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► **To cite this version:**

Robin Delbart, Laurane Roumy, Marine Harcourt, Colin Robert, Fabienne Touchard, et al. 4D Printed Composite Actuators. Fabienne Touchard; Fabrizio Sarasini. Additive Manufacturing of Polymer-based Composite Materials: Materials, Processes, and Properties, Elsevier S&T Book, 2024, 978-0-443-15917-6. 10.1016/B978-0-443-15917-6.00005-0 . hal-04477283

**HAL Id: hal-04477283**

**<https://hal.science/hal-04477283v1>**

Submitted on 16 Oct 2024

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## 4D Printed Composite Actuators

R. Delbart<sup>1</sup>, L. Roumy<sup>2</sup>, M. Harcourt<sup>2</sup>, C. Robert<sup>3</sup>, F. Touchard<sup>2</sup>, T.Q. Truong-Hoang<sup>4</sup>,  
F. Martínez-Hergueta<sup>1\*</sup>

<sup>1</sup> *School of Engineering, Institute for Infrastructure and Environment, The University of Edinburgh, William Rankine Building, EH9 3FG, Edinburgh, UK.*

<sup>2</sup> *Institut PPRIME, Dpt Physique et Mécanique des Matériaux, CNRS-ENSMA-Université de Poitiers, France.*

<sup>3</sup> *School of Engineering, Institute for Materials and Processes, University of Edinburgh, UK*

<sup>4</sup> *ESTACA, Pôle Mécanique des Structures Composites et Environnement, Ecole d'ingénieurs, France*

\*francisca.mhergueta@ed.ac.uk

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### Abstract

This chapter presents a literature review on 4D Printed Composite Actuators for engineering applications. The recent integration of particle and fibre reinforcements into liquid crystal elastomers, hydrogels, and shape memory polymers enables the development of next-generation actuators with advanced functionalities and significantly enhanced load-carrying capacity. Proofs-of-concepts considering the nature of the stimuli-responsive material are categorised, and the advantages of the particle and fibre reinforced devices are presented. The approaches are illustrated with examples, exposing their capabilities and limitations. Advanced functionalities and modelling techniques are summarised. The main challenges associated with implementing 4D printing technologies in two key engineering applications: soft robotics and morphing structures are discussed.

*Keywords:* Additive Manufacturing, Actuators, Shape Memory Polymer, Modelling

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### 1. Introduction

4D printed devices are Additive Manufactured components able to transform energy into mechanical motion in response to specific external stimuli [1]. The fourth dimension addresses the change in the shape of the 3D printed component as a function of time. The baseline technology consists of a 3D printed stimulus-responsive smart material that can shape-shift between two different stable stress states when the appropriate stimuli are applied

*Preprint submitted to Additive Manufacturing of Polymer-based Composite Materials September 20, 2023*

29 [2]. 4D printed actuators are lightweight, low-cost and relatively simple in comparison  
30 to conventional hydraulic, pneumatic, electro-mechanical or electro-magnetic actuators [3].  
31 These characteristics allow them to perform a range of unique tasks, such as self-assembling,  
32 self-repairing, and bioprinting, not achievable with conventional actuation technologies. This  
33 enables the development of novel technologies for a large variety of applications, including  
34 autonomous sensing soft robots or morphing structures.

35 Although many review articles have been published in 4D printing to date, the rapidly  
36 changing nature of this research field requires the publication of new reviews formulating the  
37 current open research questions in the field. The addition of particle and fibre reinforcements  
38 in conventional liquid crystal elastomers, hydrogels and shape memory polymers enables  
39 the creation of multifunctional materials with photothermal, magneto-thermal and electro  
40 thermal properties, facilitating the triggering of the actuation. Furthermore, the addition  
41 of a stiffer constituent increases the load-carrying capacity of the actuators, enabling the  
42 creation of next-generation actuators with superior load-carrying capacity.

43 This chapter book presents a literature review of different 4D printed composite proof-  
44 of-concept actuators. The different approaches are categorised considering the nature of  
45 their stimuli-responsive materials and are illustrated with different examples in the litera-  
46 ture. Novel original experimental and numerical demonstrators created by the authors have  
47 been included. The chapter exposes the disruptive capacity of each technology while ad-  
48 dressing the current challenges and limitations for their widespread adoption in engineering  
49 applications. The potential of advanced functionalities enabled by the reinforcement is also  
50 summarised. Finally, the future trends in two key engineering applications: soft robotics  
51 and morphing structures are discussed.

## 52 **2. 4D printing definition and approaches**

53 Different methods are available to create 4D printed objects using a variety of 3D print-  
54 ing techniques [1] but the most common widespread technique combines different smart  
55 materials in the three dimensional space resulting in at least two metastable states in a 4D  
56 printed structure: the temporary shape, and the original shape that is recovered thanks to

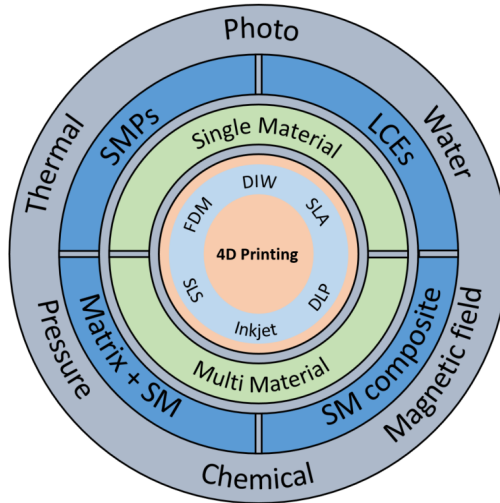


Figure 1: Summary of the different manufacturing techniques, materials and stimuli used in 4D printing.

57 an appropriate stimulus [2]. A large amount of 3D printing technologies are available in  
 58 the market compatible with 4D printing, summarised in Fig. 1. This includes Inkjet print-  
 59 ing, Fused Deposition Modeling (FDM), Stereolithography (SLA), Selective Laser Sintering  
 60 (SLS), Direct Ink Writing (DIW) and Digital Light Processing (DLP) [4, 5, 6, 7]. They have  
 61 been combined with different types of stimuli, such as water [8, 9, 10, 11], heat [12, 13, 14, 15]  
 62 or light [16, 2] to trigger the transformation. They can present multiple functionalities such  
 63 as self-sensing (the capacity to perceive information without relying on external sensors), re-  
 64 sponsiveness (the capacity to react in response to external stimuli), shape memory recovery  
 65 (the capacity to recover its original shape), self-adaptability (the capacity to autonomously  
 66 adapt its behaviour, configuration or performance in response to changing conditions) and  
 67 self-repair (the capacity to detect, diagnose and repair failure autonomously) [17, 18, 19].

68 The most critical component of a 4D printed structure is the stimulus-responsive material  
 69 used. This section summarises the main approaches available in the literature, classified  
 70 considering the different combinations of smart materials in one device and their nature. It  
 71 presents the advantages of particle and fibre-reinforced materials over their non-reinforced  
 72 counterparts. Emphasis is given to thermal-responsive approaches [20].

## 73 2.1. Shape Memory Polymers

74 Shape Memory Polymers (SMPs) are smart materials with the ability to change their  
75 shape in response to an external stimulus. This property is known as the Shape Memory  
76 Effect. The Shape Memory Effect is programmed by inducing internal residual stresses that  
77 result in a metastable temporary shape. Afterwards, the SMP is subjected to an external  
78 stimulus that allows the device to release the residual stresses and recover its original shape  
79 [10].

80 A series of programming steps lock the temporary shape to constrain the devices and  
81 induce residual stresses. As an illustrative example, in the case of thermo-responsive SMPs,  
82 the temporary shape is acquired due to the differences in material behaviour at the molecu-  
83 lar level when exposed o high temperatures. Semicrystalline and amorphous polymers have  
84 a characteristic glass transition temperature  $T_g$ , which divides the response of the materials,  
85 from the original stiff response at low temperature, into a rubbery state [21]. The program-  
86 ming procedure takes advantage of these fundamental physics to introduce internal stresses  
87 in four consecutive steps illustrated in Fig. 2: (1) heating the structure above the glass tran-  
88 sition temperature ( $T_g$ ), (2) deforming the object into a new shape (applying external load)  
89 while the temperature is kept above  $T_g$ , (3) cooling the structure below  $T_g$  while holding the  
90 external load and finally (4) releasing the structure to acquire a "temporary" shape that  
91 can recover its initial shape when (5) the structure is heated above the  $T_g$  again. In some  
92 cases, the programming steps are not mandatory and further details will be explained in the  
93 following sections.

94 A large range of stimuli is available in the literature to trigger the shape change in shape  
95 memory polymers:

- 96 • Temperature: Thermally responsive SMPs are the most conventional in literature.  
97 Their main physics have been explained in the previous paragraph, and will be further  
98 explored in this section.
- 99 • Light: UV radiation is generally employed to achieve shape fixation after deformation,  
100 photo-inducing the formation of new chemical bonds [22]. This mechanism can be

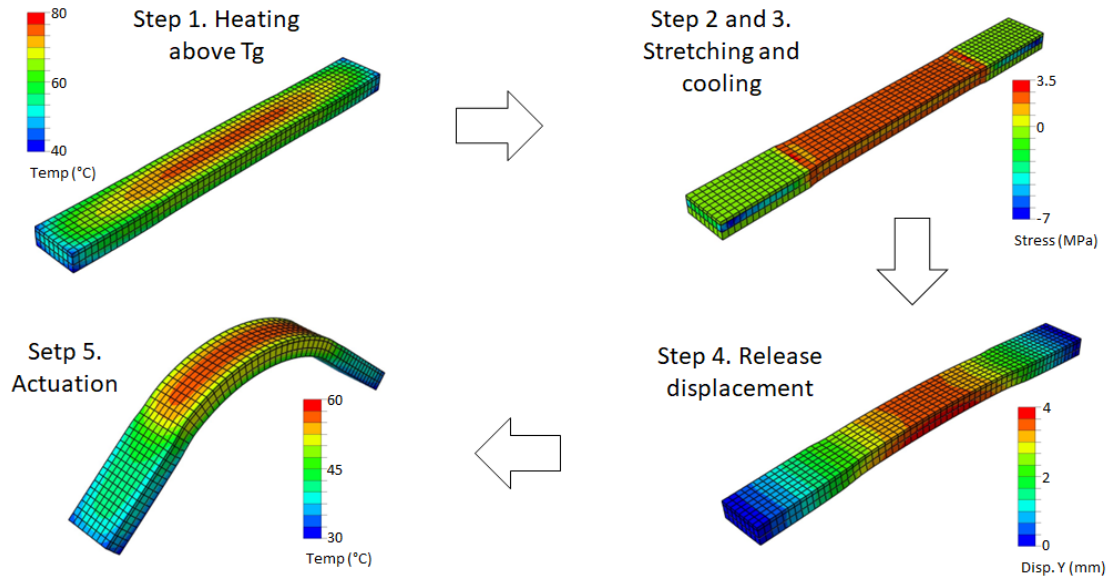


Figure 2: Programming steps and actuation of a conceptual bending actuator.

101 eventually combined with temperature-responsive motion mechanisms to achieve a  
 102 reversible trigger [23].

103 • **Magnetic Fields:** Magnetic nanoparticles can induce the shape change of a soft poly-  
 104 mer by particle realignment in the presence of a magnetic field [24]. This approach  
 105 produces patterned magnetic polarity devices compatible with soft elastomers. This  
 106 method does not incur in an increment of temperature and therefore is suitable for  
 107 bioengineering applications.

108 • **Solvents and pH:** Some SMPs respond to changes in solvent composition. This includes  
 109 water-soluble SMPs with a relatively low critical solution temperature which defines  
 110 the point at which the polymer changes from hydrophilic to hydrophobic behaviour,  
 111 inducing substantial changes in mechanical properties, transparency and shape [25, 26].  
 112 These are usually coupled to pH-sensitive shape change mechanisms, when subjected  
 113 to different acidity or alkalinity conditions. They have potential applications in drug  
 114 delivery systems, such as encapsulators of metallodrugs [27, 28].

115 For the particular case of temperature-responsive SMPs, two kinds of heating stimuli are

116 available; (i) the classical external sources of heat such as an oven, hot water or radiation  
117 [29, 30], and (ii) the indirect sources of heat such as the photothermal effect [31, 32, 33],  
118 the induction heating effect [34] and Joule’s effect [35]. The following paragraphs describe  
119 the underlying physics of the different indirect heating triggering methods, illustrating them  
120 with different 4D printing examples and analysing the main limitations of each technology.

121 Photothermal responsive SMPs are based on the phenomenon of transforming the elec-  
122 tromagnetic radiation of light into thermal energy. To provide photothermal properties to  
123 polymers, previous authors have employed SMPs reinforced with carbon nanotubes (CNTs),  
124 black ink, gold nanoparticles or nanorods [36]. These nanoparticles have a strong absorbance  
125 in the near-infrared region, high photothermal conversion efficiency and are low cost in the  
126 case of carbon nanoparticles (CNTs, graphene or carbon quantum dots). Photothermal re-  
127 sponsive SMPs have been successfully manufactured by Ge et al. [7] that used Digital Light  
128 Processing and a methacrylate-based resin to create a shape-shifting object such as an Eiffel  
129 tower able to recover from a temporary bent shape, and different objects including a grip,  
130 see Fig. 3. One of the main limitations of this technology is the ability to impose high strains  
131 above the  $T_g$  due to the brittle nature of these polymer blends. This is crucial to achieve  
132 large changes in shape and allow the implementation of technologies where large rotation  
133 angles or extensions are required. Such limitations have been found in the literature when  
134 using stereolithography or Digital Light Processing manufacturing methods since they are  
135 using photopolymers that are not very stretchable due to their high cross-linking density,  
136 which stands for the number of cross-link molecular bonds in the polymer.

137 Temperature-responsive SMPs can also be triggered by the induction heating effect,  
138 which appears when a polymer reinforced with ferromagnetic nanoparticles such as  $Fe_3O_4$   
139 is exposed to an alternating magnetic field. The rapidly alternated magnetic field goes  
140 through the object and generates an electrical current, the so-called eddy current, that flows  
141 through the material to heat it by the Joule effect, shifting to the second metastable shape.  
142 One remarkable example of this technology includes the concepts developed by Wei et al.  
143 [6] who produced a thermally and remotely actuated shape-changing SMP by direct ink  
144 writing, printing ultraviolet cross-linking Polylactic acid/ $Fe_3O_4$  composite inks, see Fig. 4.

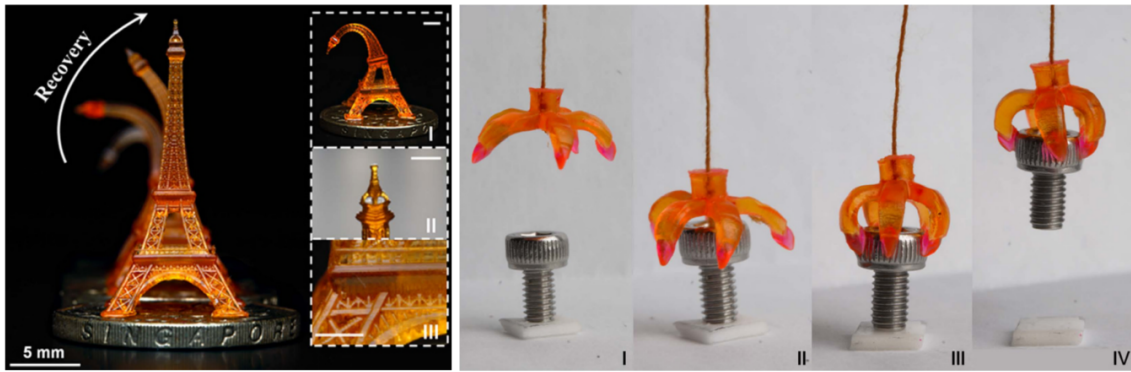


Figure 3: 4D printed photothermal responsive Eiffel tower that recovers from the temporary bent shape (left side) and the snapshots of a grabber that picks a screw (right side) [7]. Creative Commons CC BY license.

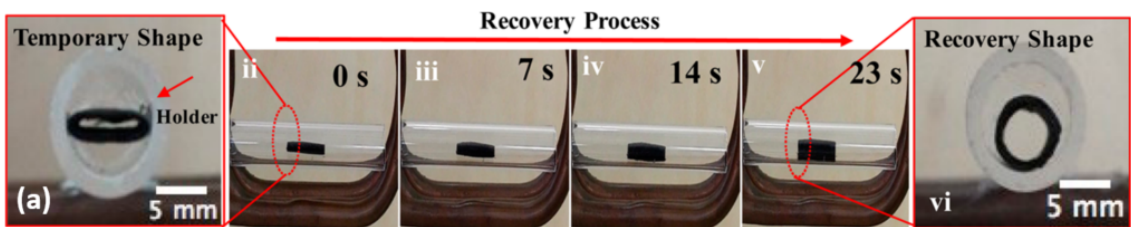


Figure 4: (a) Demonstration of the remotely-actuated shape-changing performance of a 4D printed nanocomposite cylinder in a 30 kHz alternating magnetic field. Reproduced with permission from [6].

145 This actuator has potential biomedical applications such as intravascular stents, as it is  
 146 magnetically guided and remotely triggered. However, the stent requires substantial heat  
 147 to be triggered and can cause unpleasant side effects in patients, which currently limits its  
 148 implementation.

149 In 2018, Kim et al. [24] created fast-transforming 3D printed soft materials actuated by  
 150 magnetic fields. The nozzle was modified to apply a magnetic field during the 3D printing  
 151 process of an elastomer composite containing ferromagnetic microparticles allowing the re-  
 152 orientation of the particles along the applied field to create a patterned magnetic polarity  
 153 device. This method does not require raising the temperature of the structure to trigger the  
 154 actuation, but can be only applied to soft materials with limited load-carrying capacity.

155 Very recently, few publications have reported investigations on Joule's heating [37]. This  
 156 approach relies on a multifunctional SMP able to change the temperature when subjected to



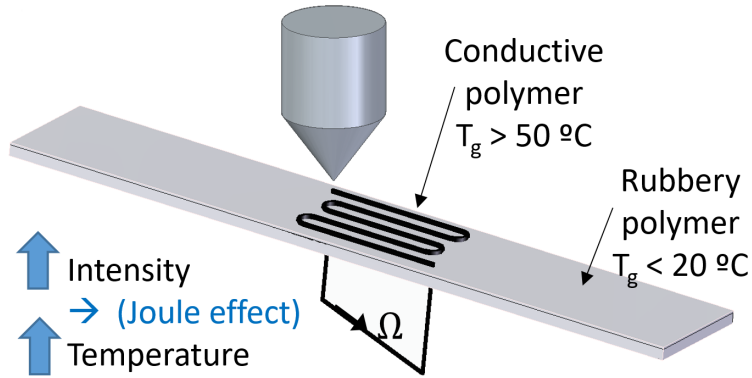


Figure 5: Electro-thermo-mechanical actuator. Conceptual design.

157 a potential difference. The concept is illustrated in Fig. 5. When a large [potential difference](#)  
 158 is induced, and the structure is self-heated over the  $T_g$ , the stiffness drastically drops such  
 159 that it can be deformed into a temporary new shape that can be locked once the electrical  
 160 generator is switched off and the structure cools down below the  $T_g$ .

161 The multifunctional SMPs are reinforced with conductive fillers such as metal nanopar-  
 162 ticles, carbon black, graphene or carbon nanotubes [38, 39, 40, 41, 42] to form a segregated  
 163 conductive network and acquire their electrical properties. The physical principle relies on  
 164 the distribution of the conductive particles to drive electricity from one pole to another,  
 165 this is called percolation [43]. Electrical conductivity develops through two mechanisms in  
 166 conductive polymers [44]: (i) electron tunnelling, referring to the “jump” of electrons from  
 167 one particle to the next one across distances through the polymer matrix and/or (ii) direct  
 168 contact of conductive particles, referring to the electron transfer via conductive filler path-  
 169 way. The Percolation theory establishes there is a minimum volume fraction of conductive  
 170 filler required for the composite to transition from insulation to conductive nature, which is  
 171 known as the percolation threshold. This threshold depends on many factors, including the  
 172 chemical structure of the matrix, and the shape, size, distribution, and conductive properties  
 173 of the filler [45, 46, 47, 48, 49, 50].

174 The electrical properties of a polymer composite can be defined by its resistivity, the  
 175 fundamental physical property that measures the inherent resistance of the material to the

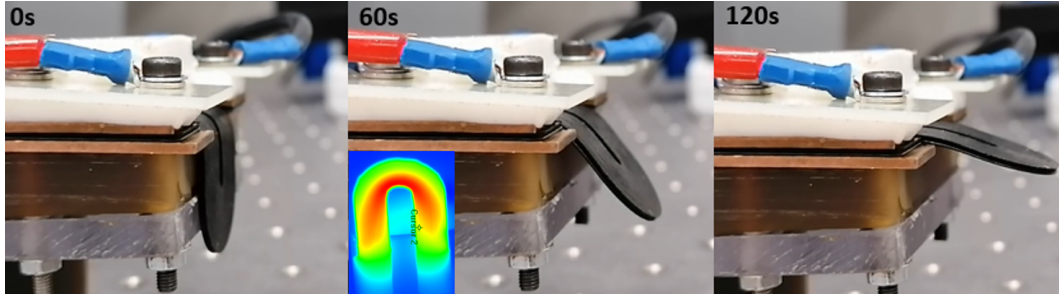


Figure 6: Snap shots of an electro-thermo-mechanical multifunctional 3D printed actuator with folding capabilities recovering its initial flat shape. Representative thermal camera temperature measurements during actuation.

176 flow of an electric current. When the electric current flows through a conductor with re-  
 177 sistance, the collisions between the moving charges and the atoms in the conductor cause  
 178 the electric energy to be converted into heat. This phenomenon is known as Joule heating.  
 179 These physical principles have been experimentally exploited recently to program [polylactic](#)  
 180 [acid \(PLA\)](#) 3D printed devices reinforced with CNTs and carbon black particles, increasing  
 181 the temperature above the  $T_g$  indirectly with an electric generator [51, 37, 52]. The studies  
 182 demonstrate the feasibility of the technology to unfold layers and recover their original flat  
 183 shape. Fig. 6 shows an electro-thermo-mechanical 3D printed folding actuator manufactured  
 184 in carbon black PLA able to recover its initial flat shape after being heated by an electric  
 185 current. The main current limitation of this approach is the difficulty to control the temper-  
 186 ature gradients and avoid irreversible damage and degradation of the electrical properties  
 187 when overheated.

188 Further efforts in the systematic characterisation of thermo-electro-mechanical proper-  
 189 ties are crucial to ensure the successful integration of self-heated particle reinforced SMPs in  
 190 engineering applications and control the motion of the devices during actuation with accu-  
 191 rate spatial and time resolutions. As reported in the literature, the resistivity of a polymer  
 192 increases with the temperature, showing drastic changes in trends above the  $T_g$  [53, 54].  
 193 Furthermore, the response of conventional actuation technologies can vary with the number  
 194 of applied cycles [55]. This progressive deviation hinders the development of a robust actu-  
 195 ation technology. The performance of the actuator may degrade over time or with repeated

196 use, which can affect its functionality and long-term reliability [51]. Addressing these issues  
197 and improving the durability of 4D printing is crucial for advancing the field and achieving  
198 reliable actuation technologies.

## 199 *2.2. Liquid Crystal Elastomers*

200 Liquid Crystal Elastomers (LCEs) are soft materials that combine the elastic properties  
201 of rubbers with the anisotropic properties of liquid crystals [56]. The physical principle  
202 behind the reversible shape variation of LCEs relies on the differences in stiffness between  
203 molecular chains along different preferential directions. Side-chain or main-chain LCEs with  
204 liquid crystal mesogenic molecules (or mesogens) can undergo a large contraction along  
205 the direction of the mesogens under external stimulus [57, 58], resulting in a prescribed  
206 deformation and motion.

207 The most conventional stimuli employed to trigger the shape-shifting functionality of  
208 LCEs are heat, organic solvents or light [59]. The exposure to such stimuli induces phase  
209 transitions of the molecular chains, aligning the molecules to modify the stiffness of the  
210 material. The main drawback to implementing this technology in engineering applications  
211 relies on the impracticality of applying the external stimuli (e.g., heating the surrounding  
212 air, immersion in hot water or exposure to infrared light) to induce the actuation process.

213 To overcome such limitations, current research efforts in LCE 4D printing are focused  
214 on the development of technologies with integrated self-triggering systems. For instance,  
215 Urayama et al. [60] developed a novel magnetic device able to align the polymer chains  
216 when subjected to an electric field. This approach led to a variable stiffness as a function  
217 of the particle orientation, and different electrooptical and electromechanical effects as a  
218 function of the cross-linking density. However, this technology currently presents a very  
219 low energy transformation efficiency, way lower than the conventional 40% ratio, since the  
220 energy applied with the magnetic field is much larger than the mechanical energy produced  
221 during the deformation of the device.

222 Alternatively, temperature gradients can be controlled by electrical circuits that can  
223 provide local heating to the structure. Yuan et al. [61] demonstrated the potential of LCEs

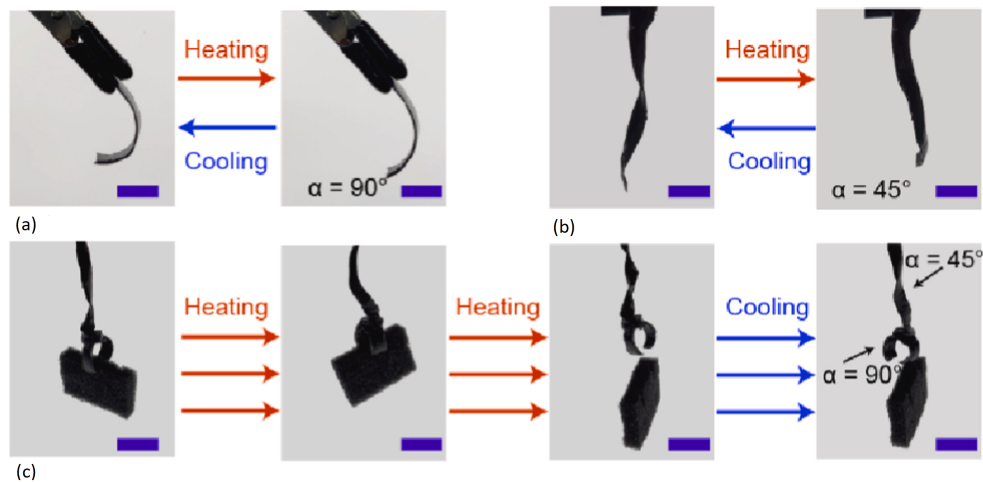


Figure 7: Thermo responsive fibre reinforced LCE actuator [66]. (a) and (b) recovery of the original shape from different twisting angles. (c) Combination of the previous actuators in a proof-of-concept grip holding a foam. Reproduced with permission from Elsevier.

224 in 4D printing to achieve reversible actuation with an integrated electric-control system. In  
 225 their work, LCE films were pre-fabricated [62, 63] and manually installed on 3D printed  
 226 objects [64, 65]. To achieve thermal actuation, conductive silver nanoparticle ink wires were  
 227 also printed at the locations where the LCEs would be installed. After inducing an electrical  
 228 current, the printed circuit produced Joule heating, increasing the temperature of the LCE  
 229 strip and resulting in an overall bending deformation of the 3D printed structure. When the  
 230 current was switched off, natural convection gradually cooled the actuator, leading to the  
 231 recovery of the original flat shape.

232 Recently, Xia et al. [66] developed fibre-reinforced LCE composites to eliminate the need  
 233 for secondary conductive wires, developing temperature-responsive grippers with modest  
 234 [load carrying capacity](#), see Fig. 7. This multifunctional composite actuator has a simpler  
 235 design in comparison to previous approaches, showing the potential of composite LCEs over  
 236 conventional LCEs to facilitate actuation. The reliability of actuation after cyclic electro-  
 237 thermal loading is a current open research question.

238 All the previous LCE approaches demonstrated reversible actuation, being a promising  
 239 solution to fully develop the field of soft robotics. However, the technology still presents

240 several limitations that hinder its implementation. First, the actuation speed which is very  
241 slow and limits its use for dynamic responsive applications. Second, the low TRL of the  
242 composite actuators, and the additional complexity of the self-triggered systems which rely  
243 on secondary elements such as metallic wires that can be damaged after multiple cycles.  
244 Finally, due to the rubbery nature of LCEs, their load-carrying capacity is limited, resulting  
245 in very compliant hinges not suitable for applications with large stress solicitations such as  
246 heavy-duty robots or morphing of primary structural components.

### 247 *2.3. 4D printed shape-shifting multimaterials and hydrogels*

248 The possibility to print objects with more than one material has allowed huge progress  
249 in 4D printing. This technology induces local strain gradients when combining materials  
250 with different mechanical properties, localising the change of shape on a specific area [67].  
251 Nowadays a large number of commercial 3D printing systems enable multi-material printing  
252 including composite hydrogels (water-responsive [8, 68] or thermal-responsive [69, 70]), SMP  
253 composites (fibre-reinforced active composites [71, 72] or bilayer SMP composites [73, 74]),  
254 multi-material SMP [14, 75] and desolvation-induced multi-materials [76, 77].

255 Hydrogels are hydrophilic materials that can be used in combination with other polymers  
256 to create an actuator. The main physical principle relies on their ability to imbibe large  
257 amounts of water making them swell. They have been used by Tibbits and coworkers  
258 [8] to create multi-material 3D printed structures composed of rigid polymer beams and  
259 hydrophilic rubber hinges that swelled when exposed to water. The hydrogel expansion  
260 created when drawn into water triggered the movement of the hinge that caused the joints  
261 to fold.

262 Hydrogels have also been used by Mao et al. [68] to create a reversible shape-shifting  
263 structure, see figure 8. Their technology combines hydrogels and SMPs in a multilayered  
264 structure, composed of an SMP top layer, an elastomer layer at the bottom, and the hydrogel  
265 layer in the middle. This hydrogel layer is 3D printed with holes to let the water go through  
266 and trigger the shape transformation. When the structure is drawn into cold water, the  
267 stiffness of the SMP is high enough to constrain the deformation induced by the swelling of

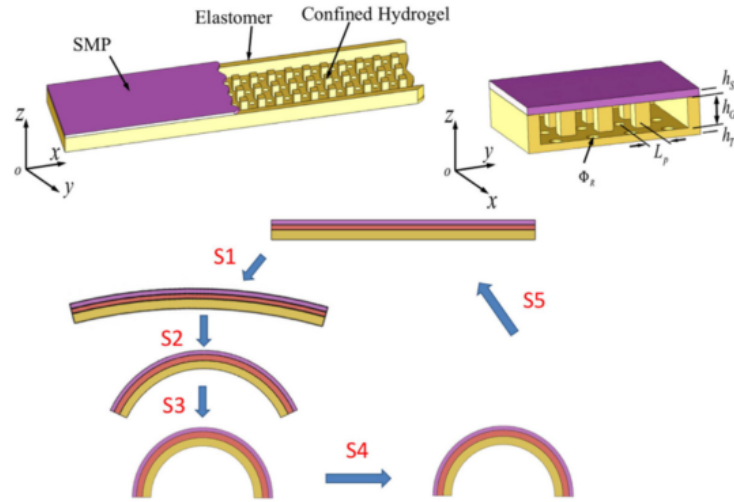


Figure 8: The reversible actuation component where the hydrogel is confined by the SMP and the elastomer layers. Schematic plots for a typical two-way actuation: In S1, the sample is immersed in water at low temperature; in S2, it is brought to a high-temperature environment and bends; in S3, it is cooled down to a low temperature; in S4, it is allowed to dry; in S5, it is heated to recover the original printed shape [68]. Creative Commons Attribution 4.0 International License.

268 the hydrogel, however, when drawn in hot water the SMP's stiffness drops drastically and  
 269 releases the hydrogel stresses, inducing the bending of the structure. As soon as the structure  
 270 is subsequently cooled and dried, it is locked in a temporary shape that is triggered with a  
 271 second heating cycle that makes it recover its initial shape, resulting in two different shapes  
 272 that can be inverted without the help of any mechanical forces. Similar methods have been  
 273 used with thermal-responsive hydrogels to create 4D printed structures [69, 70, 78]. The  
 274 main disadvantage of hydrogels consists of the reliance on water immersion to acquire the  
 275 new shape, which drastically limits the application of the technology for real world devices.

276 Unlike hydrogels, multilateral SMP actuators need to be programmed [79, 71]. Ge et al.  
 277 [12] printed a composite based on a rubbery polymer (TangoBlack, an acrylic-based material  
 278 created by Stratasys) with embedded rigid polymer fibres (veroWhite, another acrylic-based  
 279 resin created by Stratasys) using a polyjet 3D printer. With a glass transition temperature  
 280 of around 60°C, the fibres were used as SMPs and programmed to a temporary shape. Thus,  
 281 depending on the fibres distribution/fraction/orientation, complex movements were obtained

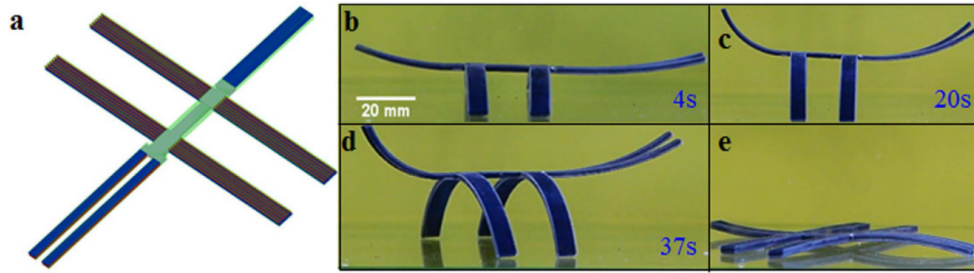


Figure 9: SMP composite actuator (a) CAD design. (b,c) The deformation behaviour of the active insect when put in hot water with temperatures of 30°C. (d) Rotated view of the structure in hot water. (e) Recovered shape in 60 °C water [15]. Creative Commons CC BY 4.0 License.

282 such as bending and twisting, to acquire coiled and folded final shapes. This concept has  
 283 been further investigated by Wu et al. [15] where different combinations of rigid and rubbery  
 284 polymers were employed to change the glass transition temperature and stiffness, resulting  
 285 in a laminated composite structure capable of achieving multiple shape changes at different  
 286 temperatures. Figure 9 shows a structure able to mimic an insect thanks to the combination  
 287 of SMPs with different glass transition temperatures. The main drawback of this technology  
 288 is the reliance on an external heat source such as hot water, which can limit its applicability  
 289 in various engineering applications.

### 290 3. Advanced functionalities

291 4D printing refers not only to shape-shifting but also to changes in material properties  
 292 or functionality over time. For example, 4D printing has transformed electronic devices into  
 293 shape-shifting functional devices [80, 77], produced devices with optical or electrical shape-  
 294 dependent properties [81], and allowed the development of devices with time-dependent  
 295 functional properties such as tissue maturation [82], biodegradability [83, 84], self-healing [85,  
 296 86, 87] or colour shifting [88]. This section summarises a variety of advanced functionalities.

#### 297 3.1. Programmable free, shape-shifting devices

298 Despite all the advantages reported for SMPs, thermomechanical programming can be  
 299 restrictive, so removing these programming steps by incorporating the internal stresses di-

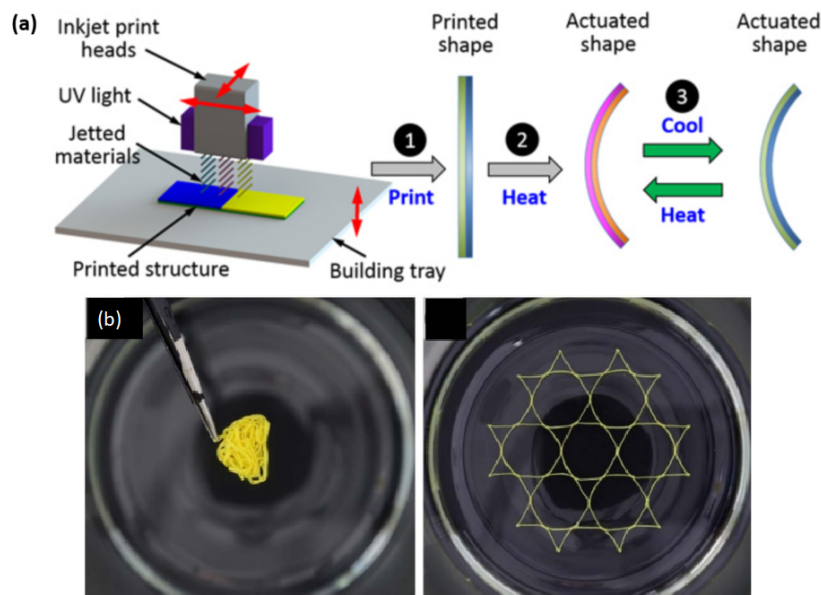


Figure 10: (a) The direct 4D printing approach exploits the ability to print multimaterial composites with internal stresses [89]. Creative Commons BY-NC 4.0 License. (b) Temporary shape of the folded 2D lattice taken out from 70°C water, then recover the original shape of the 2D lattice when plunged into 70°C water again, exhibiting a shape-memory effect [90]. Creative Commons Attribution 4.0 International License.

300 rectly during 3D printing presents a clear breakthrough in the development of autonomous  
 301 4D printing. Ding et al. [89] used this process to create a bilayer laminated strip com-  
 302 bining a soft elastomer and a variable stiffness SMP. Internal stresses were controlled with  
 303 the printing parameters and were released as soon as the structure was heated above  $T_g$ ,  
 304 changing its shape into a new permanent position, see figure 10(a). An et al. [74] printed a  
 305 so-called thermorph, a bilayer actuator composed of a PLA SMP active layer and a thermo-  
 306 plastic polyurethane constraining layer. The composite was printed flat and was triggered  
 307 to self-fold into complex 3D geometries (up to 70 faces) such as a chair.

308 Other techniques that do not require programming steps employ the heat-shrinkable  
 309 properties of SMPs. Indeed, when heated above the glass transition temperature, internal  
 310 stresses induced during the 3D printing process are released, triggering the change in shape  
 311 [91]. Zhang et al. [90] used FDM to print PLA 2D lattice structures to fold/unfold and  
 312 trigger the pattern transformation in a controllable way, see figure 10(b). The main disad-



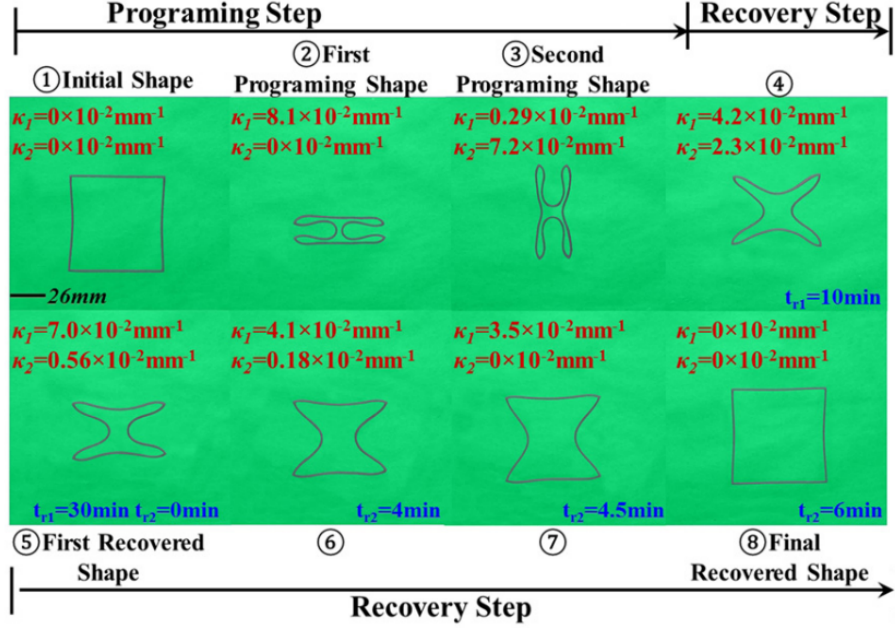


Figure 11: Triple-SME in the printed SMP box. Snapshots during the shape memory cycle, where  $K_1$  denotes the curvature of the two horizontal sides, and  $K_2$  denotes the curvature of the two vertical sides [92]. Reproduced with permission from Elsevier.

313 vantages of these techniques are the possibility to trigger the actuator only once. They are  
 314 also very sensitive to manufacturing parameters, making them barely reproducible.

315 Multi-shape actuators usually rely on a variety of transition temperatures (e.g., glass,  
 316 melting) or a broad transition range. Yu et al. [92] introduced a new method that did not  
 317 require a programming protocol, resulting in automatic shape change to multiple metastable  
 318 positions. They used an epoxy-based photocurable resin with a broad temperature range  
 319 of glass transition events around its  $T_g$  (approx 40 °C). The range of glass transition tem-  
 320 perature was used to create two temporary shapes, hence creating a multi-SME according  
 321 to the temperature [93, 94, 95, 96]. A triple shape memory square is shown in Figure 11.  
 322 Upscaling this technique is a current open research question due to the technical difficulties  
 323 to 3D print this polymer blend.

### 324 3.2. Reversible actuation

325 Most of the reversible actuation technologies available rely on Liquid Crystal Elastomers  
326 (LCEs, see section 2.2). They present a reliable solution for reversible actuation but with  
327 limited load-carrying capacity [59]. Very few examples of reversible SMP actuators have been  
328 reported in the literature. Wang and Li [97] combine the active SMP layer with a rubbery  
329 polymer to obtain a reversible actuation. They took advantage of the internal stresses  
330 induced by the 3D printing process when printing at high speeds (60 mm/s) to obtain a  
331 temporary bent SMP, avoiding the initial programming steps, able to shift to a flat shape  
332 at high temperatures when heated in an oven. The actuator included a passive confinement  
333 rubbery layer to induce internal stresses after cooling due to the different thermal expansion  
334 coefficients between the materials. As a result, the actuator recovered the post-printing bent  
335 shape, and the actuation could be triggered multiple times. The main disadvantage of this  
336 technology is the poor reproducibility of the residual stresses induced by the 3D printer, and  
337 the loose of functionality after several actuation cycles due to the progressive relaxation of  
338 the viscoplastic materials [98].

339 Reversible actuation can also be achieved by combining two SMPs with different glass  
340 transition temperatures and thermal expansion coefficients. Figure 12 (a) shows an example  
341 of a morphing structure 3D printed in conductive carbon black PLA and Acrylonitrile buta-  
342 diene styrene (ABS). The structure was composed of three beams which can be programmed  
343 at different bending angles. Figures 12 (b), (c) and (d) show the performance of a beam  
344 initially programmed by Joule's heating at 80°C, above the  $T_g$  of the PLA, but below the  $T_g$   
345 of the ABS polymer. Afterwards, it was subjected to three consecutive warming and cool-  
346 ing cycles at 100°C, the beginning of the ABS  $T_g$ . Figure 12(e) shows the evolution of the  
347 bending angle as a function of the time and the temperature. The first motion mechanism  
348 registered during actuation was the shape memory effect of the PLA, which straightened the  
349 beam from the temporary 130° bent angle, see Fig. 12(b) to 162°, see Fig. 12(c). Afterwards,  
350 during cooling, the difference in thermal expansion coefficient produced a second bending of  
351 the beam, generating a new temporary position, see Fig. 12(d). Consecutive cycles triggered  
352 reversible bending with changes of 8° in bending angle.

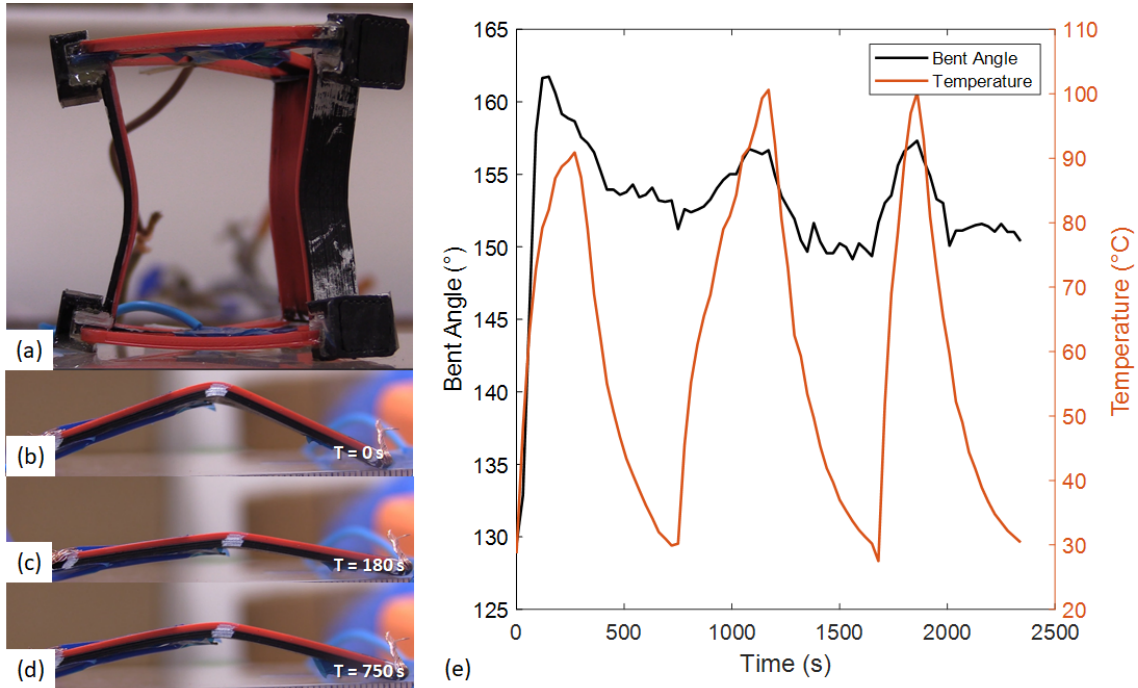


Figure 12: (a) Morphing structure prototype. (b), (c) and (d) response of the bipolymer beam at different time frames during the actuation stage. (e) Evolution of the bending angle as a function of temperature.

### 3.3. 4D bioprinting

The term 4D bioprinting refers to the 3D printing of biocompatible materials or living cellular structures that evolve over time after being printed [99]. This includes both shape-shifting biomaterials and the maturation of engineered tissues [82]. Kuribayashi-Shigetomi et al. [100] developed a self-folding origami cell formed by 3D cell-laden microstructures thanks to the cell traction force. Using microplates without a flexible joint, hollow microstructures with relatively low bending stiffness can be self-folded into shapes such as a cube, see figure 13(a). This process requires advanced 3D printers capable of manufacturing microarchitectures with resolutions of 3 to 4  $\mu\text{m}$  and must be totally bacterial free according to bioprinting standards.

3D printing of porous bioscaffolds shows tremendous potential for soft tissue reconstruction [101, 102, 103]. Kang et al. [104] reported an integrated tissue–organ printer that can fabricate stable, human-scale tissue constructs of any shape, such as in vivo bioprinted ear

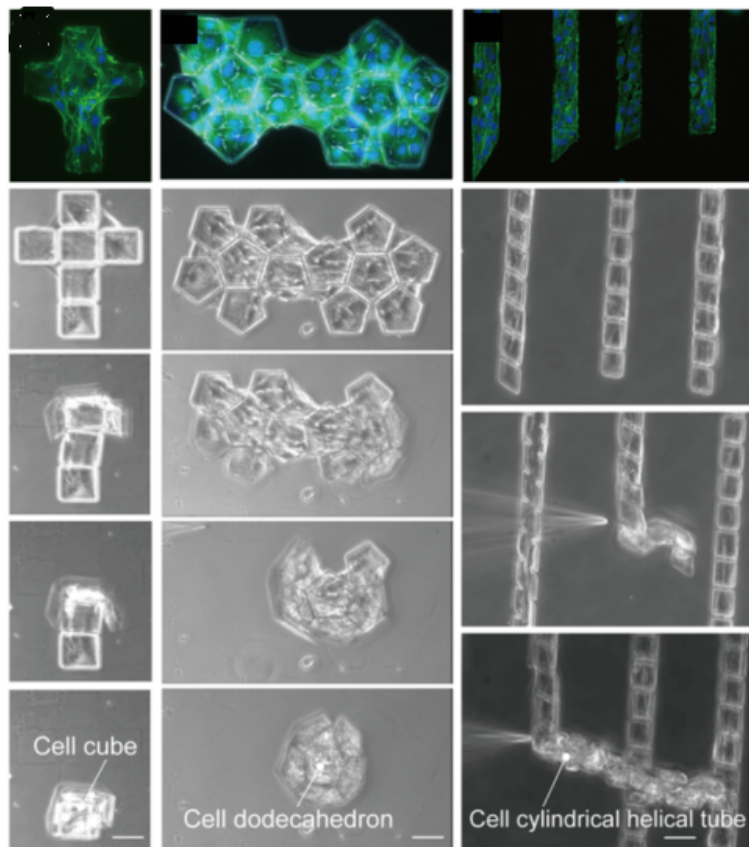


Figure 13: (a) Regular tetragon, regular dodecahedron and cylindrical tube with self-folding capabilities [100]. Creative Commons CC BY 4.0 License.

366 constructs. However, in addition to the previous limitations, this method has limitations in  
367 the size and stability of the printed organs, and it is currently unaffordable in comparison  
368 to conventional prosthetics.

#### 369 *3.4. 4D self-healing*

370 In order to extend the lifetime of material systems, self-healing materials have been  
371 fabricated to enable structural restoration and function recovery [105, 106]. Healing occurs  
372 at molecular level due to dynamic bond exchange reactions. These are typically achieved by  
373 using different types of dynamic covalent bonds, such as the Diels–Alder (DA) reaction [107,  
374 108], transesterification [109, 110], Schiff base chemistry [111, 112] and disulfide metathesis  
375 [113, 114].

376 Recently, Kuang et al. [87] developed UV-assisted direct ink writing (DIW) printing to  
377 fabricate a semi-interpenetrating elastomer network with a complex structure, high strain  
378 shape memory effect, and repeated self-healing capability. The diffusion and re-entanglement  
379 of linear polycaprolactone (PCL) chains in crack interfaces upon heating led to repeated  
380 crack healing, see figure 14(a). In addition, the effect of high strain shape memory could  
381 help heal large cracks in Vitrimers or covalent adaptive networks, changing their network  
382 topology through bond exchange reactions [115].

383 Yang et al. [117] developed a reversibly cross-linked thermoset based on furan-maleimide  
384 DA links for FDM printing. DA thermosets could be de-cross-linked and melt during printing  
385 at elevated temperatures ( $>90$  °C) and re-cross-linked at lower temperatures due to their  
386 entropically favourable state [118, 107]. Appuhamillage et al [86] blended a DA polymer  
387 with commercially available PLA to achieve remarkable mechanical properties by using the  
388 reversible DA reaction to improve the interfacial bonding properties after printing.

389 More recently, Zhang et al. [116] used a two-step polymerization strategy to develop 3D  
390 printed reprocessable thermosets that impart reshapeability, repairability, and recyclability  
391 into 3D printed structures see figure 14 (b). These recyclable and healable 3D printing  
392 methods contribute to alleviating environmental challenges associated with the continuous  
393 increase in consumption of 3D printing material.

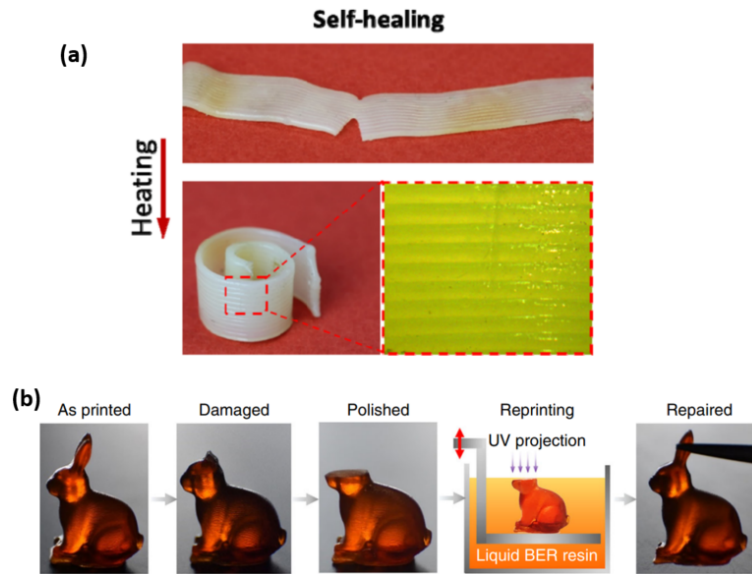


Figure 14: (a) Self-healing process for printed Archimedean spiral structure. Reprinted with permission from [87]. Copyright 2018 American Chemical Society. (b) Demonstration of the ability of the material to repair flawed printed structures: surgery on a damaged rabbit [116]. Creative Commons Attribution 4.0 International License.

394 To date, the development of composite actuators with self-healing capabilities has been  
 395 poorly reported in the literature. Delbart et al. [53] investigated the self-healing capacity  
 396 of a multifunctional carbon black-reinforced PLA composite subjected to Joule heating, see  
 397 Fig. 15. The evolution of void content was analysed by tomography, showing a progressive  
 398 reduction in void content when increasing the temperature of the device above the  $T_g$  due  
 399 to the resin flow. Reductions of 63 % in void/material pixel ratios were registered after one  
 400 Joule heating cycle, with the possibility to reduce up to 96 % of void content for annealing  
 401 processes. Despite the potential, the use of the previous technology to remove and heal  
 402 larger cracks is a current open research question, but it could be employed to repair these  
 403 actuators, for instance, after low-velocity impacts.

#### 404 4. Modelling

405 Considering the complexity of the couple electro/magnetic/photo/thermal-mechanical  
 406 response of these novel actuators, modelling can play a crucial role to gain deeper insight into

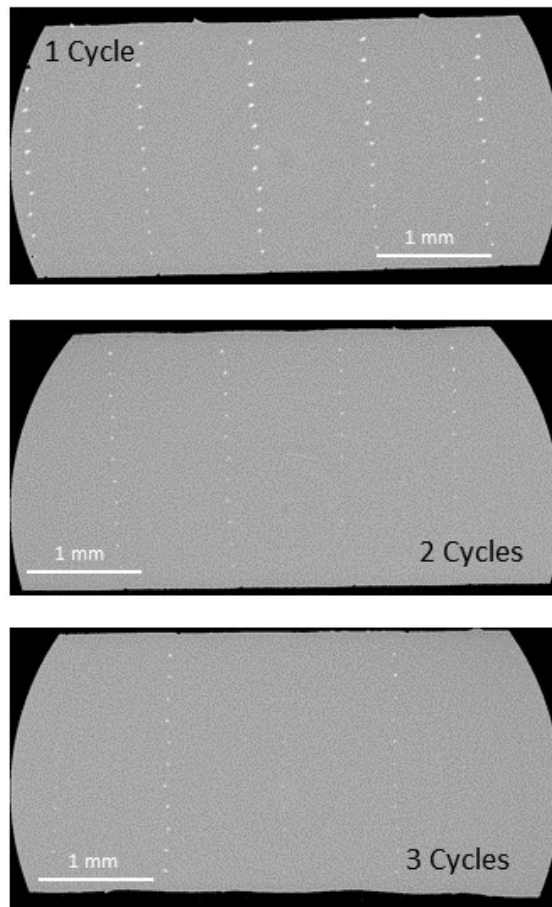


Figure 15: Representative tomographies showing the progressive reduction of void content in a carbon black PLA 3D printed component subjected to successive Joule heating cycles.

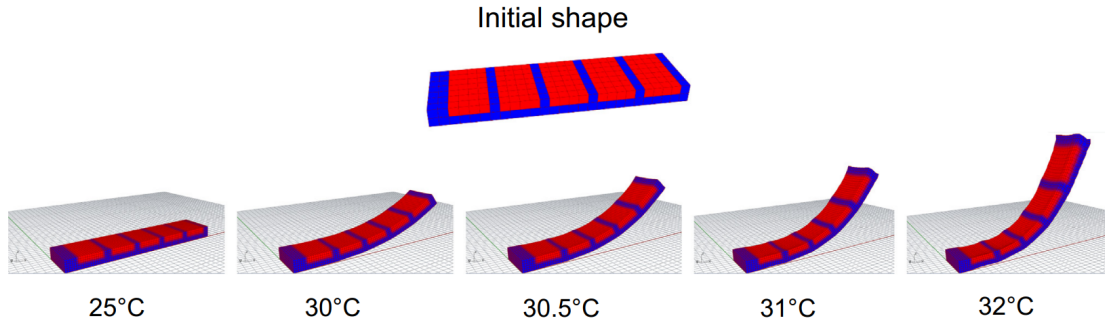


Figure 16: Numerical simulation of the hydrogel actuator [119]. Creative Commons CC-BY-NC-ND license.

407 the underlying physics involved in the mechanical response during the different programming  
 408 steps and actuation.

409 Sosson et al [119] developed a modelling framework for simulating smart materials con-  
 410 sidering a voxel basis. The modelled smart materials were limited to nonprogrammable  
 411 shape-shifting SMs, including piezoelectric materials, electro-/magneto-/photostrictive ma-  
 412 terials, and hydrogels. They used the material properties characterised by Westbrook and Qi  
 413 [120] to predict hydrogel shrinking and imposed bending. This motion was created by pat-  
 414 terning the hydrogel (red) into another non responsive hydrogel (blue) as shown in Fig. 16.  
 415 For instance, this framework has been employed to optimise the final shape of a plane wing  
 416 [121], see figure 17.

417 Other approaches based on the Finite Element Method (FEM) focused on the devel-  
 418 opment of actuators for soft robotics [122, 123] or morphing structures [124], providing an  
 419 efficient method to optimise the topology and the motion of the devices. The FE mod-  
 420 els provide insights into the actuator behaviour under different inputs such as pressures  
 421 [122] or temperature [125]. The bending angles and output forces of the actuator can be  
 422 characterised experimentally and used to validate the models. In addition, FEM can pre-  
 423 dict the force distribution for different sets of design parameters, considering geometrical  
 424 characteristics such as wall thicknesses and cross-sectional widths [122].

425 The level of complexity in the modelling approaches found in the literature is directly  
 426 linked to the complexity of the physical system being studied. Therefore, technologies that



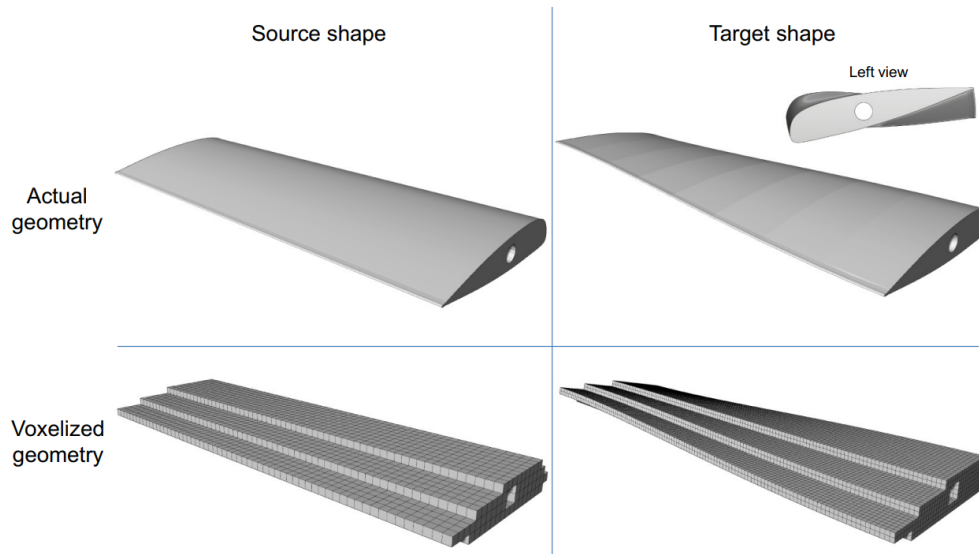


Figure 17: Wing twist problem formulation [121]. Creative Commons CC-BY license.

427 involve internal self-triggering mechanisms and couple the thermo-mechanical behaviour  
 428 with electro/photo or magnetic properties pose greater technical challenges and demand  
 429 additional efforts to achieve the necessary level of accuracy for engineering predictive tools.

430 For example, one action to improve the accuracy of the prediction of temperature-  
 431 responsive SMPs is to populate the existing experimental database of material properties,  
 432 with a focus on the plastic response of polymers across a large temperature range. This is  
 433 challenging experimentally, in particular above the  $T_g$  since the equipment for mechanical  
 434 polymer testing does not conventionally adapt to the rubbery response on that range of  
 435 temperatures. Most of the authors usually employ Dynamic Mechanical Analysis to charac-  
 436 terise the storage modulus of the polymer, and afterwards obtain indirectly the equivalent  
 437 Young's Modulus [97, 126], but that methodology does not provide information on the plas-  
 438 tic response of the polymer, which is crucial in 4D printing applications undergoing large  
 439 deformations. Conducting additional experiments and measurements to gather data points  
 440 that accurately represent the material's behaviour under different temperature conditions  
 441 is therefore necessary for the implementation of temperature-dependent constitutive models  
 442 with improved predictive capabilities.

443 Another limitation to model 4D printing relies on the development of robust multiscale  
444 modelling approaches. The development of mesoscale models that discretise the individ-  
445 ual filaments and their interaction with the adjacent ones is crucial to predict the internal  
446 stresses obtained during the 3D printing process which influences the behaviour of the sam-  
447 ple during actuation [127, 128]. However, the computational cost of such models is currently  
448 prohibitive, thus homogenised macromechanical approaches are used instead, losing resolu-  
449 tion of the physics occurring at microscale level [129, 130]. Furthermore, microscale models  
450 [131] that can account for the stochastic distribution of voids and defects in 3D printed  
451 samples are key to unravelling the role of each filament in the overall motion of the actua-  
452 tor, providing crucial information to optimise the internal architecture and design actuators  
453 with enhanced reliability.

## 454 **5. Engineering applications and future trends**

455 4D printed actuators offer several advantages over conventional hydraulic, pneumatic,  
456 electromechanical, or electro-magnetic actuators [3]. They are lightweight and compact,  
457 enabling the implementation of morphing technologies not compatible with complex me-  
458 chanical systems. Compared to traditional actuators, 4D printed actuators are low-cost  
459 and require fewer parts, resulting in reduced material and energy costs. Their simplified  
460 design and construction also leads to shorter assembly times and lower maintenance re-  
461 quirements. Potential engineering applications may include the deployment of origami solar  
462 arrays [132, 133], soft robotics [134, 123], intravascular and trachea stents [135, 24, 136],  
463 reflector antennas [137, 138] or tissue engineering [139, 140].

464 Despite the previous advantages, it is important to note that the current capabilities  
465 of 4D printing do not match the performance level of conventional actuators [141]. While  
466 smart materials can adapt to the environment, their functionality is limited compared to the  
467 capabilities of mechanical actuators, which offer a reliable and adaptable motion for various  
468 applications. 4D printed actuators require careful design and customization for specific  
469 applications, and their versatility is limited compared to the wide range of applications that  
470 can be achieved by adjusting operational parameters of off-the-shelf mechanical actuators.

471 For example, mechanical actuators can vary rotational speed simply by adjusting intensity,  
472 and pneumatic actuators provide a holding force by adjusting the pressure. The following  
473 sections will outline the main challenges associated with implementing 4D printing in two  
474 key engineering applications: soft robotics and morphing structures, and how 4D printed  
475 composites can contribute to advance the field.

### 476 *5.1. Soft robotics*

477 The development of advanced autonomous soft robots requires the integration of a sens-  
478 ing control system that can dynamically adapt the response to different robotic operations  
479 such as gripping or human-machine interaction [142]. Conventional approaches in the liter-  
480 ature incorporate electromechanical servos [143] or pneumatic actuators [144] into the 3D  
481 printed structure, however, the integration of these actuators results in complex components  
482 with relatively higher manufacturing, assembly, and maintenance costs than the counterpart  
483 emerging 4D printing technology.

484 The field of soft robotics can be potentially transformed by disruptive 4D printing tech-  
485 nologies. Soft smart materials possess an adaptive shaping capacity, which is particularly  
486 advantageous for mimicking biological behaviours [145, 146]. To date, most of the con-  
487 cepts for soft robotics incorporate LCEs to create sensing robots that can shape-shift when  
488 subjected to external stimuli such as light [145] or heat [147, 61]. Despite the potential,  
489 LCEs on their own are not compatible with conventional control systems used in robotics.  
490 Considering the previous, composite LCEs with multifunctional characteristics can enable  
491 the implementation of closed-loop control technologies [66], without the need to rely on  
492 secondary heating circuits based on copper or silver to warm the structure and trigger the  
493 actuation [147, 61].

494 The development of alternative 4D printing technologies based on composite LCEs and  
495 SMPs integrated within an electrical circuit is crucial to exploit the future abilities of soft  
496 robots. 3D printing allows the implementation of electrical circuits [39, 148, 149, 150] and  
497 integrated sensors [151, 152, 153] that can be employed to control the actuators and provide  
498 autonomy to the devices through an integrated control system [134]. Current efforts to

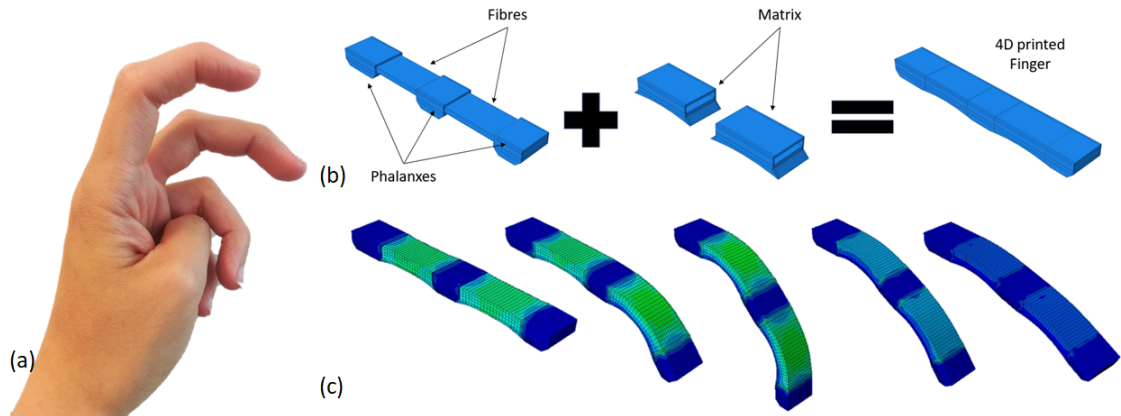


Figure 18: Design of a 4D printed finger. (a) Example of phalanxes rotations. (b) 4D printed finger concept. (c) Numerical simulation of the mechanical response during a bending cycle, showing a good correlation with the physiological response.

500 replace these conventional actuators are focused on the development of 4D printed joints  
 501 using multifunctional SMPs [154]. Fig. 18 shows the conceptual design of a simplified 4D  
 502 printed finger with two phalanxes composed of a rubbery matrix and SMP fibres. The  
 503 virtual component is able to mimic the rotations of the phalanxes, showing a bending cycle  
 504 during actuation. This technology can be potentially manufactured with multifunctional  
 505 temperature-responsive composites to allow controlling the movement and rotations as a  
 506 function of an applied voltage.

507 Despite the previous evidence of disruptiveness, further efforts are needed to ensure these  
 508 technologies are (i) reliable when subjected to continuous cycles, (ii) fully reversible without  
 509 the need for additional programming steps and (iii) responsive at given rates with improved  
 510 actuation speed to adapt to dynamic robotic operations.

## 510 5.2. Morphing structures

511 Morphing structures refer to systems that can autonomously alter their shape, mim-  
 512 icking organisms in nature [155]. They can undergo controlled transformations, enabling  
 513 them to adapt and optimise their performance to changing environmental conditions or user  
 514 requirements. The change in shape can be triggered by external stimuli or internal con-  
 515 trol mechanisms that will induce either a tensile or flexural deformation of the material, or

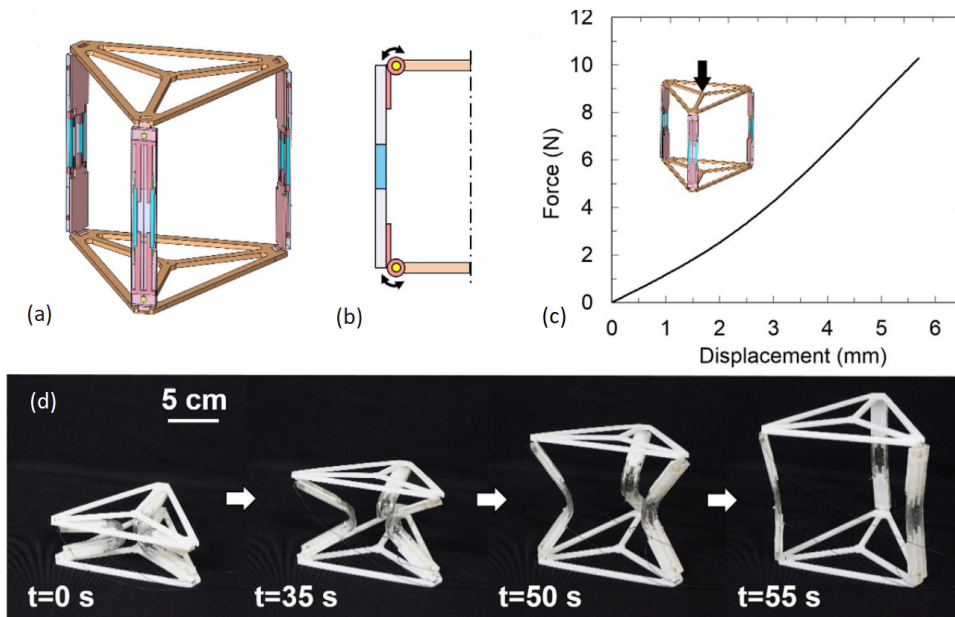


Figure 19: Triangular deployable trusses. (a) Conceptual design. (b) Sketched of the hinge smart joints. (c) Load carrying capacity of the deployed structure. (d) Snap-shots of the deployment process [160]. Creative Commons Attribution-NonCommercial-ShareALike 4.0 International License.

516 folding at creases [141].

517 The development of 4D printing plays a crucial role in providing the actuation capability  
 518 necessary for hierarchical motion in a variety of applications such as variable aerodynamic  
 519 drag [156, 157], variable stiffness [158] and deployable structures [159, 160], see Fig.19.  
 520 However, there are several challenges to address before 4D printing is implemented into  
 521 those real-world applications. This section focuses on load-carrying capacity, recovery of  
 522 folded creases and simultaneous actuation for large structures.

523 One of the main limitations of 4D printing is its overall poor load-carrying capacity. This  
 524 limitation arises from the inherent trade-off between compliance and stiffness in traditional  
 525 structural materials. Shape-changing objects require a certain level of compliance to undergo  
 526 large rotations and deformations, which is often at odds with the requirements for structural  
 527 integrity and load-bearing capabilities. The majority of morphing structures found in the  
 528 literature employ either soft polymers [124], or Kirigami principles [161, 162]. Incorporating  
 529 soft polymers or adding cuts and holes in stiff layers allow for increased compliance of

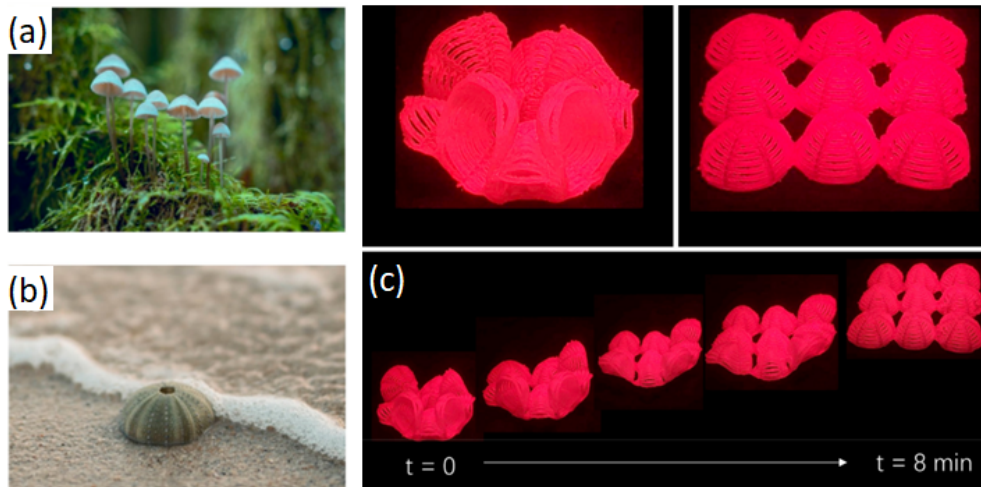


Figure 20: Example of mimetic morphing structures with energy absorption capacity based on dome-shaped creatures. (a) mushrooms and (b) sea urchins. (c) The recovery process of the deformed sample when the temperature is increased [165]. Reproduced with permission from Elsevier.

530 hinges, enabling significant deformation and rotations in the morphing structure. However,  
 531 these approaches may not meet the loading requirements of heavy-duty robots or structural  
 532 applications.

533 To address this limitation, innovative approaches have been developed to combine the  
 534 benefits of shape-changing materials with structural materials such as metals or composites.  
 535 This includes the integration of hybrid materials, such as Shape Memory Alloys [163] or  
 536 composites [164] that incorporate both compliant and stiff elements. Dezaki et al. [163]  
 537 found a balance between load-carrying capacity and shape-changing capabilities to manu-  
 538 facture a gripper with improved load-carrying capacity able to hold a weight up to 300 gr.  
 539 Biomimetic morphing structures have also been developed to improve the impact resistance  
 540 [165]. Shape-morphing domes with variable mechanical properties have been produced and  
 541 evaluated, finding a correlation between the number of circular units and the energy absorp-  
 542 tion capacity, see Fig. 20.

543 Aiming at the same objective but focusing on a different approach, certain authors are  
 544 developing smart actuators that do not rely on the Shape Memory effect. Their rationale  
 545 is based on the idea that by avoiding operational temperatures above the glass transition

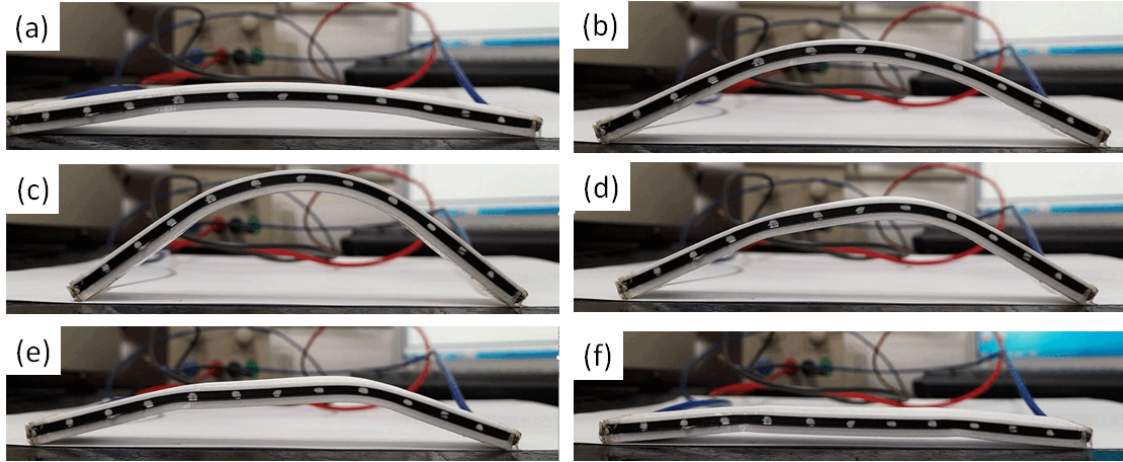


Figure 21: Bending actuator presenting a bending cycle. Figures (a), (b) and (c) at temperatures below the  $T_g$ , exhibit a larger bending due to the differences in thermal expansion coefficients. Figures (d), (e), and (f) at temperatures above  $T_g$ , show the recovery of the original flat shape due to the shape memory effect.

546 temperature ( $T_g$ ), actuation can still be achieved while preserving the mechanical proper-  
 547 ties of the active polymer layer. Recent investigations focus on the difference in thermal  
 548 expansion coefficient between adjacent materials that could produce a significant motion  
 549 below the  $T_g$ . Compared with the Shape Memory Effect, this mechanism is reversible and  
 550 quite repeatable, since it limits the degradation of the materials [37]. This principle has  
 551 been employed by Zhang et al. [166] to create a self-folding lattice structure with variable  
 552 global stiffness, and by Shao et al. [167] to manufacture grippers. The main disadvantage  
 553 of this approach is the scalability of the bi-polymer layered structure since it usually relies  
 554 on a passive rubbery layer that is difficult to produce. Fig. 21 shows a bilayer TPU/PLA  
 555 bending actuator that presents both motion mechanisms. For temperatures below  $T_g$ , a  
 556 larger bending is induced due to differences in the expansion coefficient. Above the  $T_g$  the  
 557 actuator recovers each original printed flat shape.

558 4D printing technologies also present limitations on folding and recovery capabilities,  
 559 which is crucial for the manufacturing of autonomous self-deployable origami structures  
 560 [168]. Origami allows folding truss or membrane structures in a relatively flat volume fol-  
 561 lowing a corrugation pattern, so they can be confined in small spaces. Some examples include  
 562 car airbags [169], paper shopping bags or sophisticated solar panel arrays [170].

563 The term rigid origami applies to structures with non-negligible thickness and consists  
564 of rigid panels divided by creases that act as hinges [171]. The stress-state on a crease is not  
565 uniform and varies along its entire length. The magnitude of the strain gradient for a given  
566 material depends on the crease geometry in terms of angle and radius, which defines the  
567 sharpness of the fold. The main implications for 4D printing consist of the generation of large  
568 plastic irrecoverable deformations at the creases due to the large strain gradients when the  
569 structure is folded. This problem is worsened for thermo-responsive devices programmed at  
570 temperatures above the  $T_g$  since the behaviour of the material is dominated by a viscoplastic  
571 response at that stage.

572 This plastic deformation avoids the full recovery of the original 4D printed shape. The  
573 recovery ratio is a trade-off between the compliance of the hinge and the curvature of the  
574 fold. For rigid SMP layers, full recovery for 4D printed layers is reported in the literature for  
575 very large curvature radii [172], and a progressive loss in the recovery function is reported  
576 as the curvature decreases [51]. This issue can be alleviated by lowering the programming  
577 temperatures, ensuring the plastic deformation imposed in the fold is minimised, but still  
578 allowing the development of the Shape Memory effect [37]. The optimisation of the program-  
579 ming temperatures to increase the sharpness of the fold and implement origami solutions  
580 with rigid 4D printed hinges is a current open research question.

581 Finally, the simultaneous triggering of multiple actuators in large self-deployed struc-  
582 tures is also a field that requires further research efforts. Most of the proof-of-concepts  
583 in the literature focus on small devices that cannot be scaled to large structures such as  
584 solar origami arrays. For instance, it might be difficult to operate simultaneously thermal-  
585 responsive actuators distributed along large deployed structures due to the presence of sub-  
586 stantial temperature gradients [8]. To conceive feasible 4D printing technologies for large  
587 morphing structures it is needed to further develop control systems that integrate actuators,  
588 sensors and control algorithms to actively drive and regulate the morphing process, ensuring  
589 autonomous, precise and reliable transformations.



## 590 **6. Conclusions**

591 The field of actuator technologies has seen significant advancements with the emer-  
592 gence of 4D printing and the utilisation of smart materials. 4D printed actuators offer  
593 lightweight, compact, and cost-effective solutions compared to conventional hydraulic, pneu-  
594 matic, electro-mechanical, or electro-magnetic actuators. They provide the ability to achieve  
595 complex motions, shape changes, and adaptability in response to stimuli. However, the cur-  
596 rent capabilities of 4D printing and smart materials still fall short of the performance level  
597 offered by conventional actuators. In particular, they lack the versatility, accuracy, reliabil-  
598 ity, functionality and load-carrying capacity of conventional actuators.

599 Composite 4D printing has the capacity to alleviate some of the previous issues. Com-  
600 posite 4D printing refers to actuators reinforced with particles or fibres, or hybrid structures  
601 composed of more than one material. The incorporation of conductive and magnetic fillers  
602 into conventional soft materials, such as shape memory polymers, liquid crystal elastomers,  
603 or hydrogels, offers the potential to acquire multifunctional properties. These fillers enable  
604 self-triggering capabilities, eliminating the need for external stimuli or secondary triggering  
605 systems to activate the shape memory effect. In the case of thermal-responsive devices,  
606 this enables self-induced shape changes due to the photothermal, induction, or Joule effect.  
607 Electro-thermo-mechanical devices are of particular interest due to their possibility to be  
608 controlled by an electrical circuit compatible with the close-loop technologies developed for  
609 autonomous robots.

610 Hybridisation with a second constituent (responsive or not) with different physical prop-  
611 erties has been employed to incorporate additional motion mechanisms. This includes dif-  
612 ferences in thermal or swelling expansion coefficients. Those motion mechanisms can be  
613 combined with the shape memory effect to create multi-shape actuators with advanced  
614 functionalities, such as programmable free, multi-shape and reversible actuators. Further-  
615 more, soft materials have been hybridised with stiffer constituents such as carbon fibres to  
616 increase the load-carrying capacity, at the expense of limiting the motion of the devices.

617 Despite the previous significant developments, the current capabilities of 4D printing

618 still do not match the performance of conventional actuators, and further research efforts  
619 are necessary to mature 4D printing technologies and implement them in fields such as soft  
620 robotics and morphing structures. Several open research questions still exist in the field,  
621 primarily focused on three key areas: the development of new materials, virtual optimisation  
622 and design exploration and integration with sensing and control systems. Additional ther-  
623 moplastic polymers, including unidirectional reinforced composites, should be considered in  
624 future stages. Reinforcing with continuous fibres would increase the loading capacity, how-  
625 ever, the continuous fibres will need to be strategically placed to maximise the motion of the  
626 actuator. The reinforcement would also influence the electrical conductivity of the printed  
627 actuator, therefore it needs to be carefully selected to ensure a low electrical resistance while  
628 being compatible with the glass transition temperature of the polymer matrix. Numerical  
629 approaches should focus on the development of high-fidelity element models. Multiscale sim-  
630 ulation frameworks with temperature-dependent constitutive models need to be developed  
631 to enhance the accuracy and predictive capabilities of actuator designs, and provide valu-  
632 able insights into the behaviour and performance of these systems. Finally, multifunctional  
633 4D printed composites could integrate 3D printed sensors to monitor the temperatures and  
634 deformations during the programming and triggering stages. Future investigations can focus  
635 on the development of a closed-loop control and autonomous operation, able to self-regulate  
636 the applied voltages to achieve a desired thermo-response and motion. This technology  
637 should enhance the actuator's responsiveness and precision.

## 638 **7. Acknowledgements**

639 The authors would like to acknowledge the economic support of the Defence Science  
640 and Technology Laboratory (Dstl), UK (grant number DSTLX1000144021R), the Defence  
641 Innovation Agency (AID) of the French Ministry of the Armed Forces (grant number AID-  
642 2021 65 0045) and the UK Space Agency (grant number UKSAG23-0001M). The scientific  
643 contributions of Marcelo Dias and Niamh O'Neill-Berest are kindly acknowledged.

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