs migrate reaching the lowest energy state and therefore more
 391 likely to be used in engineering applications. However, if no external magnetic field is applied
 392 during the curing process, the resulting material is isotropic. Recently, Garcia-Gonzalez et al.
 393 [327] showed that the PDMS-based soft polymer and the platinum catalyst-based crosslinker
 394 were put together in such a way that the matrix chains increased their crosslinking degree.
 395 Insights of this study showed that a preferred direction of the CIP particles aligned with the
 396 field was achieved demonstrating more mechanically stiffer behavior of PDMS/CIP material
 397 along a magnetic field direction.

398 3.1 Magneto-active polymers

399 Magneto-active polymers (MAPs) usually contain MPs within the soft polymer matrix, which
400 triggers the application of magnetism [328]. These polymers are synthesized by uniform or
401 non-uniform distribution of MPs within the non-magnetic polymer matrix before the curing
402 [329]–[331]. Additionally, these particles can be aligned in a desirable direction upon the
403 application of a magnetic field during the solidification process. MAPs are also referred to as
404 magneto-sensitive polymers, magneto-active elastomers, magneto-sensitive elastomers, or
405 magneto-rheological elastomers. Based on the hysteresis loop of MPs and their coercivity,
406 MAPs are further classified into hard MAPs and soft MAPs [332]–[334].

407 The MPs of soft MAPs have a low magnetic coercivity and these particles do not adequately
408 reserve the magnetization under a null external magnetic field [335]. Some common examples
409 of these MPs include a Si-Fe alloy and Fe-Al series of alloys. In these polymers, MPs move
410 due to dipole-dipole interactions between particles in the presence of a magnetic field [336].
411 Such movements and rearrangements of MPs introduce some internal stresses that induce
412 deformations and change the mechanical properties. Soft MAPs can only help in achieving
413 simple and limited actuation for soft robotics applications [337]. On the other hand, the MPs
414 of hard MAPs featuring high coercivity like neodymium–iron–boron (NdFeB) can sustain
415 magnetism even after the removal of an external magnetic field. Consequently, upon applying
416 a further magnetic field, these particles tend to align themselves in the field direction,
417 introducing internal torques within these responsive polymers [338]. Therefore, hard MAPs
418 are preferred for soft robotics applications, as the relatively stable magnetism of these
419 polymers permits directly amendable magnetic fields to generate specific programmable
420 responses [339]–[341].

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

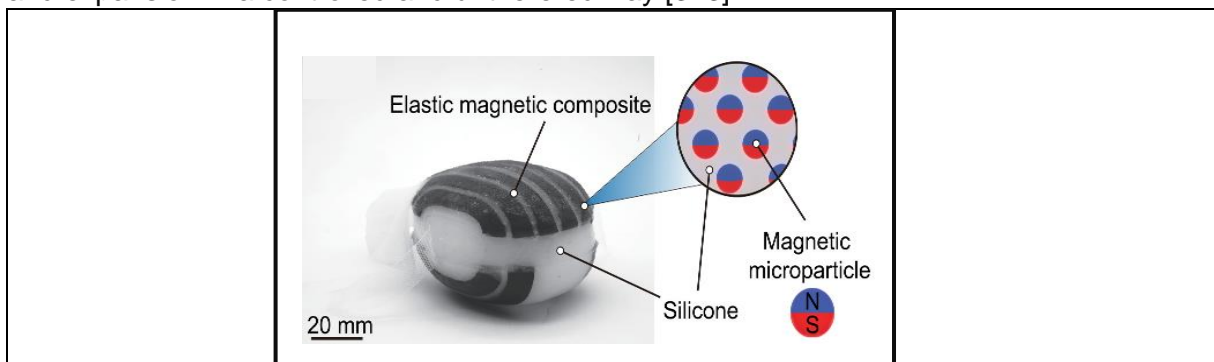


Figure 7. Magneto-responsive composites composed of MPs and pure silicone are used to develop soft bladder robots for assisting urination (adapted from ref. [344], under the terms of the Creative Commons Attribution license).

430 3D printing of magneto-active soft composites can be useful for producing soft structures with
431 good mechanical properties. Nowadays, magnetorheological elastomer (MREs) composites
432 which are filled with MNPs such as CIP, and Fe_3O_4 exhibit tunable rheological and viscoelastic
433 properties for meeting the demand of novel applications such as soft robotics, self-deployable

434 structures, actuating damping devices, vibration isolators, medical inserts, and flexible
435 electronics [345]–[347].

436 3.1.1 Shape morphing magneto-active composites

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

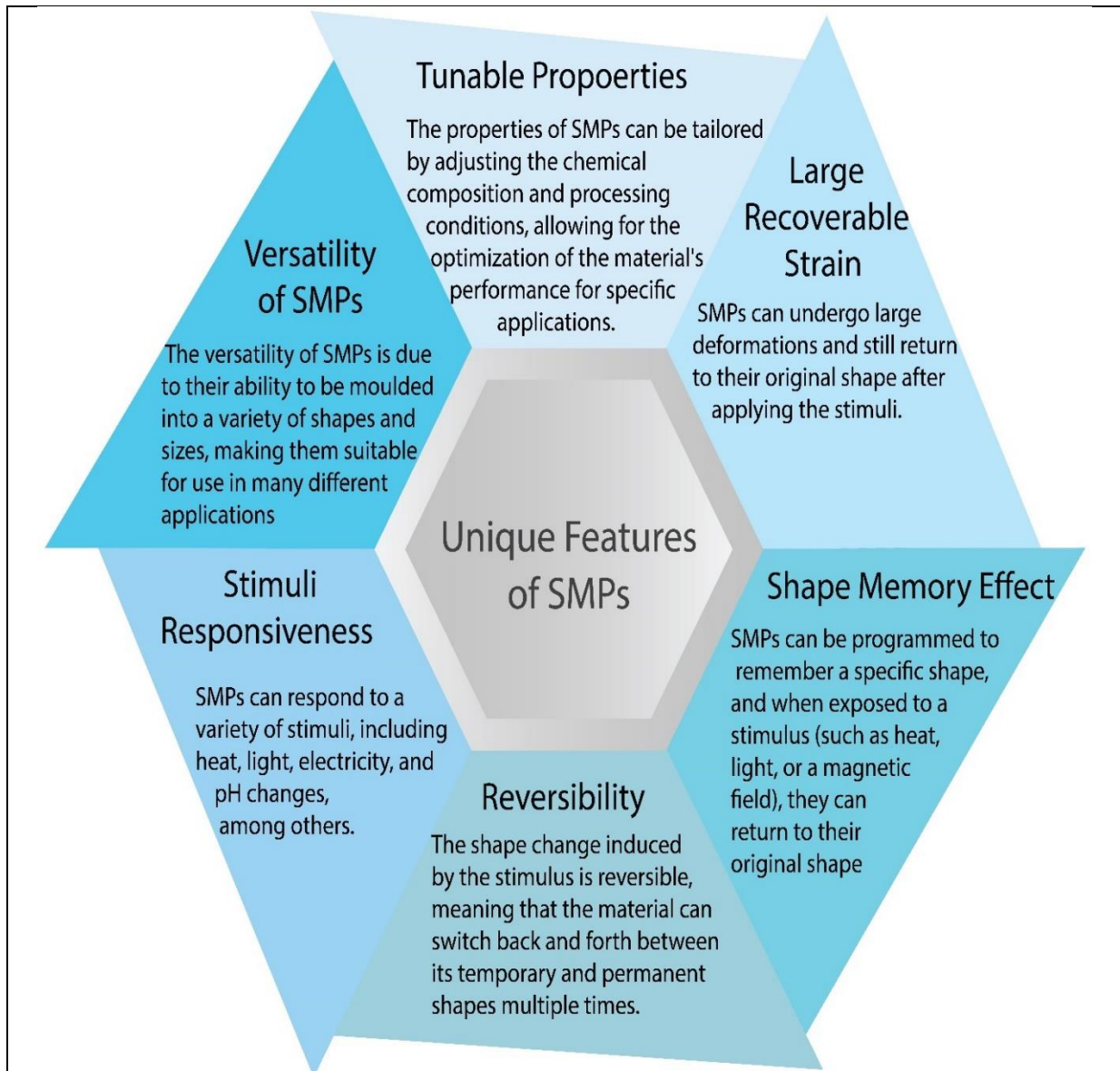
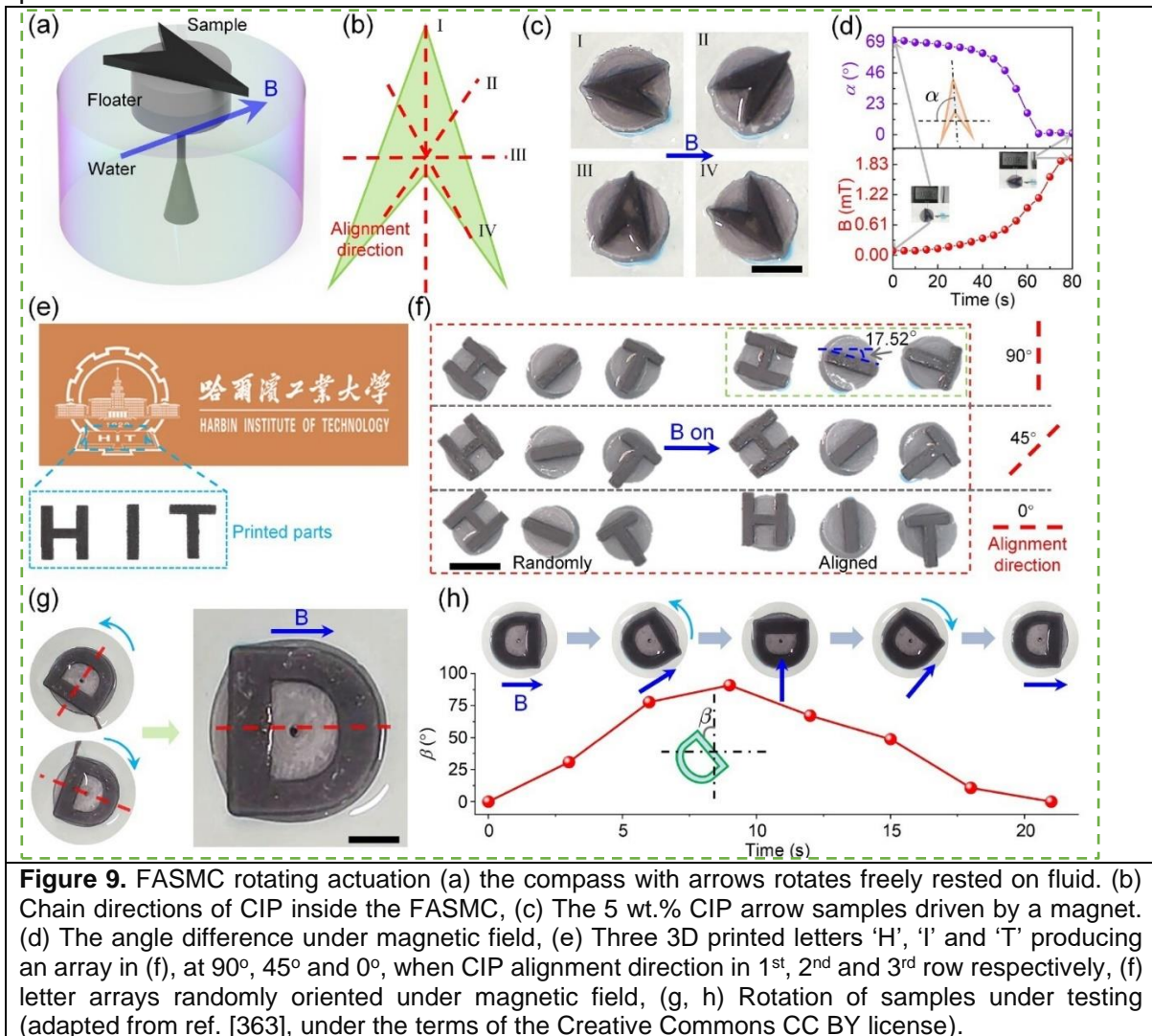


Figure 8. Prominent features of SMP enabling 3D printing of smart materials (Figure drawn with the help of ref. [353])

443 SMP-based composites are highly tunable for controlling many shape memory properties
444 [310]. For instance, the addition of various 2D materials such as graphene, CNTs, manganese
445 dioxide (MnO₂), iron oxide and silver nanowires etc, multifunctional features such as robust
446 self-adhesion, feasible 3D printability, rapid self-healing ability, and electrical conductivity of
447 composites can be improved for developing novel wearable devices [354]–[358]. Moreover,
448 various SMPCs such as citric acid-based SMPC, polyester urethane (PEU), acrylamide, N,N'-

449 dimethyl acrylamide (DMAA), ethylene glycol, dimethacrylate, and silicone: Ecoflex and silicon
 450 elastomer are commonly employed in combination with each other and some other materials
 451 as a potential SMPC [359], [360]. The interest in 3D printing of SMPC is steadily growing in
 452 many fields covering soft robotics biomedical devices, and flexible electronics [361]. Most of
 453 the SMPCs are based on the magnetic stimulus by embedding MNPs into the polymer
 454 matrices, usually ferrite and soft magnetic materials. The shape of SMPCs can be
 455 conveniently adjusted by applying an external magnetic field to achieve various characteristics
 456 including facile controllability, rapid response time, and reversible behavior for broad
 457 application prospects [362]. Recently, Wu et al. [363] prepared a flexible anisotropic soft-
 458 magnetic composite (FASMC) through DLP-based printing using flexible long-chain acrylic
 459 resin monomer and soft CIP-based MNPs. Insights of this study showed that multiple complex
 460 structures of FASMC with strong anisotropic magnetic properties exhibited large deformation,
 461 controlled motion, anti-deflection, variable stiffness metamaterial, and array assembly, as
 462 depicted in **Figure 9**. These behaviors of FASMC are particularly attractive when targeting
 463 next-generation sensors and actuators with superior magnetic properties in one or more
 464 specified directions.



465 Soft magnetic composites have been orderly deposited using an advanced 4D printing
 466 technique to build deformable actuators under low-strength magnetic field [364]. Reisinger et
 467 al. [365] introduced a novel technique for controlling the temperature of dynamic bond
 468 exchanged in covalently crosslinked polymer networks. Later, light-mediated curing was used

469 for printing various functional objects, as presented in **Figure 10(a₁)**, through DLP-based 3D
 470 printing, with spatially controlled reshaping capabilities. Furthermore, fiber-reinforced, and
 471 highly filled magneto-active thiol-ene polymer composites were effectively used for on-
 472 demand activation of dynamic transesterification with various reshaping capabilities (referring
 473 to **Figure 10(a₂)**), which gives rise to the potential use of 3D-printed magneto-active materials
 474 in various active and soft devices.

475 In another novel study, encoding of various shapes and forms by magneto-/electro-active
 476 SMPC structures was explored using carbon black-filled conductive PLA and iron-filled
 477 magnetic PLA through FDM [366]. The shape recovery technique was exploited under
 478 temperature and the magnetic field for a unique composite actuator was investigated. Results
 479 proved that the 4D-printed composite actuator achieved a maximum bending angle of 59°
 480 under a low external magnetic field and was fast enough to revert to its original shape when
 481 powered by a power supply, as presented in **Figure 10(b₁)-Figure 10(b₄)**. This research
 482 proved that the 4D-printed composite actuator strategy has broad application prospects in the
 483 field of soft robotics by keeping in line with sustainability rules.

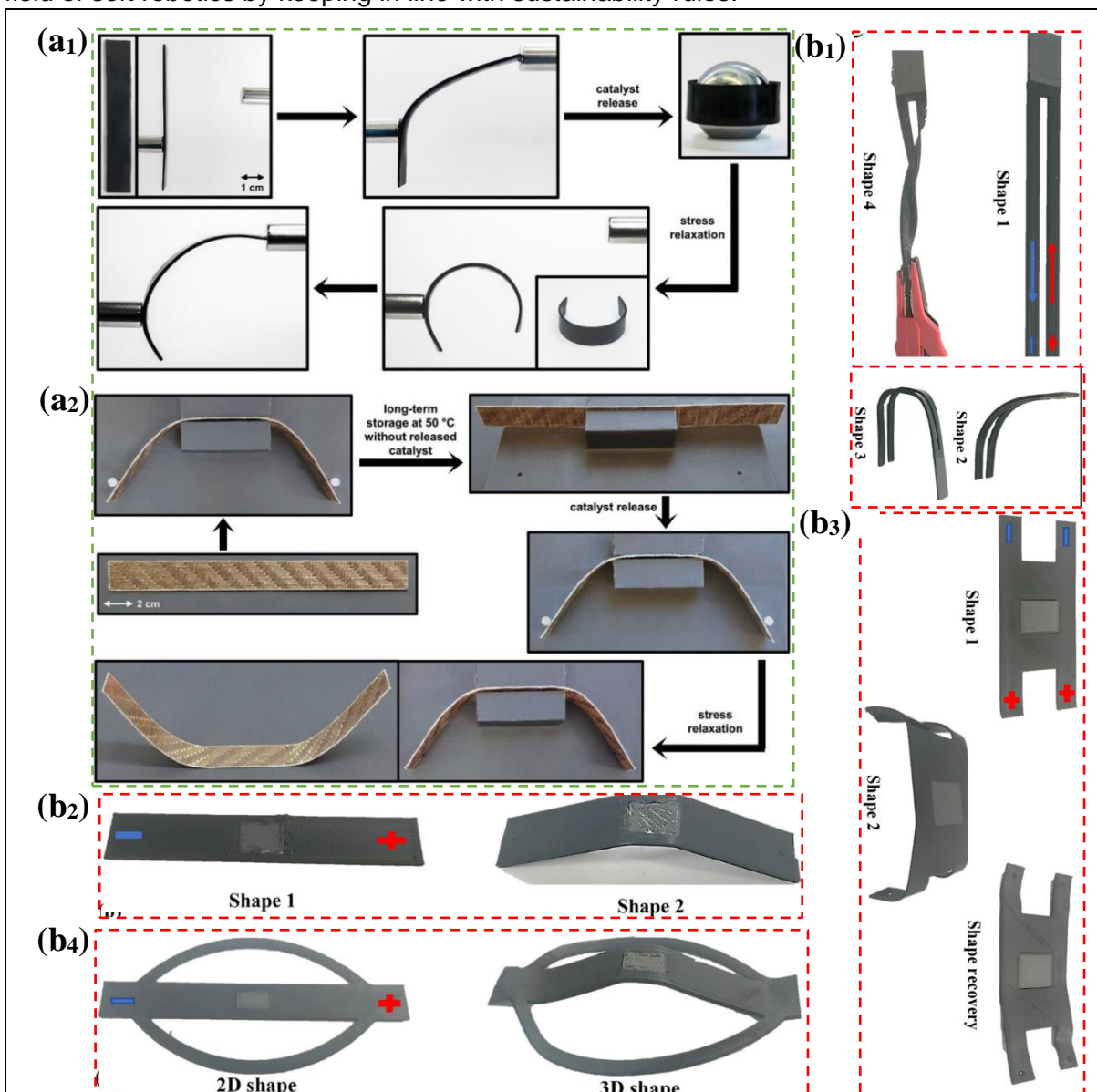


Figure 10. (a₁-a₂) Permanent reshaping of composite structures, (a₁) Magnetically assisted reshaping of a Fe₃O₄ particulate composite, (a₂) Reshaping of a fiber-reinforced composite (adapted with permission from ref. [365], copyright 2023, Wiley-VCH GmbH); (b₁) Different shapes of a 2D U-

shape materials, (b₂) Transformation of 1D beam shape to 2D shape under 60 V power supply and the permanent magnet, (b₃) Conversion of a 2D rectangular shape into a 3D structure (93% shape recovery), (b₄) Programming a 2D pyramid into a 3D structure (adapted from ref. [366], under a Creative Commons Attribution 4.0).

484 **3.2 Magneto-active multifunctional composites**

485 The world is continuously exploring novel smart materials with more versatile functionalities
486 [367]. As a result, it is a promising initiative to integrate the advantages of multi-active
487 ingredients into a single material or structure, through monolithic [368] or layered forms [369].
488 Compared to conventional MAPs, magneto-active multifunctional composites can developed
489 by integrating the advantages of LCEs and MREs [370]–[372]. For instance, LCEs exhibit high
490 work density and large strains (up to 400%) to multiple environmental stimuli like heat, light,
491 and electric field [373], [374]. Valiant efforts were made by researchers to combine the distinct
492 features of LCEs and MREs for developing soft materials with enhanced and unparalleled
493 functionalities [375]–[379]. For instance, Zhang et al. [377] developed an untethered miniature
494 12-legged robot, via a facile fabrication process (casting and soft lithography) by integrating
495 three distinct configurations of LCEs and MREs, as illustrated in **Figure 11(a)**. The results
496 revealed that this robot responded to wireless stimuli of a controlled magnetic field and
497 surrounding temperature. Thus, complex shape morphing behaviors with anisotropic material
498 properties can be achieved by using the multi-responsiveness of these soft composites.
499 Similarly, Zhang et al. [378] developed a multi-responsive actuator with accurately controlled
500 deformation through the integration of MREs and PDA-coated LCEs. This facile material-
501 structural synergetic design triggered complex and multimode programmable deformation
502 including shrinkage/bending, bidirectional bending, twisting/bending, and rolling/bending.
503 Additionally, this shape-morphing behavior could also be manipulated locally and sequentially,
504 thanks to its photo-sensitive feature.

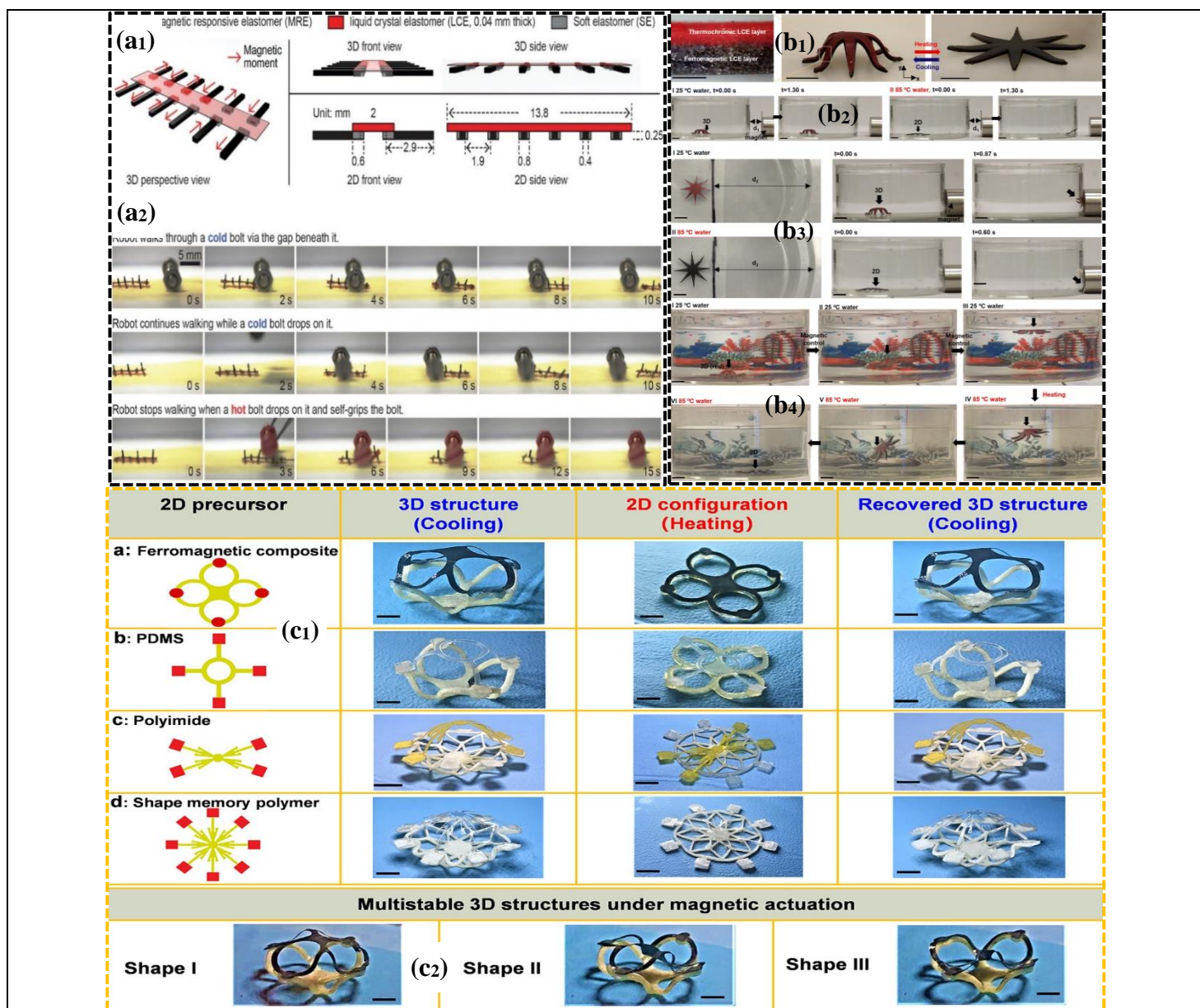


Figure 11. (a₁) Schematics demonstrating the design of a miniature 12-legged robot; (a₂) Robot movement and self-gripping of the hot bolt (adapted from [377], under the terms of the Creative Commons CC BY license); (b₁) Bilayer structure consists of ferromagnetic and thermochromic layers; (b₂) Magnetic actuation of octopus structure at different water temperatures; (b₃) Adaptive motion of octopus structure, when water temperature changed from 25°C to 85°C, under the same magnetic stimulus; (b₄) 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); (c₁) Diverse assembled 3D mesostructures and their configurations under heat stimulus; (c₂) Multistable 3D mesostructure under magnetic stimulation (adapted with permission from ref. [381], copyright 2021, American Chemical Society)

505 These soft composites can also be used to develop multifunctional structures with
 506 synchronous color-changing and shape-morphing properties such as biomimetic camouflage
 507 devices. For instance, Li et al. [379] reported a versatile and facile strategy to develop
 508 reconfigurable thermochromic biomimetic structures, such as chameleon and butterfly, as
 509 illustrated in **Figure 11(b)**. The single biomimetic structure contained a combination of LCEs,
 510 and MREs embedded with multiple color-changing dyes, which enabled the thermo-magnetic
 511 dual response of an octopus structure along with a camouflage feature. This response helped
 512 it to achieve adaptive and diverse biomimetic motions (rotating, rolling, swimming, and
 513 crawling), accompanied by a color camouflage. Thus, multifunctional magneto-active soft

514 composites are highly suitable to fabricate bilayer multi-stimuli actuators capable of complex
515 and accurately controlled deformations, and these actuators can be used in versatile fields
516 including biomedical, camouflage, and soft robotics.

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

526 **3.3 Magneto-active hydrogels**

527 The development of magneto-active hydrogels (MAHs) is considered a panacea for
528 developing more complex parts with excellent biodegradability and crack-healing properties
529 [383]–[385]. Recently, 3D-printed hydrogels have gained significant attention due to their
530 simple, accurate, and repeatable manufacturing. In this regard, polydopamine (PDA) hydrogel,
531 poly(3,4 ethylenedioxythiophene):polystyrene sulfonate (PEDOT:PSS) and polyacrylamide
532 (PAAM) are widely used for achieving toughness, and biocompatibility and validating the 3D
533 printability of such a hydrogel into customized architectures [386]–[388]. Moreover, hydrogel
534 products with excellent multiscale architectures and improved binding affinity at the interface
535 of other polymer chains [389]. Mostly two networks of hydrogels and polymers termed static,
536 and dynamic are extensively used to develop smart structures. Static dealing structural
537 integrity of materials or dynamic coping mostly with self-recovery and self-healing properties
538 [390].

539 Different natural and synthetic polymers or their combinations are used to develop hydrogel
540 chains through different cross linking ways [391]–[393]. MAH was first proposed in 1996 and
541 has been extensively researched ever since. Magnetic hydrogels with unique and distant
542 magnetic manipulation are captivating, particularly for hydrogel-based flexible and soft
543 actuators [394]–[396]. These hydrogels contain hydrogel chains embedded with nano-/micro-
544 scaled ferromagnetic or paramagnetic fillers that permit rapid actuation in response to an
545 external magnetic field. These hydrogels easily entrap MPs and exhibit excellent stability and
546 processability [397]–[400]. Magnetic response appears in MAHs due to the addition of MPs
547 [401]. These hydrogels have distinct advantages such as wireless actuation, facile operation,
548 complete biosafety and biodegradability, self-adaptability, intelligence, highly controllable
549 magnetic responsiveness, fully reversible response, and compatibility with miniaturization and
550 integration [267], [402]–[404]. Thus, 3D printing of MAHs has an enormous prospect in remote-
551 controlled and untethered soft actuators, bionics, soft robotics, flexible electronics,
552 hyperthermia cancer therapy, deployable micro-devices, and minimally invasive surgery
553 [405]–[409].

554 **4 Applications**

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

564 **4.1 Soft and intelligent robots**

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

571 Intelligent magnetic soft robots can change their structure in programmable and
572 multifunctional modalities depending on material architectures and methods for controlling
573 magnetization profiles [426]. Particularly, pneumatic soft actuators [427], and pneumatic
574 origami actuators were explored due to their unique attributes for producing a large
575 deformation of patterns with highly energy-efficient devices and safe tissue interaction [428].
576 However, there is a price to pay for the universal soft gripper, as its vulnerability limits its
577 lifespan (50,000 grips), particularly when sharp objects are present (5000 grips) [429].
578 However, soft magnetic actuators offer versatile locomotion modes including walking, crawling
579 swimming, rolling and jumping motions have shown great potential for emerging applications
580 [187], [430], [431].

581 Soft robotics are usually constructed of inherently flexible materials which improve their ability
582 to adapt to complicated situations and cooperate interactions with humans and soft actuators
583 [432]–[434]. **Figure 12** shows the key features and their dynamic behavior of soft robotics
584 under a stimulant environment. Traditionally, MPs are incorporated in soft robotics for
585 introducing anisotropy in two ways First, after the fabrication of the soft robot and second while
586 fabricating the soft robot. However, the starting material such as the magnetic composite of a
587 soft resin and MPs remains the same for both methods.

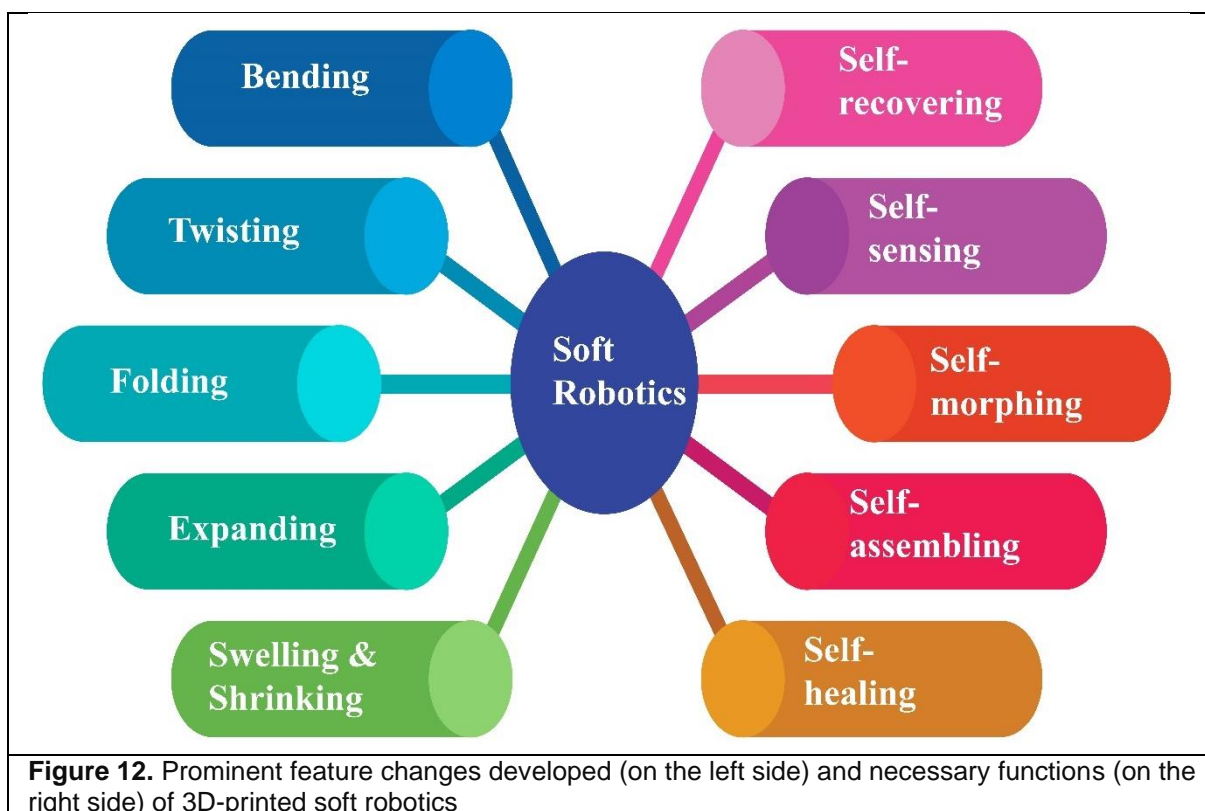


Figure 12. Prominent feature changes developed (on the left side) and necessary functions (on the right side) of 3D-printed soft robotics

588 Recently, a pneumatic origami structure using liquid silicone rubber was printed through an
589 industrial 3D printer. The proposed industrial printer directly printed the 3D folded structure

590 (origami-inspired structure) to maximize the design freedom for grasping various objects [435].
591 Urs et al. [436] studied unique two quasi-direct-drive actuators weighing 8–15 kg robots made
592 from 3D-printed components for an overall cost of less than USD 200 each. These thermal
593 actuated actuators were subjected to 420k strides of gait data which nearly doubles the
594 thermally driven torque and is useful in high-speed legged robots while matching the
595 performance of traditional metallic actuators. These 3D-printed designs are regarded as highly
596 customizable and reproducible soft actuators [437], for potential applications in robot legs.
597 Recently, Wan et al. [438] studied three kinds of pneumatic soft actuators for fabricating an
598 out-pipe crawling soft robot. Results revealed that the pipe robot realized omnidirectional
599 turning and could adapt to diverse shapes and sizes of pipes with a movement speed of 2.85
600 mm/s. Moreover, the small in size, low in mass and has a higher degree of freedom the soft
601 robotic arm achieved omnidirectional bending and a specific range of grasping work, for
602 potential applications in underwater pipe soft robots. Li et al. [439] studied multilayer DLP-
603 based printing for patterning MNPs including micro-structure through 2PP using gelatin
604 methacryloyl (GelMA)-based hydrogel with neodymium-iron-boron (NdFeB) or iron particles in
605 the ultraviolet (UV)-curable PDMS-based polymer matrix. Results showed that magnetic
606 torque actuation produced various shape changes such as gripping, swimming, rolling, and
607 walking, as depicted in **Figure 13(a₁)-Figure 13(a₃)** are induced by programming
608 heterogeneous magnetization within discrete multilayer robot segments. Moreover, the
609 opening angle of a capsule-like robot under magnetic actuation, as depicted in **Figure 13(a₄)-**
610 **Figure 13(a₅)** was useful for drug delivery. Thus, the proposed facile approach is feasible for
611 the creation of versatile 3D multi-material actuators for broader applications.

612 MASMs are reconsidered as fast, untethered, and reversible shape reconfiguration attractive
613 for novel soft robotics [440]. For instance, Qi et al. [441] investigated a heat-assisted magnetic
614 reprogramming approach for developing 3D-printed magneto-active soft matter using CIP as
615 a soft-magnetic reinforcing filler with the elastic matrix silicone rubber. The magnetic
616 reprogramming approach relied on heating PCL-based thermoplastic matrix above its melting
617 point and applying magnetic fields during cooling for reorienting soft MP chains for achieving
618 multiple deformation modes with unique shape-morphing features, as presented in **Figure**
619 **13(b₁)-Figure 13(b₂)**. Moreover, the proposed approach was successfully employed for
620 multiscale and reprogrammable soft machines such as adaptive grasping of a soft gripper with
621 the tunable actuation response, as presented in **Figure 13(b₃)**. Lastly, the unique sensing
622 performance of triboelectric skin (due to the use of CNT as a conductive filler) was also
623 demonstrated by using electrical signals to identify the deformation and contact behaviors.
624 Thus, the magnetic reprogramming approach provides a new concept for designing new active
625 materials for broader applications in soft robotics.

626 In another novel study by Simińska-Stanny et al. [442] soft actuators were fabricated using
627 printable magnetic hydrogel ink through multi material DIW. Results showed that magnetic
628 hydrogels had good mechanical stability, unique magnetic responsiveness, highly porous as
629 well as noncytotoxic towards fibroblasts. Moreover, 3D-printed magnetic actuators
630 demonstrated excellent actuation behavior, as depicted in **Figure 13(c₁)- Figure 13(c₂)** by
631 magnetically induced jumping rolling and bending. The proposed 4D printing of magnetically
632 responsive hydrogel strategy would provide an efficient way to fully capitalize on the role of
633 biocompatible materials for developing a wide range of soft actuators.

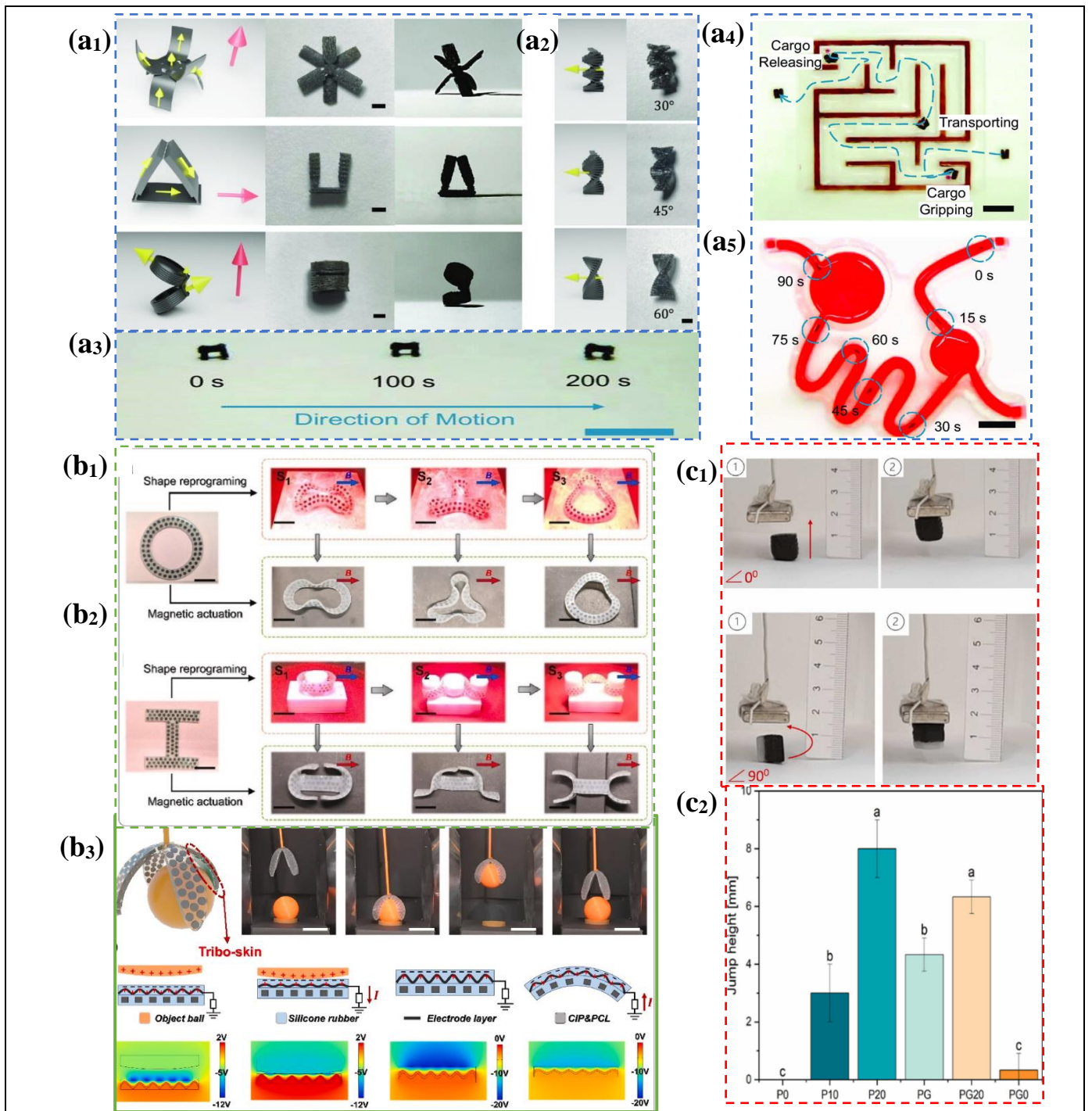


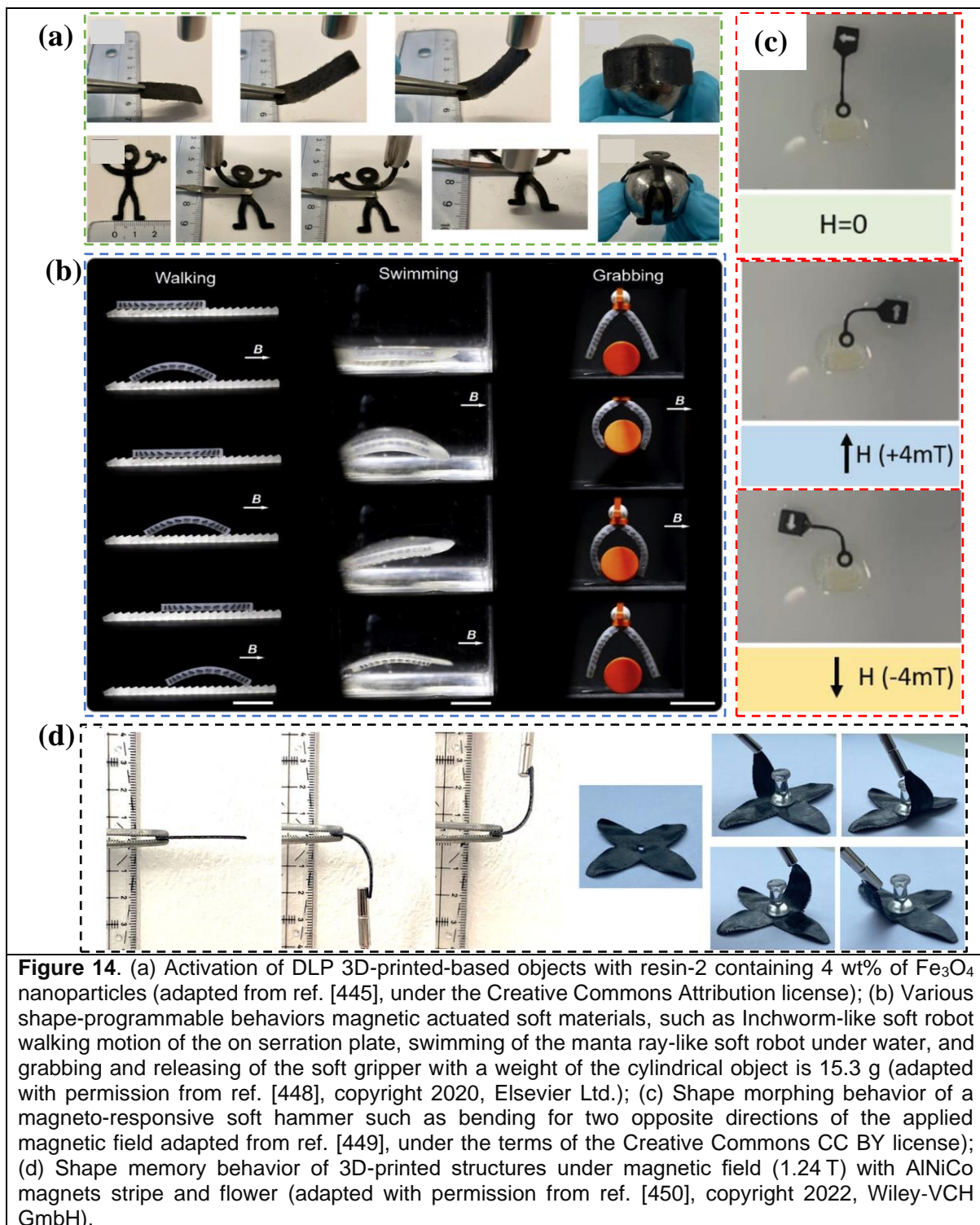
Figure 13. (a₁) Images of the 3D-printed robots under magnetic actuation (actuation field highlighted with red arrow) with encoded magnetization profiles (magnetization direction highlighted with the yellow arrow at each segment), (a₂) Helical robots with various helix angles, (a₃) Motion of the robot with the oscillatory frequency of 2Hz under actuation field, (a₄) Navigation of capsule-like robot in a maze map for cargo manipulation including gripping, transporting, and releasing, (a₅) Navigation of helical robot in a vascular model (adapted from ref. [439], under the terms of the Creative Commons Attribution License,); (b₁-b₂) Reprogramming and magnetically actuated shape morphing behavior of 3D printed various characters (b₁) "O", (b₂) "H" under magnetic field 400 and 300 mT, respectively, (b₃) Snatching function of four leaves-based soft gripper under 300 mT (adapted with permission from ref. [441], copyright 2022, Elsevier Ltd.); (c₁) Various jumping behavior of magnetic hydrogel under a magnetic field, (c₂) Difference of jumping heights for various 3D-printed cubes (adapted from ref. [442], under the Creative Commons CC-BY-NC-ND license).

634 Soft robotics always suffer permanent damage from irregular external stimuli and repetitive
 635 motions during their long service life [443], thus the self-healing ability of smart material is

636 highly desirable for overcoming these issues [444]. Cazin et al. [445] explored the magnetic
637 response with thermo-activated healability using Fe_3O_4 nanoparticles in a dynamic
638 photopolymer network (thiol-acrylate resins containing magneto-active fillers) through DLP-
639 based 3D printing. Results demonstrated that the healing performance of 3D-printed
640 structures was observed due to the recovery of magnetic and mechanical properties under
641 temperature-triggered mending. As a proof of concept, the 3D-printed magneto-responsive
642 structures were thermally healed, reshaped, and activated under magnetic field stimulus, as
643 presented in **Figure 14(a)**.

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

653 Lantean et al. [449] investigated complex macroscopic gear-based devices through DLP using
654 MAPs containing Fe_3O_4 . Insights of this study revealed magneto-responsive hammer-shape
655 actuators, as presented in **Figure 14(c)** with different stiffnesses demonstrating various
656 motions including rotation and bending. Thus, magneto-responsive gears made from MASMs
657 have advantages in broader applications including linear actuators, gear-trains, and micro
658 grippers. Rossegger et al. [450] explored magnetic-driven actuators through DLP-based using
659 magneto-responsive thiol-click photopolymers containing Fe_3O_4 . The thiol crosslinker further
660 imparts softness and flexibility to magnetic actuators. Moreover, as proof of concept, various
661 3D prints such as strips and flowers, as depicted in **Figure 14(d)** showed magnetically driven
662 movement for their promising role in soft robotics and other fields.



663 3D-printed magnetic actuated soft robotics offers an unprecedented geometric configuration
 664 with more degree of freedom due to the programmable magnetization profile [377], [451]. For
 665 instance, Bayaniahangar et al. [452] fabricated 3D-printed soft magnetic helical coil actuators
 666 using PDMS embedded with iron oxide particles. The developed complex helical coil
 667 structures were supported with Pluronic f-127 hydrogel and had 30 % iron oxide particles. This
 668 allowed linear magnetic actuation with 360 % device's linear actuation and 80° bending
 669 actuator in helical coils. Insights of this study also revealed that the 3D-printed helical coils
 670 under magnetic field stimulus demonstrated untethered soft robot locomotion as presented in
 671 **Figure 15(a₁)- Figure 15(a₂)** on 45- and 90-degree inclines. Pavone et al. [453] printed

672 support-free actuators to exploit the Lorentz Force: permanent magnets and Gallium for
 673 effective movement of the actuator. The insights of this study revealed that 3D-printed actuator
 674 has a wide range in numerous fields such as limb prosthesis wearable devices, and human
 675 motion. Moreover, at a maximum current of 6.10 various actuator movement (displacement of
 676 20 mm and acceleration of 1.10 m/s^2) was observed as presented in **Figure 15(b)**.
 677 Soft actuators are made of flexible or compliant materials and give large deformation and high
 678 stability for many applications [454], [455]. Recently, Cao et al. [456] developed ultra-flexible
 679 magnetic actuators through a facile FDM-based 3D printing technique using thermoplastic
 680 rubber (TPR) pellets/CIPs. Also, the 3D-printed magnetic actuator exhibited highly
 681 functionalized manipulations and controllable deformation of the sucker and pump actuator for
 682 sticking objects and pumping liquid as presented in **Figure 15(c)**. Thus, multifunctional, and
 683 ultra-flexible magnetic actuator offers a promising strategy for fabricating highly complex and
 684 controlled deformable structures for soft robotics applications.

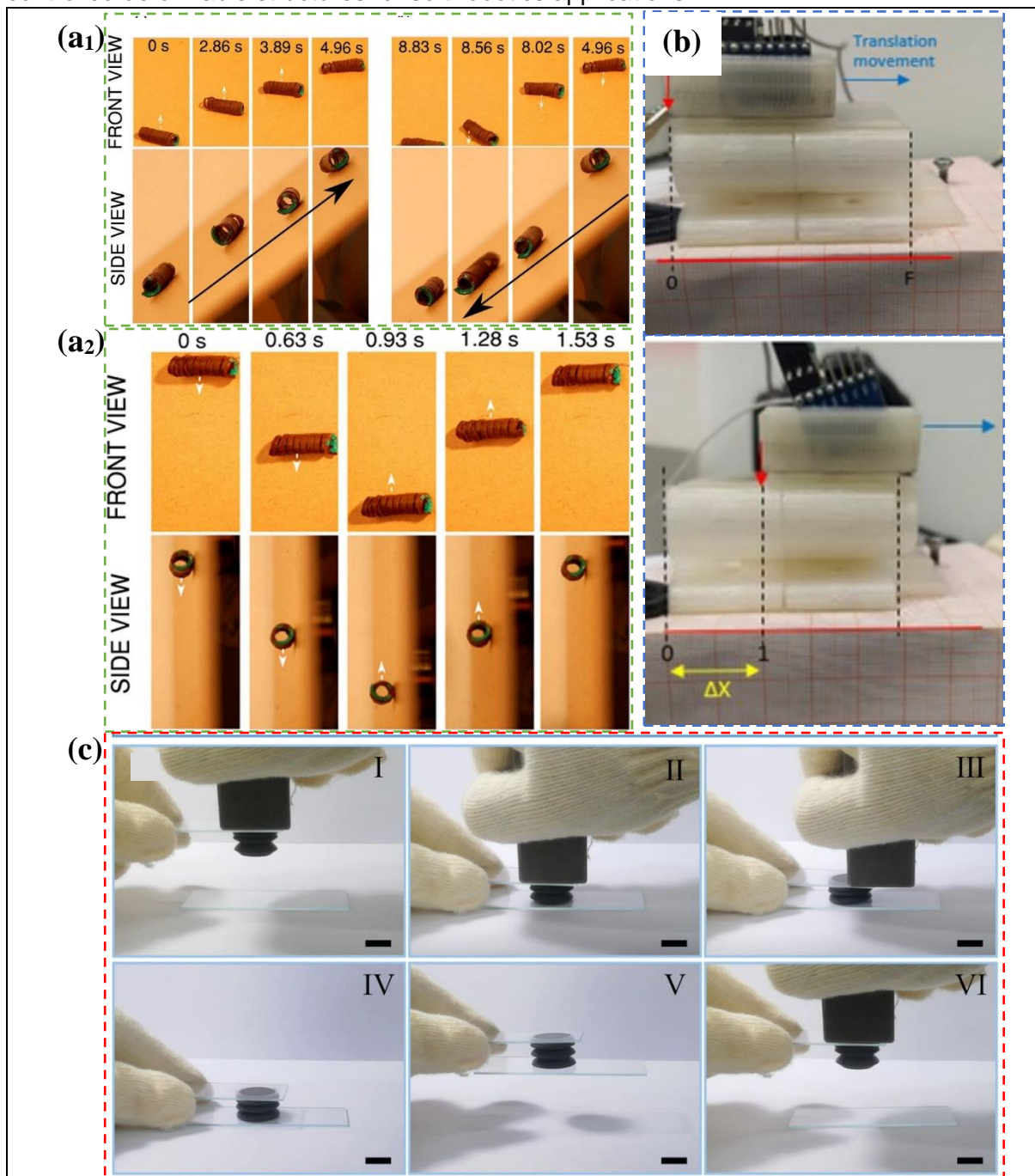


Figure 15. (a₁-a₂) Images of untethered locomotion of helical coil on a 45° incline (a₁) upward, downward, and (a₂) on the 90° vertical wall, front view and side view (adapted with permission from ref. [452], copyright 2020, Elsevier B.V.); (b) Translation movement of the actuator with and without current 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).

685 4.2 Untethered microrobots

686 Microrobots are robots whose dimension reaches in micron-sized realm for performing
687 necessary tasks at a micron scale including sensing, object manipulation, and improved
688 navigation under external stimuli or environmental sources [186], [457]–[459]. The science of
689 robotics is accelerating towards the conception of microrobots with new functionalities,
690 especially under magnetic properties to control the motion of microrobots [460]. In this regard,
691 3D printing techniques are captivating for making perfect microrobots ensuring their
692 satisfactory performance. Among them, 2PP is regarded as the best technology for producing
693 microrobots due to its highest resolution at the nanometric scale, and the creation of
694 monolithically 3D complex structures using diverse materials including inorganic and organic,
695 passive, and active [461], [462]. Untethered microrobots due to their small size and mobility
696 have enormous prospects for localized diagnosis, in minimally invasive surgery, targeted
697 delivery of agents, tracking, imaging, and sensing, micromanipulation, cell delivery, and
698 biopsies [463]. Among them, magnetic actuation exerts magnetic force and torque on
699 magnetic materials in microrobots to actuate and control them, which has the advantages of
700 fuel-free, simple direction, speed control, and harmless penetration through living tissues
701 [464]. Microrobots are now considered the pioneer in the development of advanced healthcare
702 systems in personalized medicine [465]. For instance, Jang and Park [466] developed an
703 untethered milli-gripper fabricated from 3D-printed biodegradable chitosan hydrogel ink
704 coated with citric acid superparamagnetic iron oxide nanoparticles (SPIONs). Results showed
705 that a 3D-printed gripper was promising for gripping and releasing cargo under an applied
706 electromagnetic field, as presented in **Figure 16(a₁)- Figure 16(a₂)**. Moreover, the untethered
707 milli-gripper demonstrated a precise position control due to the high magnetization of the citric
708 acid-coated SPIONs. Thus, the proposed work proved that the biomimetic untethered milli-
709 gripper also be employed as a minimally invasive small soft robot in vivo for numerous
710 biomedical applications including targeted drug delivery. Pétrot et al. [467] fabricated remotely
711 actuatable NdFeB-based MNPs. Reported results demonstrated that magnetically deformable
712 3D culture substrate actuated under a magnetic field and bends back and forth along its
713 longest axis, as presented in **Figure 16(b)**. Also, these structures had soft, curved, and
714 dynamic properties of tissues in vivo for potential applications in micro-actuator field.
715 Soft robotics driven from AM of naturally available materials have proved to be more effective
716 in achieving complex structures in a more deterministic manner [468]. For instance, Zhang et
717 al. [469] exploited wirelessly actuated programmable microfluidic cilia using naturally available
718 materials FePt Janus microparticles/silk fibroin (SF) hydrogels. Insights of this study showed
719 that high tunable actuation performance of proposed material for various arrangements
720 (antiplectic, symplectic, and diaplectic metachrony) and 2D arrangements (circular and
721 triangular) was achieved, as presented in **Figure 16(c)**, under less than 10 mT external
722 magnetic field. Such robust integration of the multi-material including FePt and SF rendered
723 cilia system allows researchers to use them for future applications in biomedical and health
724 care devices.

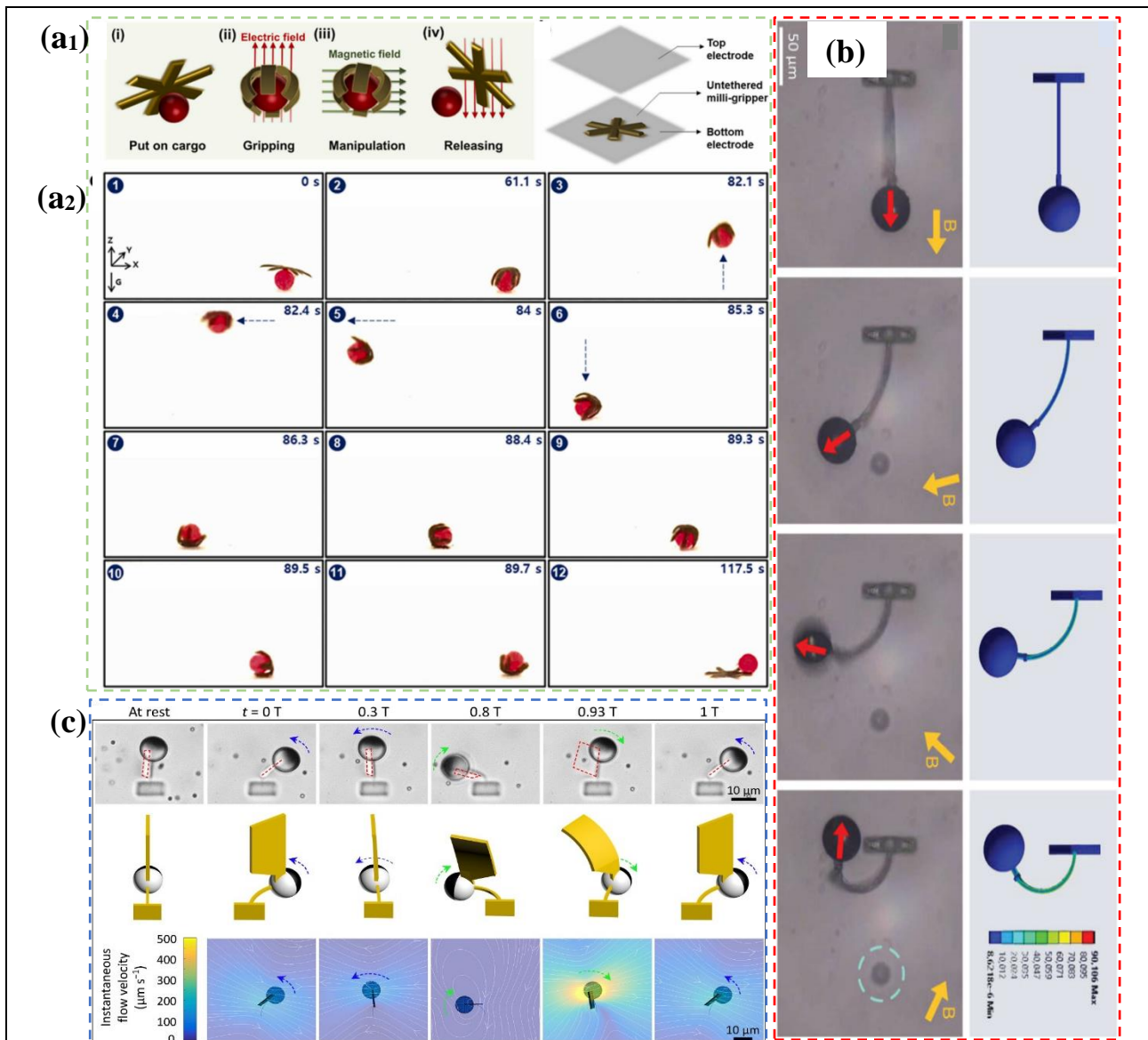


Figure 16. (a₁-a₂) Untethered milli-gripper used for cargo delivery test: (a₁) Schematic diagram illustrating the untethered milli-gripper on cargo stimulated by a magnetic field and releasing of the cargo induced by an electric field, and schematic diagram showing the electrode system used in the cargo delivery test, (a₂) 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)).

725 Miniature robots can be deployable on the water surface for achieving high controllability for
 726 various applications. Richter et al. [470] proposed novel microscale magnetic soft actuators.
 727 Insights of this study showed that ultrathin (80 μm) and lightweight (100 gm^{-2}) magneto-
 728 responsive actuators could lift, tilt, pull, or grasp near each other under electromagnetic near-
 729 field, as presented in **Figure 17(a)** at low energy consumption (0.5 W). It was envisioned that
 730 such soft micro magneto active robot would serve as a pioneer for next-generation soft robots
 731 in various prevailing applications in both biomedical and engineering sectors.
 732 Ansari et al. [471] printed anisotropic soft structures using magnetic ink containing a UV-
 733 curable resin and MNPs using an extrusion bioprinter. A custom electromagnetic coil system

734 was used during extrusion for orienting the magnetic moment of the particles in the ink. Results
 735 exhibited that with 1:1 particle-to-resin ratio in the magnetic ink under a 20 mT field for
 736 orientation for printed structure demonstrated a preferential magnetization index up to 0.99. It
 737 was shown experimentally that soft structures have tremendous promise in shape morphing
 738 capabilities for an object using a folding cube robot through loading, carrying, and dropping,
 739 as presented in Figure 17(b₁)- Figure 17(b₂). Lin et al. [472] studied a novel magnetic-driven
 740 folded diaphragm inspired by the locomotion of earthworms having various radial
 741 magnetization properties for controlling the contraction and stretching between body
 742 segments. Experimental results showed that the developed folded diaphragm exhibited
 743 distinctive features for producing different shapes including untethered soft robotic systems
 744 as soft drivers (actuators) for their practical applications such as soft biomimetic robots and
 745 diaphragm pumps under a magnetic field, as illustrated in Figure 17(c₁)- Figure 17(c₂). This
 746 approach unravels many opportunities to fabricate multifunctional robots including the
 747 swimming robot inspired by squid and bio-earthworm crawling robot.

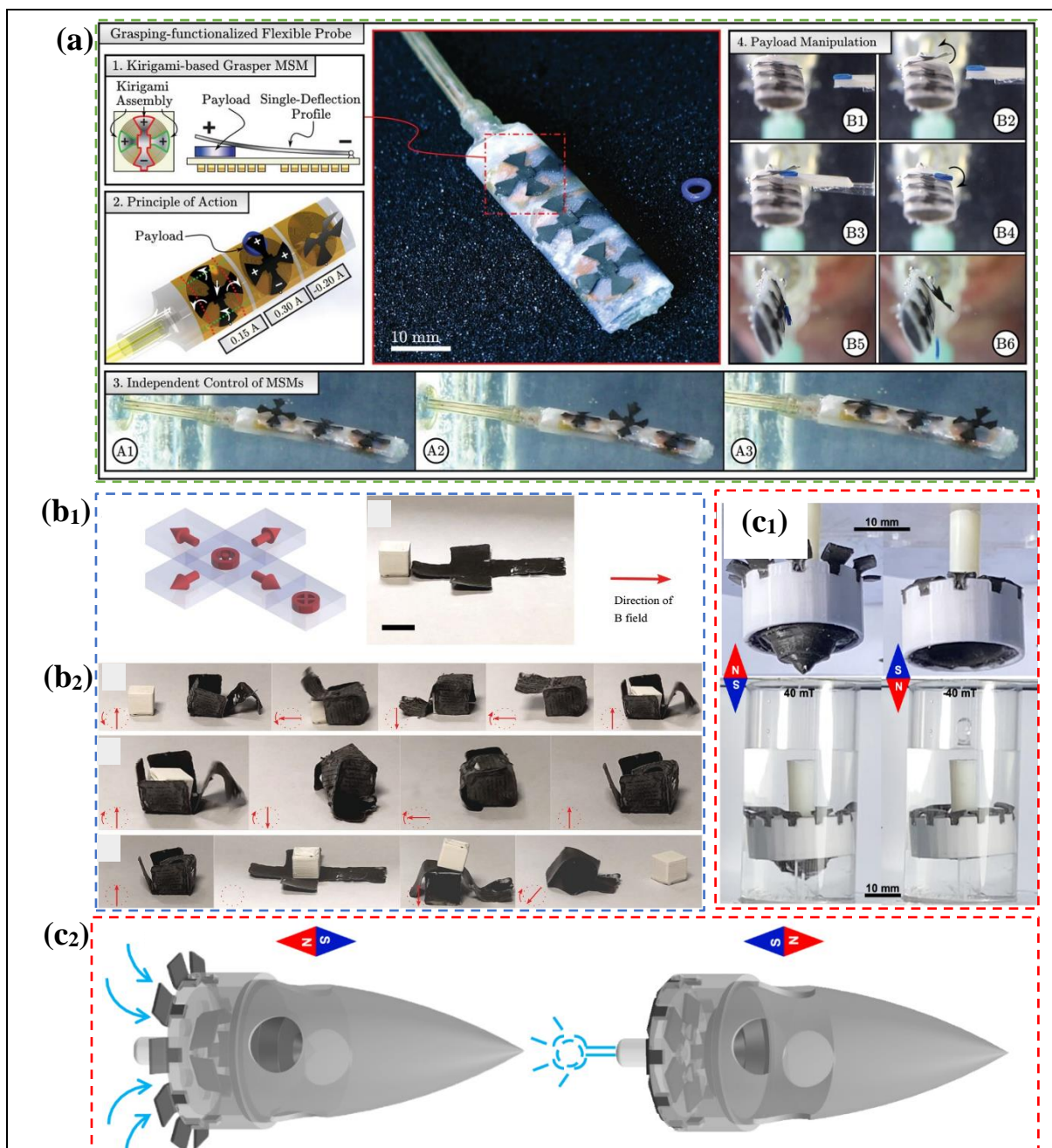
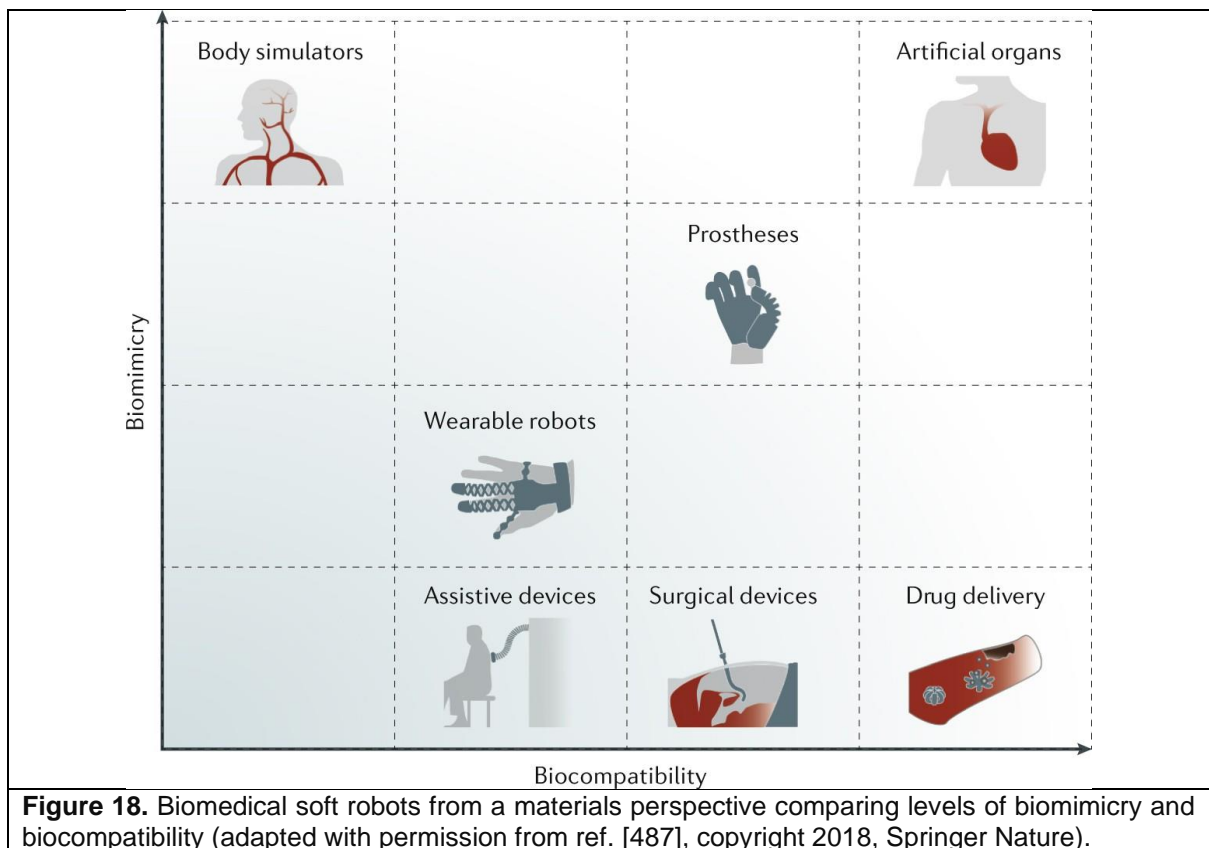


Figure 17. (a) A flexible tip functionalized for payload (20 mg) grasping and release using near-field magnetic soft machines (adapted from ref. [470], under the terms of the Creative Commons CC BY license); (b₁) Schematic diagrams of the printed sheet with six various magnetization directions on the six faces of the cube, and the printed sheet with said magnetizations placed alongside a cubic object of 0.5 g (b₂) 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); (c₁) 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), (c₂) Sucking and schematic diagram showing swimming robot driven by the harmonic magnetic field (adapted from ref. [472], under a Creative Commons Attribution 4.0 International License).

748 **4.3 Biomimetic devices**

749 Biomimetic is a type of human-made actuation material or device that can initiate motions
750 under force [473]. Different bioinspired designs of scale shapes and arrangements result in
751 various types of anisotropic friction, providing a means of switching the robot's locomotion for
752 desired conditions [474]. Moreover, due to the huge demand for recreating human skin with
753 the functions of the epidermis and dermis for interactions with the physical world [475], soft
754 actuators have attracted considerable interest in the biomimetic field for many biomedical
755 applications [476]. Magnetic robots actuated wirelessly and rapidly under an external magnetic
756 field for non-invasively access and navigation in difficult-to-reach areas inside the human
757 body. This is because of deformation 3D-printed smart structures which have unique
758 implemented actions such as gripping and lifting as well as self-healing ability [477]. Using this
759 facile strategy, other smart biomaterials could be designed which is in great demand and used
760 for a variety of applications, such as bionic grippers [478], open-channel microfluidic chip for
761 controllable liquid transport [479], tissue engineering [480], and drug delivery [481]. These soft
762 robots can be precisely actuated at target sites for intelligent cargo release under a magnetic
763 field [482] and applications related to neurological disorders such as motor and sensory
764 deficits [483]. Thanks to their intelligent responsiveness, researchers have rationally designed
765 magnetic actuated soft robots that can encapsulate therapeutic agents for biomedical
766 applications [484]. Now, 3D-printed biomimetic-based devices especially those made from
767 biodegradable materials have captivating adaptivity, complex designability and stimuli
768 responsiveness [485] and have brought significant advancements for various biomedical
769 applications [486], as highlighted in **Figure 18**.



770 Biomimetic devices are usually flexible, reconfigurable, compliant, and adaptable to switch
771 between various states (flexible to stiff) for demanding applications such as targeted drug
772 delivery [488]. For instance, Choi et al. [489] proposed the idea of a soft carrier using through
773 fabricating the lid, border, and hemisphere using a thermo-responsive poly(N-
774 isopropylacrylamide) (PNIPAM)/polyethylene glycol (PEG) hydrogel and SPIONs using 3D
775 printing. Results showed that the hemisphere allowed the successful storage and transport of
776 cargo (soft carrier) under dual stimuli such as near-infrared (NIR) light and magnetic field with
777 different shapes and numbers of cargo, as presented in **Figure 19(a₁)- Figure 19(a₂)**.
778 Cao et al. [490] studied biomimetic magnetic actuators through an FDM-based 3D printing
779 technique using TPR particles and CIP. Insights of the study showed that various shape
780 transformations of magnetic actuators such as the predation behavior of octopus tentacles,
781 the flower blooming behavior of the plant and the flying behavior of the butterfly, as presented
782 in **Figure 19(b₁)-Figure 19(b₂)**. It was anticipated that the 3D-printed MASMs could open new
783 avenues for the fabrication of a diverse range of soft robotics with multiple functions.

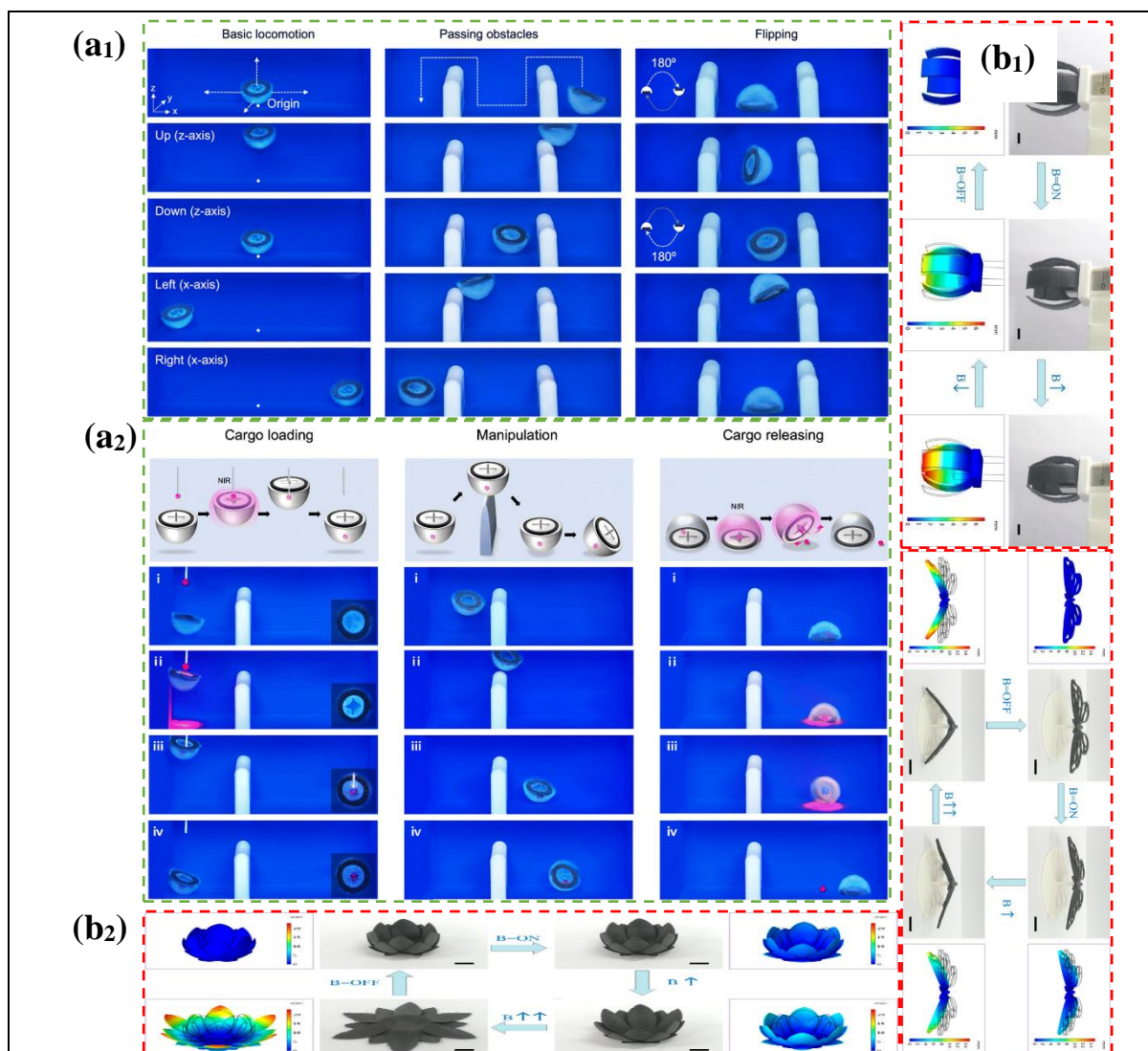
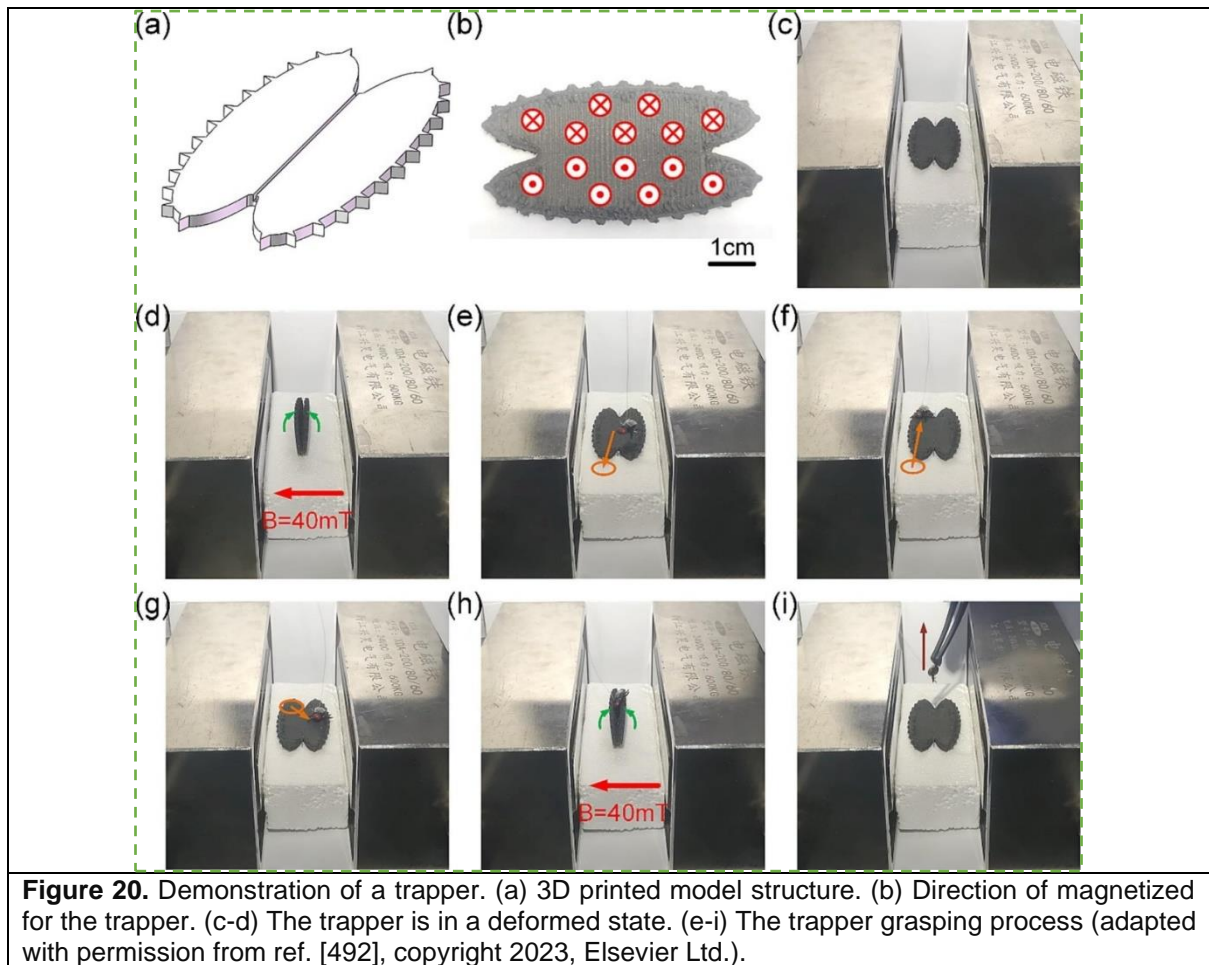


Figure 19. (a₁) Various images of basic locomotion, passing obstacles, flipping the smart soft carrier flips by 180° using a magnet without relying on an external wall such that the downward-facing lid faces upward, (a₂) Cargo delivery test of the smart soft carrier, cargo loading, schematic of manipulation, and cargo releasing (adapted with permission from ref. [489], copyright 2023, Royal Society of Chemistry); (b₁-b₂) Magnetic field-induced deformation and finite element simulation of various biomimetic magnetic actuator: (b₁) tentacle and butterfly, (b₂) flower (adapted with permission from ref. [490], copyright 2021, American Chemical Society).

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



793 Wang et al. [493] printed a millimeter-scale magnetic soft robot (referring to **Figure 21a-Figure**
 794 **21b**) using NdFeB/PDMS, multiwalled carbon nanotubes (MWCNTs)/PDMS and reduced
 795 graphene oxide (rGO)/PDMS integrated with temperature, tactile and electrochemical sensing
 796 functions. Furthermore, the shape morphing behavior (**Figure 21c**) of the robot showed
 797 remarkable sensing performance such as linearity of 3.383 k Ω /°C, and electrochemical stimuli
 798 with a low detection limit of 0.036 mM for NaOH solutions. Thus, the proposed study
 799 anticipated the performance of such a robust soft robot for next-generation targeted drug
 800 delivery, as presented in **Figure 21d- Figure 21e**.

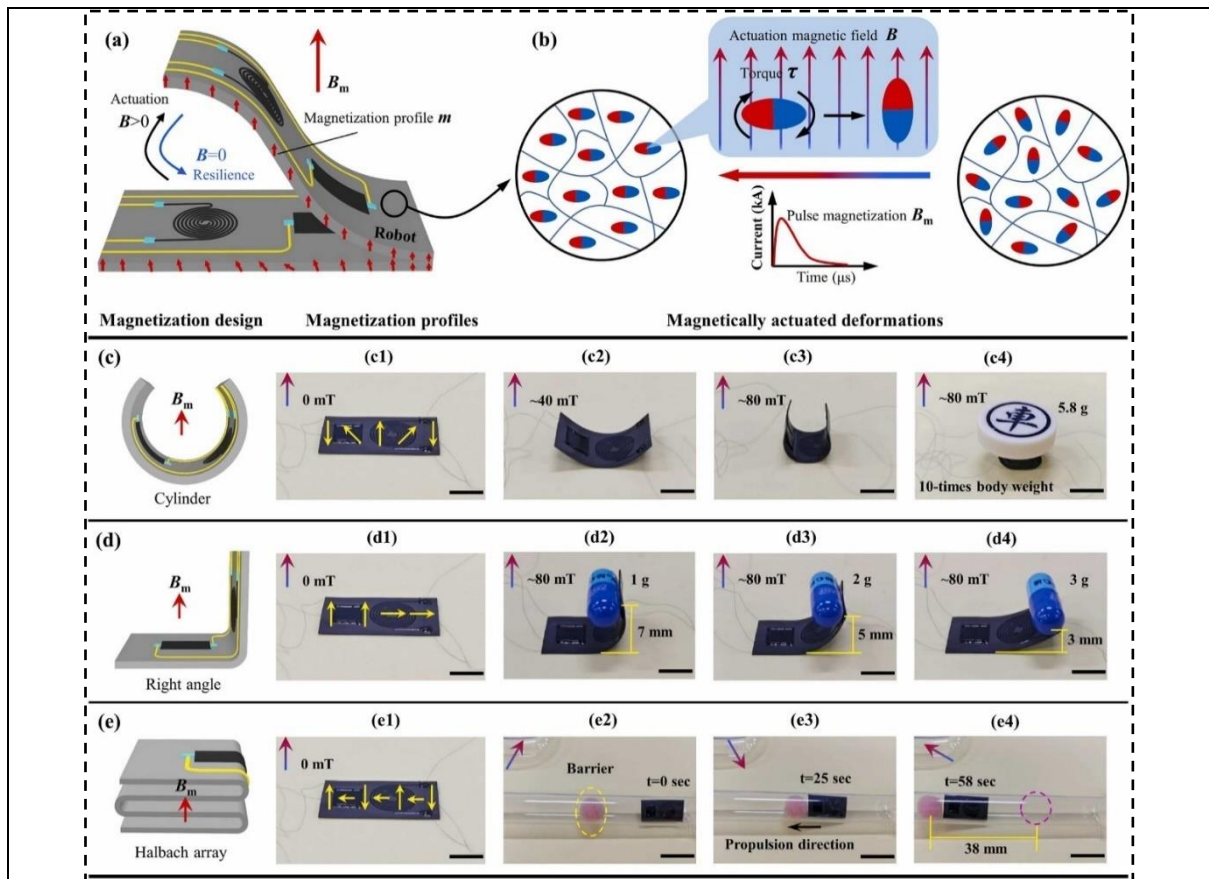


Figure 21. Magnetic soft robot multi-dimensional deformations and actuation mechanism. (a) The folded robot was magnetized under a magnetic field having a unidirectional pulse B_m and implanted into its body with the magnetic profile. (b) Illustration of magnetization and actuation mechanism. The robot 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.).

801 Intelligent tactile sensing is critical for soft robotics so that they can interact safely with
 802 unstructured environments and produce desired motions [494] under many shapes such as
 803 bionic flowers, and bionic worm robots [495]. Wang et al. [496] used a highly viscous magnetic
 804 composite ink for designing various bionic soft robots. Various actuator prototypes with various
 805 magnetization orientations and profiles have been fabricated such as bionic soft robots and
 806 magnetically powered electrical switches to successfully perform different operations including
 807 dragonflies and inchworms as presented in **Figure 22(a₁)- Figure 22(a₃)**. Thus, the proposed
 808 study confirmed that the magnetic responsive materials with programmable patterning fulfil
 809 the future of soft robotics in functional and practical applications.

810 Wang et al. [497] explored an insect-scale magnetoelastic robot using PDMS embedded with
 811 NdFeB-based MPs having improved controllability designed. The robot produced a
 812 controllable jumping motion by tuning magnetic and elastic strain energy. Results showed that
 813 on-demand actuation was applied for precisely controlling the pose and motion of the robot
 814 during the flight phase for effectively performing numerous tasks with integrated functional
 815 modules, as depicted in **Figure 22(b₁)- Figure 22(b₄)**.

816 Yao et al. [498] studied the diversification of actuation modes of magnetic-active actuators
 817 using blending matrix of PCL and thermoplastic polyurethane (TPU) and soft CIP-based MNPs
 818 as fillers through 3D printing. The results showed that 3D-printed magneto-active structures
 819 have excellent shape fixation, shape recovery rates, exceptional flexibility, and
 820 magnetorheological effects, as presented in **Figure 22(c)**. The shape-morphing behavior was

821 an excellent match with the simulation results and has an ideal role in numerous fields such
 822 as intelligent flexible robotics and biomedicine.

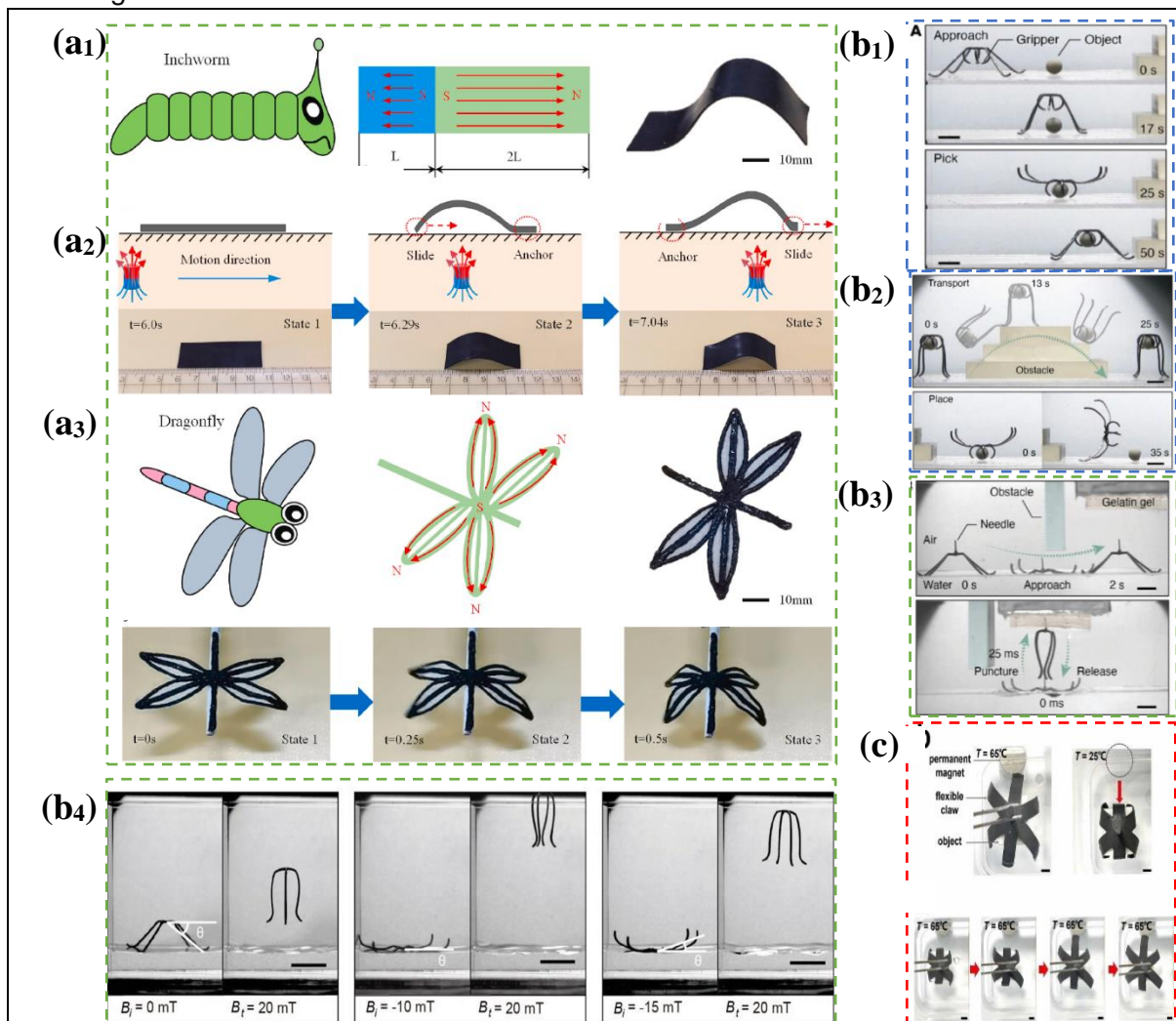


Figure 22. (a₁) Inchworm bionic soft robot schematic diagram, a plate-shape magnetic actuator magnetization domains of the bionic inchworm robot, and the bionic inchworm robot (prototype) drive by the magnetic actuation, (a₂) The bionic inchworm robot three-step motion stages with corresponding experimental results, (a₃) 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.); (b₁-b₃) A robot with a soft gripper picks, transports, and places a tiny object in water, (b₂) A robot with a needle overhead performs adaptive locomotion and targeted puncturing, (b₄) 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).

823 Magneto-active metamaterials or field-responsive novel origami structures whose shapes or
 824 properties modulated under a magnetic field hold great promise for many applications [499].
 825 For instance, Moonesi et al. [500] reported novel 3D-printed origami-inspired scaffolds using
 826 Fe₃O₄ and cellulose acetate (CA). Results demonstrated the cells' favourable surface
 827 morphology, superparamagnetic behavior, wettability, and appropriate compressive stiffness
 828 for cell proliferation, prominently decreased degradation, and acceptably low iron ion release
 829 of the printed scaffolds. Moreover, an optimized foldability with varying scaffold architecture
 830 was observed under magnetic field stimulus due to the presence of Fe₃O₄ magnetic particles
 831 which further allowed the scaffold folding, as presented in **Figure 23(a)**. Guan et al. [501]

832 developed a magnetically assisted DIW using alumina micro-platelets and fumed silica for
833 printing various structures. The printed structures had the ability to be turned into ceramics
834 with anisotropic properties, including their magnetic response, high electrical conductivity, and
835 self-shaping ability, as depicted in **Figure 23(b₁)-Figure 23(b₂)**. This work showed that
836 multilaterals with their magnetic response can be employed for multifunctional devices with
837 tailored and improved properties.

838 Luo et al. [502] prepared various magnetic-controlled liquid block structures with the ability to
839 program and reconfigure precisely under an external magnetic field. Liquid biomaterial inks
840 were prepared by gelatin methacryloyl (GelMA)/alginate (ALG) and carboxyl modified Fe₃O₄
841 MPs. Results showed that various liquid blocks including H-type and the spinal column-like
842 scaffolds demonstrated biomimetic morphologies and various functions, as presented in
843 **Figure 23(c₁)-Figure 23(c₃)**. Thus, considering the outstanding biomimetic functions from
844 natural materials the mentioned liquid blocks above together with the essence of the
845 magnetically controllable show great application potential for tissue engineering.

846 Zhao et al. [503] prepared personalized 3D printing of a bio-designed tracheal scaffold using
847 shape memory PLA/Fe₃O₄ composites filament under the magnetic stimulus. Results showed
848 that a 3D-printed tracheal scaffold with glass sponge microstructure exhibited higher strength,
849 and shape fixation for its unique ability to adapt the complex environmental conditions in the
850 soft tissue of patients. Moreover, 3D-printed scaffolds changed to a temporarily deformed
851 configuration and deployed back into a conformed shape under magnetic field stimulus, as
852 presented in **Figure 23(d₁)- Figure 23(d₂)**.

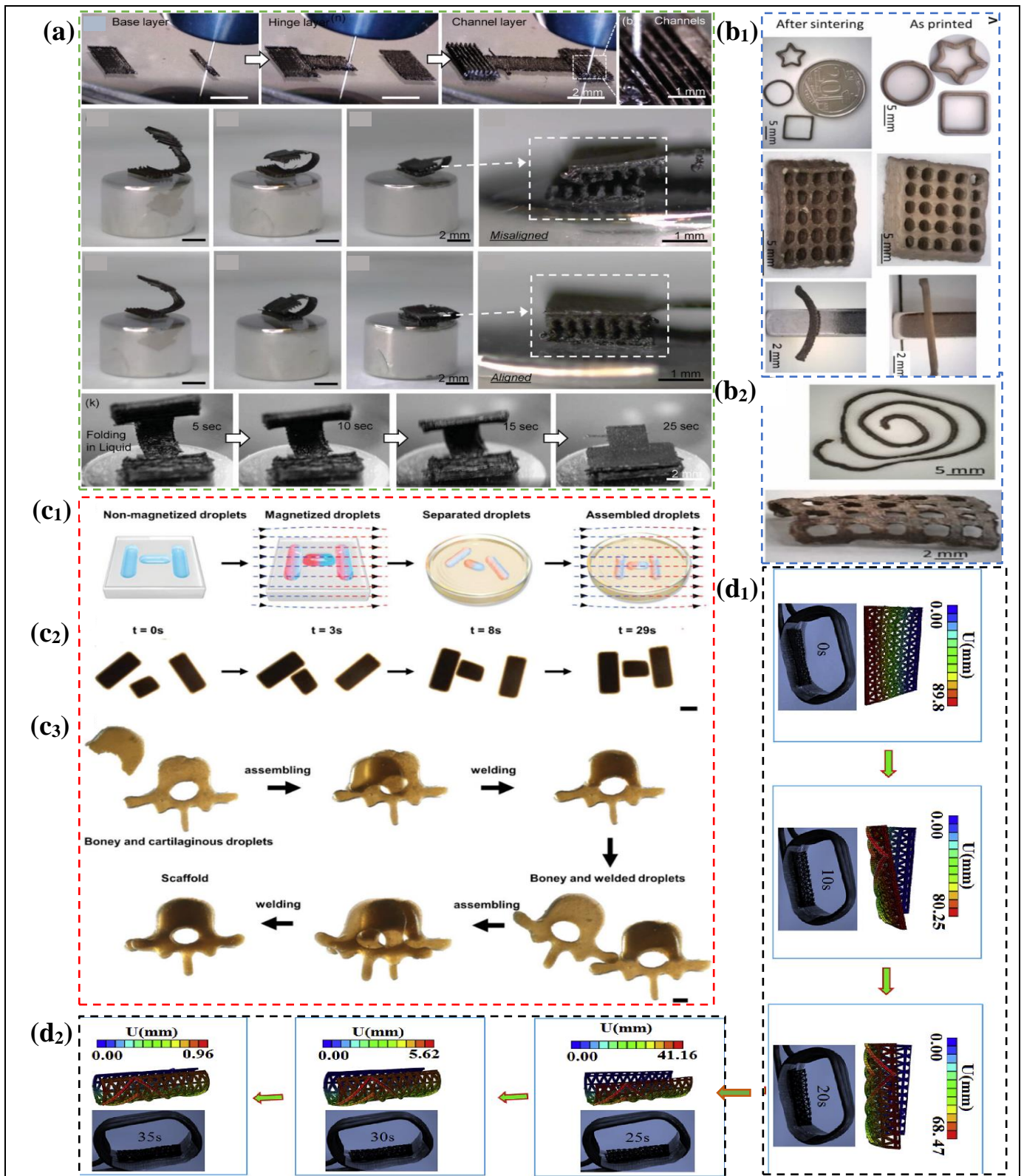


Figure 23. (a) Scaffold printing and foldability: on a Petri dish via solvent casting DIW and folding as a time lapse are shown with a Fe_3O_4 -MNPs with 7 mm long hinge and 15- Fe_3O_4 -MNPs base layers (adapted with permission from ref. [500], copyright 2023, Wiley-VCH GmbH); (b₁) Photos of 3D printed samples as printed and after sintering, (b₂) Complex 3D structures obtained after self-shaping during the sintering process (adapted with permission from ref. [501], copyright 2022, Elsevier B.V.); (c₁-c₂) Fabrication of "H"-shaped liquid blocks through all-liquid molding, magnetizing and patterning, c₃) 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); (d₁-d₂) Function verification of bioinspired tracheal scaffold in vitro actuated under magnetic field (adapted with permission from ref. [503], copyright 2019, Elsevier Ltd.).

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

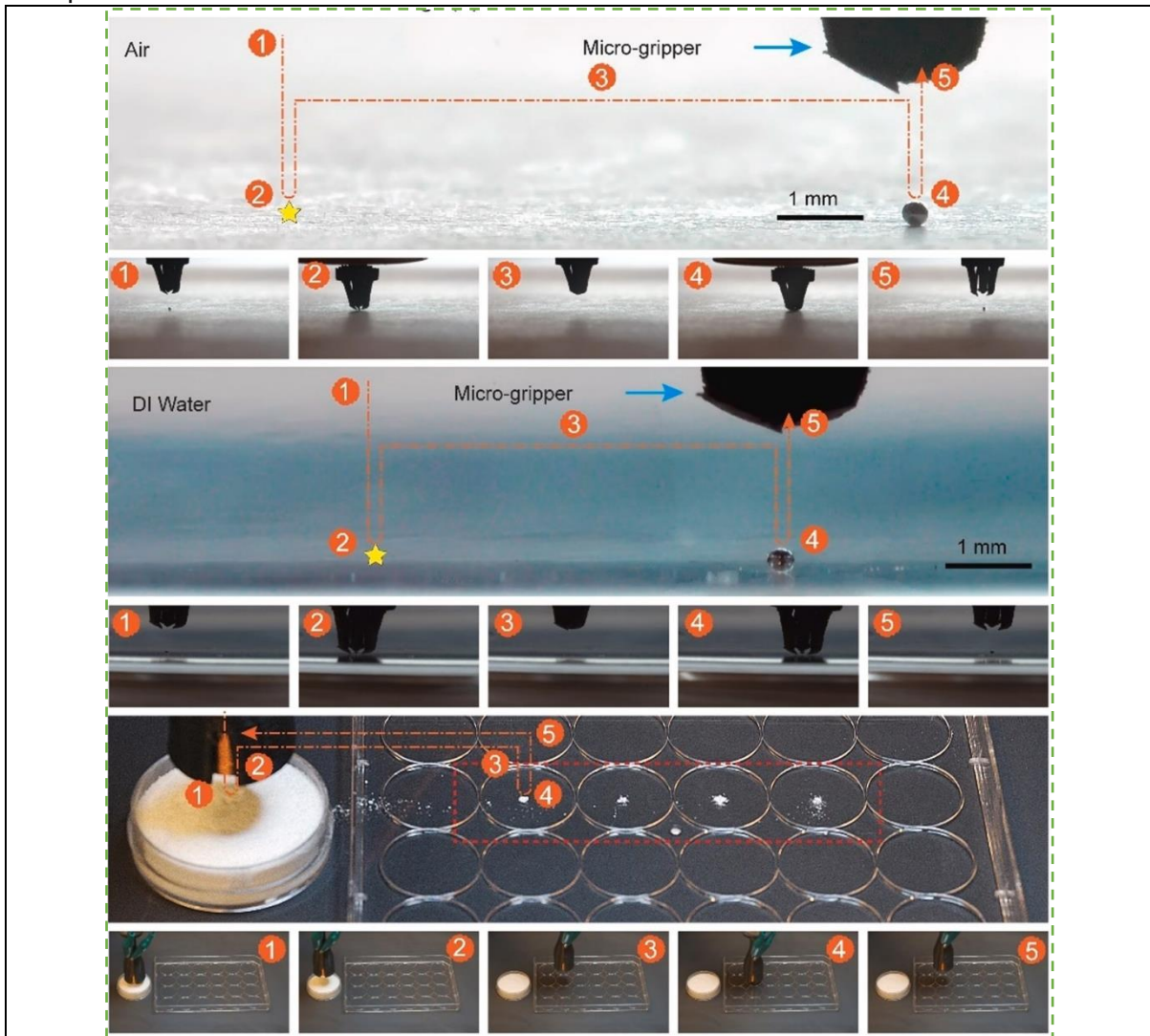


Figure 24. Manipulating and transporting tin microspheres in various mediums such as in air, in DI water and lastly for salt powders (adapted with permission from ref. [504], copyright 2021, Elsevier B.V.).

861 4.4 Advanced sensors and flexible electronics

862 In addition to performing many intelligent functions, stimuli-responsive smart sensors can
863 perform many tasks such as self-validating, self-testing, self-identifying and self-adapting as
864 part of their task or responsibility [505]. As opposed to conventional sensors, smart sensors
865 can manage their functions by being stimulated by external factors (external environment) in
866 which they are located and thus manage a variety of conditions. These features of smart
867 sensors are particularly attractive for achieving self-adaptation, advanced learning, and signal

868 processing architecture, within a single integrated circuit. Smart sensors are crucial for
869 designing stretchable electronics such as wearable monitoring systems, skin electronics,
870 invasive electrophysiological recordings, and prosthetics [506].

871 Flexible electronics-based devices are an emerging area and have extreme importance in
872 both engineering and biomedical sectors [507], [508]. Not limited to this, smart grippers,
873 flexible sensors, intelligent devices stretchable ionotropic devices and many more which have
874 not discovered yet are often required similar processing mechanisms for their operation [509],
875 [510]. There are various difficulties in these devices' fabrication through traditional 3D printing
876 techniques such as in unbalancing printability, shape fidelity, static nature, ionic conductivity,
877 stretchability, and other functionalities [511]–[513]. Such devices from 3D printing of smart
878 materials (4D printing) can greatly benefit from the remarkable patterning capability, complex
879 design, and shape-changing behaviors. More importantly, many smart materials in 3D printing
880 such as LCE demonstrate excellent recoverable shape-morphing organisms which are best
881 suited for applications such as grippers, valves, sensors, soft robotics, etc. [514]. Recently,
882 Han et al. [515] investigated novel magnetic microfibers, using NdFeB and PLA through
883 filament extrusion-based printing. The printed ferromagnetic microfibers were magnetized to
884 achieve various deformations of microfibers under magnetic fields. Moreover, the thickness,
885 mixing ratio, and length of the magnetized microfibers provided unique and customized
886 deformation of the microfiber for numerous applications in smart sensors and actuators.
887 Zhang et al. [516] developed a fully flexible soft robot through a light-cured 3D printing
888 technique using a tentacle-integrated liquid metal spiral wire with Nd₂Fe₁₄B magnetic
889 powders/Ecoflex (liquid silicone) composites. The various fabrication parameters were
890 optimized for achieving good energy transmission efficiency between the two tentacles of soft
891 robots. Moreover, printed soft robots demonstrated unique motion under an external magnetic
892 field as depicted in **Figure 25(a)**. Also using electromagnetic induction these soft robots can
893 transmit electric signals to the oscilloscope.

894 Another novel study by Dezaki et al. [517] explored 4D-printed MRE composite actuators using
895 silicone resins loaded with strontium ferrite-based MNPs and a thin conductive carbon black
896 PLA. The developed composite actuator with programmable magnetic patterns showed
897 excellent shape memory behavior such as electroactive under Joule heating and magnetic
898 fields. Moreover, the printed actuator (1.47g) can lift weights to 200 g. As such, the developed
899 printing process provided highly remotely controlled shape-memory features of 3D-printed
900 composite actuators. Also, Sundaram et al. [518] fabricated complex actuators (>106 design
901 dimensions) through multi-material drop-on-demand 3D printing using both soft and rigid
902 polymers with MNPs. Results showed that developed multi-material 3D printing with optimized
903 topology allowed complex actuators to use them in liquid interfaces as highlighted in **Figure**
904 **25(b)**. **Table 4** summarizes the state-of-the-art 4D printing technologies which are recently
905 been studied for various smart sensors and actuator-based applications. Likewise, Huang et
906 al. [519] used an interesting approach for fabricating Fe₃O₄ driven fiber-Tip multimaterial
907 microcantilever-based magnetic field sensor using an advanced femtosecond laser-induced
908 2PP technique. Insights of this study showed proposed sensor exhibited a minuscule size and
909 a high magnetic sensitivity of 119 pm mT⁻¹ in the range of 0–90 mT. Moreover, these sensors
910 showed the false-color scanning electron microscopy (SEM) images of the polymeric magnetic
911 microcantilever from the top view and the side view as presented in **Figure 25(c₁)-Figure**
912 **25(c₂)**. Thus, this new facile approach can be employed for different stimulus-responsive
913 microsensors and micro-actuators on the fiber tip. Saiz et al. [520] showed that magneto-
914 responsive PCL/Fe₃O₄ inks containing up to 10 wt% Fe₃O₄ can be employed for high level
915 microstructures with fiber diameters of 9.2 ± 0.6 μm using novel melt electrowriting-based 3D
916 printing technique. Reported results demonstrated that printed samples exhibited tunable
917 magnetic responses under various MNP concentrations and multi-material designs, as
918 presented in **Figure 25(d)**. This methodology can bridge the wide-open gap for designing

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various complex structures at the microscale level using different active fillers combined for many mysterious applications.

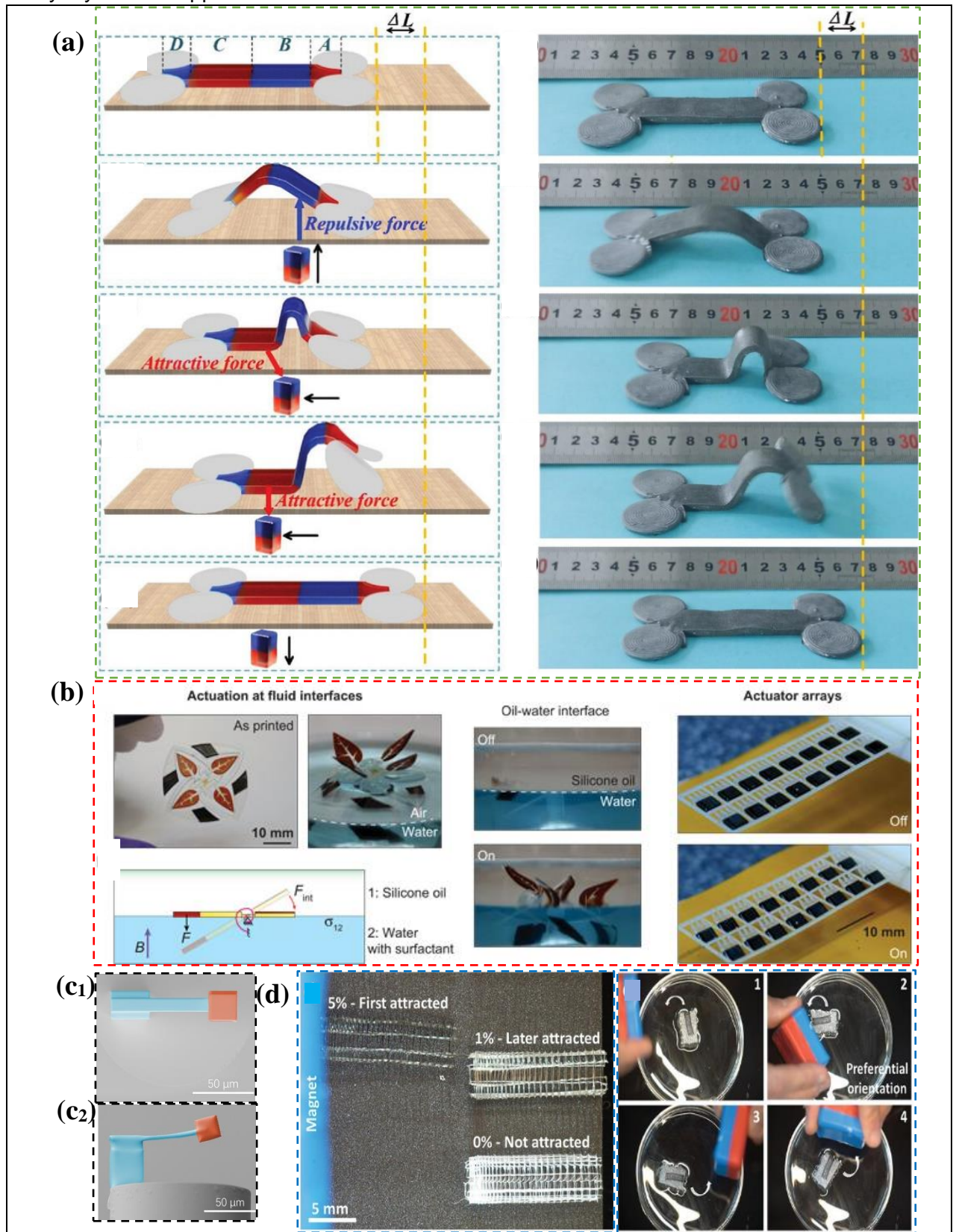


Figure 25. (a) Schematic and real-time crawling motion of the soft robot at various moments in a cycle (adapted with permission from ref. [516], copyright 2023, Wiley-VCH GmbH); (b) Magnetic actuator arrays capable of deforming under applied field use in liquid interfaces, some panels return to their 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); (c₁-c₂) 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% Fe₃O₄ 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)

921 **Table 4.** Summary of some recent works from 2020 to now on 3D printing of MASMs for soft robotics
922 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 ₂ O 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 ₃ O 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/MWCN T/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 ₃ O 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 ₃ O ₄	Magnetic field and heat	Bending	Soft robots	[539]
2022	FDM	PEEK/Fe ₃ O ₄	Magnetic field	Folding and bending	Electrical motors for space-compliant	[540]
2022	DIW	TPU/PCL/Fe ₃ O ₄	Heat and magnetic field	Bending and grasping	Flexible robotics	[541]
2022	SLA	PCL/Fe ₃ O 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 ₃ O ₄	Magnetic field	Rolling, jumping, and bending.	Soft robotics	[442]
2021	FDM	PHB/PCL/CNFs/Fe ₃ O ₄	Magnetic field	Bending	Smart actuators	[544]
2021	FDM	PLA/Fe ₃ O 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	GeIMA/CoFe ₂ O ₄ /BiFeO ₃	Magnetic field	-	Micro-swimmers for differentiation of neuron-Like cells	[547]
2020	SLS	PA-12/γ-Fe ₂ O 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]

923 5 Contemporary challenges and prospects

924 When the shape of a 3D-printed structure is designed to morph over time, it's referred to as
925 4D printing. These geometry shifts can be induced in any number of ways, with some of the
926 most common being electrical stimulation, heat, and moisture [551]–[553]. Mostly DIW and
927 DLP-based 4D printing methods are currently available and studied. However, novel 3D
928 printing techniques such as 2PP and micro-printing may provide a breakthrough in multi-
929 responsive tactics for complex shapes and efficient control over their shape-morphing
930 behaviors [554]–[556]. In a bid to emulate the movement mechanism of the printed structures,
931 the researchers employed computational design techniques that used selectively printing
932 'bend lines' into the geometry of the multilayer structures [557]–[560]. Material choice was also
933 crucial in 3D printing as the actuation of the smart material would only be possible with a
934 material responsive to any stimuli. Many 3D-printed objects are pre-programmed to morph
935 using intelligently placed layers and folds, which can contract and expand to give the desired
936 effect [561]–[563].

937 Most of the studies discuss only single material-based printing techniques while multi-
938 materials have huge potential in actuators for soft robotics, kirigami/origami and complex
939 structures, and controlled sequential folding [564], [565]. Furthermore, 3D printing at the micro-
940 scale has excellent potential to demonstrate various shape-morphing behaviors for the
941 possibility of releasing and trapping micro-objects. Various micro-shapes such as smart box-
942 like 3D microstructures, and microspheres can be useful for high-tech applications such as

943 on-demand drug delivery [566]–[568]. Also, soft devices are promising candidates in extreme
944 environments where human interaction is not possible. To date, their mechanical properties
945 are not up to the mark and thus 3D-printed soft robotics have limited use [569], [570]. The
946 time-dependent thermomechanical properties of soft actuators are also a promising field.
947 Furthermore, the soft actuators support heavy loads only at low temperatures but the load-
948 carrying capability at high temperatures is quite limited [571]–[573].

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

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

967 Soft materials such as polymers are prone to structural damage under external factors that
968 affect cracking, embrittlement, external loading, and eventual functional degradation. This
969 lowers their overall lifespan. This can be avoided through recovering functional performance
970 such as “self-healing” after incurring (minor) structural damage. One way to achieve this is
971 “self-heal ability” using polymer chemistries involving reversible primary and/or secondary
972 bond networks or embedded monomer reservoirs that use bio-inspired features [577].
973 Focused research is needed on sustainable soft actuators for achieving high performance and
974 mitigating environmental issues in terms of their waste at their end life [578].

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

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

991 Recently, the emergence of 2D materials allows us to achieve high mechanical properties of
 992 3D complex structures by mixing 3D printing and 2D materials such as graphene
 993 montmorillonites, carbon nanotubes, cellulose nanocrystals, carbon nanofibers, and so on,
 994 thereby forming shape memory polymeric nanoarchitectures [583] generally through DIW
 995 [584]. These novel 2D materials even at low concentrations such as 0.1 wt.% graphene
 996 nanoplatelets improved significantly shape recovery behavior [585].

997 A great deal of progress has already been made with stimulus-responsive magnetic actuators.
 998 For further improvements in their functions and to broaden their practical applications, there
 999 is still much to be done, as summarized in **Figure 26**. First, the 4D stage of soft actuators is
 1000 not mature enough to realize practical applications. However, overcoming the main bottleneck
 1001 including the fabrication of large parts and, mainly, the regulated transformations and
 1002 movement of actuator parts under external stimuli can pave the foundation for more practical
 1003 applications [586]. Second, it is still a challenge to produce more complex deformations for
 1004 precisely controlling their local stimuli response, particularly material handling. It is expected
 1005 that mag-bots used in remote, confined spaces with more complex designs for various
 1006 purposes such as material handling [587]. Third, besides macroscopic deformation, changes
 1007 in their other macroscopic properties such as color change could also be useful for opening
 1008 many avenues [588]. Fourth, commercialization of the printed actuators involves the synthesis
 1009 of novel SMP characterized by various types of response and advanced printing skills, all of
 1010 which are a major part of 4D printing [589]–[591]. Due to the lack of soft materials, their
 1011 commercial introduction is still at an early stage. Thus, significant attention needs to be paid
 1012 to the variety of 3D printers and the availability of smart materials for 4D printing perspective.

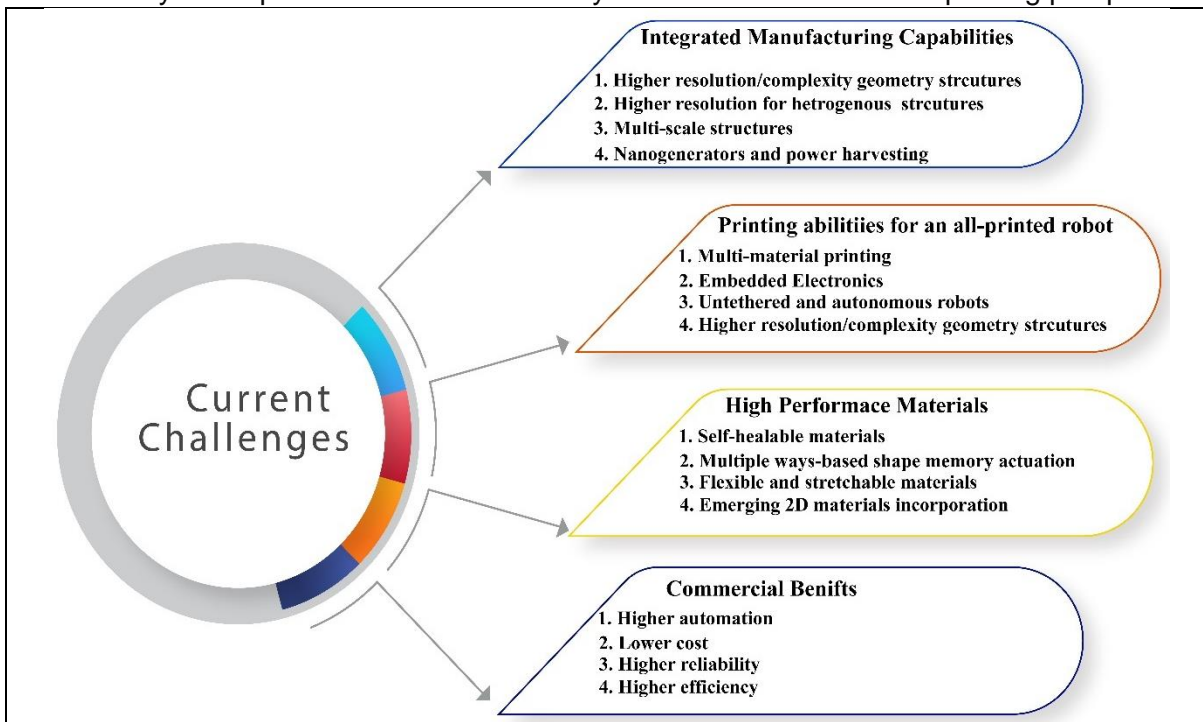
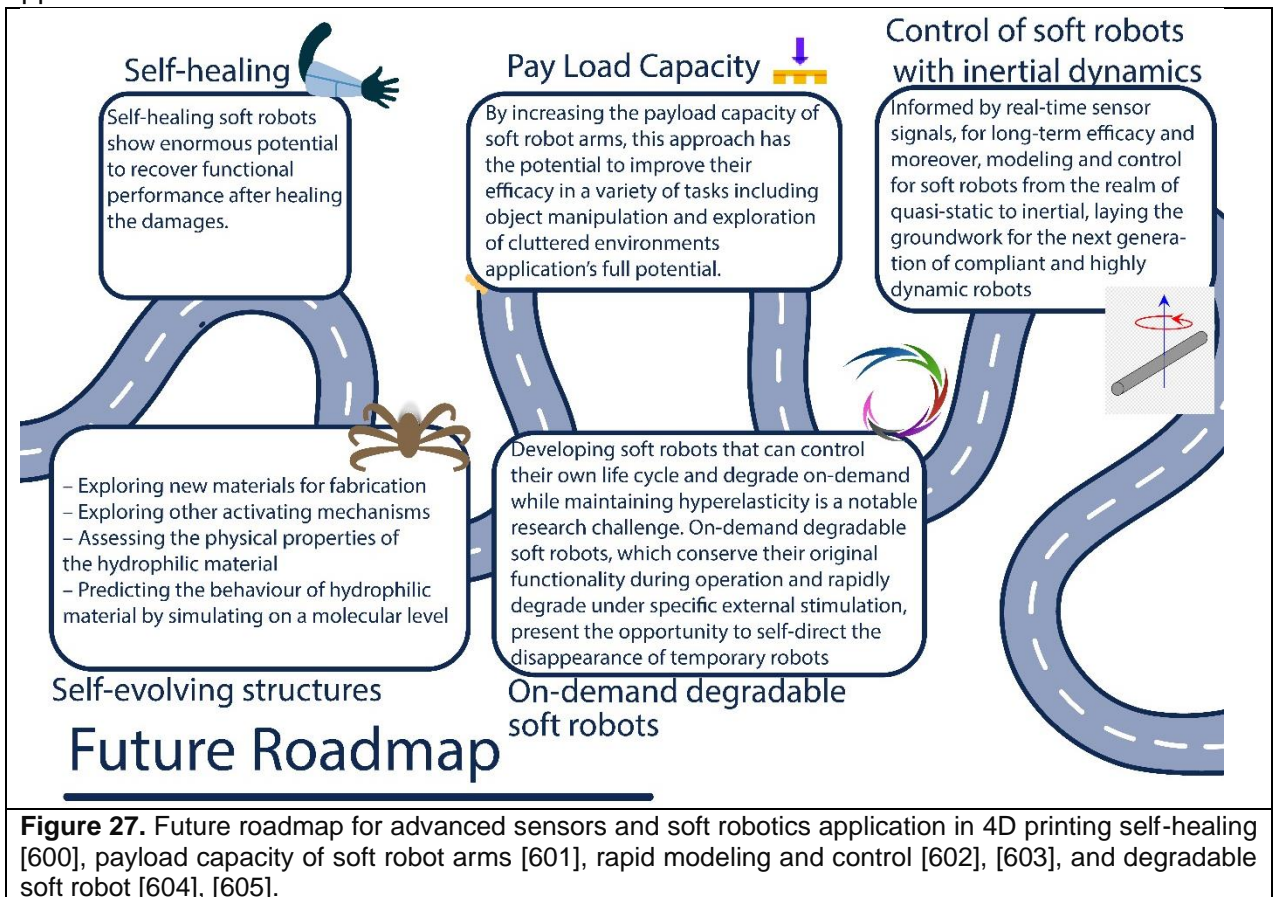


Figure 26. Future roadmap for advanced sensors and soft robotics application in 4D printing self-healing

1013 From laboratory evaluation to clinical application, safety aspects and regulatory pathways
 1014 should be considered. Due to the complexity of the human body, future research should
 1015 increasingly focus on the clinical use of microrobots as well as nanorobots [592] for alleviating
 1016 various challenges related to them such as detoxification, biocompatibility [593], biological
 1017 barriers, biosensing, biodegradation propensity and functioning in complex biological fluids
 1018 [594]–[596]. Biomedical applications often require magnetic soft robots to navigate in

1019 unstructured aquatic-terrestrial environments [597], [598]. Furthermore, for precise positioning
 1020 and efficient operation, the miniature magnetic robot needs to be enhanced both in terms of
 1021 controllability and agility. Recently, a 4D printed shape-programmable soft robot with near-
 1022 infrared light and magnetic stimulation was effectively employed for remote manipulation of
 1023 placing drugs, particularly in the application of hazardous chemical operations [599]. For
 1024 future research, we anticipate that several challenges related to the following areas need to
 1025 be addressed, as summarized in **Figure 27**. This will improve the functionality as well as the
 1026 performance of today's state-of-the-art soft robotics (referring to **Figure 28**) for many unknown
 1027 applications.



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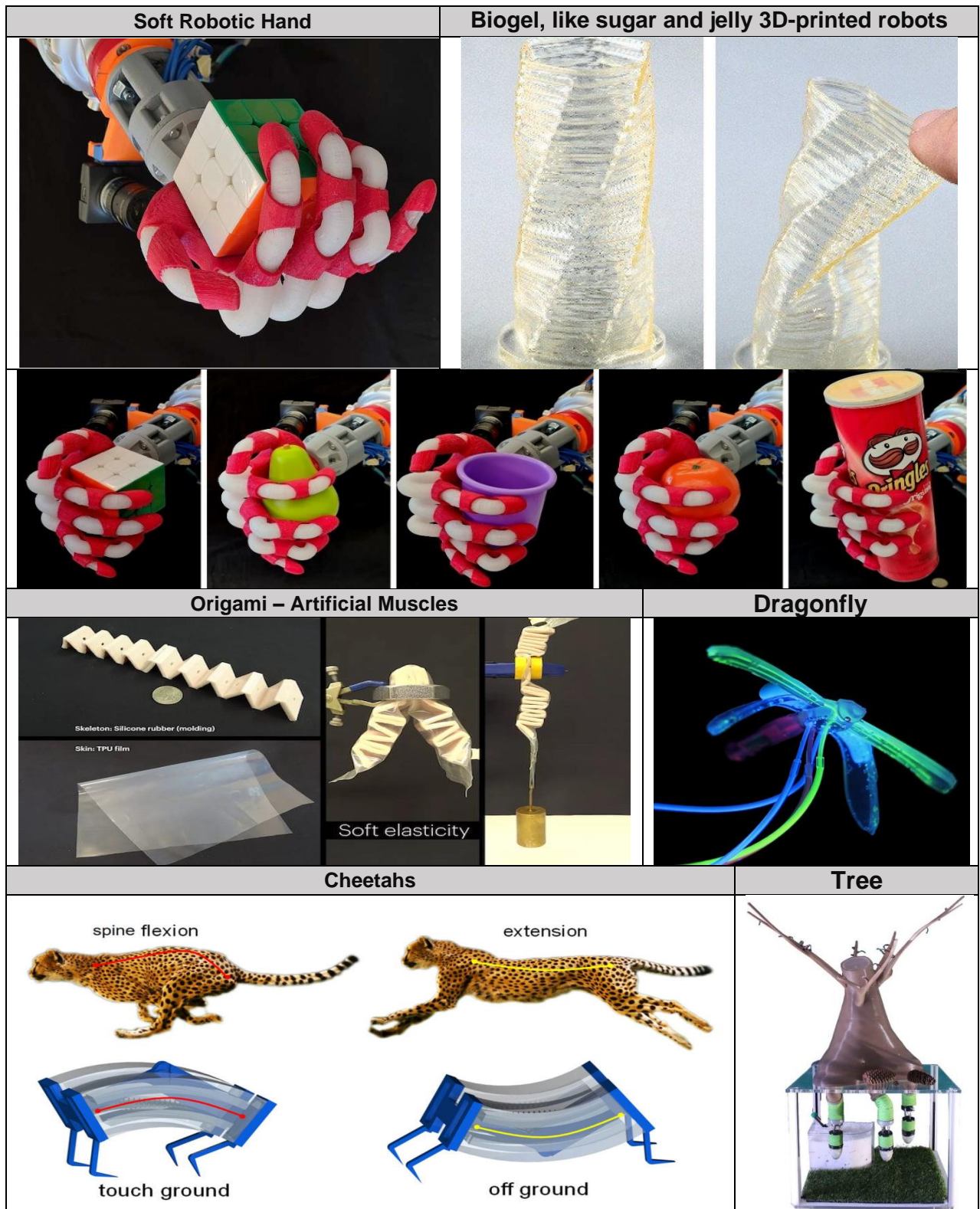


Figure 28. Emerging soft robotics in various shapes including soft robotic hand (adapted from [606] (Pic credit Elvis et al.), Biogel, like sugar and jelly 3D printed robots (Credits: A. Heiden et. at the Johannes Kepler University), Origami inspired Artificial Muscles and Origami Gripper, Dragonfly (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]).

1044 **6 Summary**

1045 Interestingly, we can learn a lot about shape morphing behavior of smart materials by drawing
1046 inspiration from nature. In this review, we have highlighted various 3D printing methods; new
1047 MASMs, and fabrications of various functional structures including sensors, and soft actuators,
1048 for broad applications in flexible electronics and biomedical. Particularly, this review study
1049 focuses on the justification of 3D printing of smart materials under magnetic stimulus for
1050 developing the state-of-the-art in soft robotics and providing recent breakthroughs in the
1051 proposed field. The 3D printing technology is replacing many traditional manufacturing
1052 techniques in the development of unthinkable, complex shapes and multifunction advanced
1053 sensors and actuator applications. It has been observed that the potential of 3D printing in the
1054 development of soft robotics has been significantly expanded due to emerging materials such
1055 as LCEs, polymers and their composites, and hydrogels for producing advanced intelligent
1056 devices. Furthermore, explications of the shape morphing mechanisms such as bending,
1057 twisting, and folding are easily achievable under the magnetic stimulus, which permits the
1058 printed actuators to gain control of their various soft robotics functions. Lastly, we provided
1059 some of the current 3D printing challenges such as low mechanical properties, response under
1060 multi-programming and stimuli that need to be addressed in future studies. Finally, we provide
1061 future perspectives, for the designing of the next generation of 3D-printed biodegradable and
1062 sustainable soft robots with much higher payload capacity. Thus, there is significant
1063 improvement required in the arena of 3D printing of MASMs, with more focused research
1064 towards its practical applications.

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1068 **Declaration of interest**

1069 The authors declare that they have no known competing financial interests or personal
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1071 **Data availability**

1072 No data was used for the research described in the article.

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