

Chapter 12: 4D Printing of Smart Magnetic-Based Robotic Materials

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Abstract

Four-dimensional (4D) printing has been rapidly emerging in robotic manufacturing. In order to tackle delicate jobs, in various adaptive environments, such as sorting food, manipulating human organs and tissues, and locomotion in inaccessible terrain, stiff actuators, sensors, and even central controller units are often replaced with soft and flexible materials in traditional robots. Additive manufacturing of robots gained researchers' attention in recent years. Soft polymer materials, like poly-dimethyl siloxane (PDMS), have been developed and additive manufacturing has made it easier to practice them. Recently, a broader range of smart robotic materials with the capability of response and adaptation to external stimuli have been introduced in additive manufacturing. Soft robotics may now be actuated by external stimuli of electric, magnetic, chemical, thermal, or photonic origin thanks to the improved functionalities of smart materials that induce programmable modulations in size, shape, and stiffness. In operating soft robots, a magnetic field provides unique benefits, such as penetrating a variety of materials, remote commanding, and fast response time, with applications in an enclosed location, such as surgical catheters or untethered drug-delivering robots working in the human body. This chapter presents and categorizes numerous state-of-the-art electromagnetic robotics materials made using 4D printing. The operating principles, manufacturing processes, and their associated benefits and disadvantages, as well as future potential, are outlined.

Keywords: 4D printing; additive manufacturing; electro-magnetic; robotics; soft materials

Introduction

Robots may be seen in a new context as something that shares a collaborative environment with humans for medical and assistance means. This cannot be realized by conventional rigid robots and manufacturing [1-3]. This is one of the future themes, concentrating on factors such as robot flexibility and intelligence embodiment for adaptive interaction, compliance, and safety recently achieved via soft robotics. This sort of robotics relies heavily on the use of smart materials. But to deliver proper smart functionalities of the future robots one could optimize the controlled movement and force using smart robotic materials and four-dimensional (4D) printing.

Advancements in smart material functionality and additive manufacturing enable programmable modulations in size, form, and stiffness that allow sophisticated soft robots to be actuated by external stimuli of electric, magnetic, chemical, thermal, or photonic sources, expanding the possibilities of 4D printing of soft robotics.

Responsive materials that can change their size and/or stiffness in response to electrical and magnetic fields are the essence of electromagnetically powered soft robots. These materials must be able to endure mechanical stress while maintaining a low elastic modulus.

Magnetism may be used to direct the soft actuator's translational and rotational motion in a variety of ways. Elastomers and hydrogels are often mixed with micro- or nanoparticles [4]. In elastomeric composites, the dispersion of local electromagnetic properties may enhance motion control precision. Continuously distributed magnetization has been established to form sections with arbitrary magnetic sections that may be used to regulate motion [5]. These techniques can be used to generate novel robotic materials for soft actuators using 3D printing, origami, and kirigami [6, 7] as shown in Fig. 1(a).

The electromagnetic smart robotic materials suitable for 4D printing are categorized in this chapter, from the material point of view, into: magnetorheological elastomers (MREs), magnetic-based shape memory polymers (MSMPs), magnetic-based hydrogels (EMHs).

Magnetorheological Elastomers (MREs) in Robotic 4D Printing

A magnetic particle suspension in a viscous liquid is represented by an MRE, which is a physical representation of an MR fluid. Seismic vibration dampers and artificial joints are all examples of robotic applications where MREs are often employed [8]. The ordering force of magnetostatic particle-particle interaction, in MREs, outweighs the randomizing force of particle thermal motion, resulting in chain-like particle aggregates oriented along the direction of the magnetic field. An increase in the MR fluid's modulus is caused by the creation of chain-like aggregates, which stiffens the fluid.

Magnetic Particle Dispersion Strategies in MREs

Optimal characteristics of magnetic elastomers and hydrogels are achieved when the magnetic particles are evenly dispersed without aggregation. Blending, in situ precipitation, and grafting are the most popular procedures for dispersing magnetic particles [9]. The blending, which is called physical doping, involves combining magnetic particles, uncrosslinked polymer chains and crosslinking agents in a single container that is usually filled with a solvent or nonpolymerizing silicone oil [8]. During the crosslinking process, the magnetic particles, which do not form chemical connections with polymeric components, are held in place. In spite of its simplicity, the blending process still has limitations with dispersion and particle agglomeration control. Weak chemical bonds between the particles and the polymeric network make bleaching magnetic particles in swelling gels more difficult.

Hydrogels, but not elastomers, may be suitable for using in situ precipitation. Precursor solution containing magnetic metals is infused into a polymeric gel network, and then a chemical reaction is triggered to precipitate magnetic particles in the gel. The in-situ precipitation approach produces more uniformly dispersed magnetic particles than the blending

method, making it easier to include a large number of particles. This approach, on the other hand, is only acceptable for hydrogels that are able to withstand alkalines [10].

Magnetic elastomers are often made via the grafting-onto approach, in which the magnetic particle functions as a crosslinker by possessing chemically functional groups on its surface. The magnetic composite material made via this approach is more stable because of the covalent connection between the particles and the polymer chains [11].

Magnetic particles may have an isotropic or anisotropic distribution. While it is simpler to make isotropic materials, a magnetic field may be used to linearly organize the particles while the elastomer matrix is curing. The actuation behavior may be given directionality by such an arrangement of magnetic particles [8]. Magnetic actuation may be directed and efficient if the magnetic particles are trapped in micro-sized domains [12]. Only particular areas of the actuator are affected by an applied field, due to the distribution of magnetic particles inside predefined regions. By 3D printing patterns encased in nonmagnetic silicone, the effects of design and orientation on the efficacy of vibration damping in relation to applied magnetic field magnitude and direction were studied [13].

To 4D print MREs soft robots, the ferromagnetic particles are dispersed inside silicon- or rubber-based polymers (such as PDMS) that serve as excellent electrical insulators, such as iron, cobalt or neodymium-iron-boron (NdFeB) [14]. The external magnetic field also aligns the randomly oriented particles, resulting in changes in stiffness and damping [15] and actuations. Magnetic fields may improve the stiffness and shear modulus of MREs based on the particle size, concentration, and magnetic properties as well as the elastomer matrix [16, 17]. In the presence of a magnetic field, experiments have shown that randomly distributed magnetic particles exhibit less stiffness variation than anisotropic MREs [18]. Also, it was found that the volume fraction of the magnetic particles could be varied to find the optimum value for the maximum stiffness increase in soft manipulators with the introduction of the magnetic field [19].

Particle concentration and MREs soft robots compliance were shown to be linearly inversely related [20]. Therefore, the design of soft robotic MREs requires a trade-off between stiffness amplification and desired conformance. Magnetic particle alignment and the degree of crosslinking affect the deflection and mechanical properties of MRE soft actuators [21, 22]. To see how well a magnetic soft actuator held up under different magnetic fields and filler concentrations, the gripper was developed and tested [22]. The results of experiments revealed the direct relationship of the soft gripper bending and the magnetic filler concentration up to a certain level of the magnetic field, 80 mT. After this level, the bending of the gripper remained quite stable. The gripping force of the magnetic soft gripper, however, was reported to be quite low, around 0.1 N, in response to magnetic field of 80 mT.

To manage stress concentration and adhesion in deformed robotic bodies, MREs have been used as sensing components in soft robotics. Highly stretchable MREs made from a mixture of Ecoflex 00-10 (Smooth-On, PA, USA) and iron powder was integrated into a soft pneumatic actuator as an inductive coil-based wireless sensing mechanism to measure external loads and internal pressure in the precision of micrometres' deformation of the soft robot body [23]. The developed MREs sensor showed the capability of measurement of actuator's deformation with no hysteresis caused by either internal pressure or external contacts. Liquid metal-filled MRE

was also developed as a sensing module exhibiting maximum resistivity in the rest state while it is dropping drastically under the mechanical deformations [24].

Electro-Magnetorheological Elastomers (EMREs) in Robotic 4D Printing

In comparison to other soft robotics research, the EMREs have received significantly less attention. Composites of metal and/or magnetic particles and elastomers having electro-magnetic properties are widely used to build soft actuators based on EMREs principles, which respond to electrical stimulation in the presence of a magnetic field.

In research of soft robotics, a Ni nanowire/silicone nanocomposite actuator with electro-magnetic properties was tested in response to the magnetic force direction [25]. Increased concentrations of metal particles, according to the findings, led to greater actuation strain due to improved electrical responsiveness [25]. In addition, the strain response of EMREs soft actuators dropped with reasonable phase lag when the input voltage frequency was increased [25]. The EMRE soft actuators have demonstrated manageable hysteresis [25]. However, they do not respond effectively to the low voltage in high frequencies [26]. Active vibration control of soft robots might potentially benefit from the EMREs soft actuators. It is, however, the inherent time delays in these systems must be addressed.

Soft robots that use electrorheological fluids with dispersed magnetic particles and electric and magnetic fields for actuation modulation are also developed [27, 28]. MR fluid was injected into the 3D-printed hollow struts in a field responsive 3D-printed metamaterial architecture (Figure 1.b) [29]. The product displayed dynamically regulated movement under an applied magnetic field. Slightly quicker soft robots, however, still face hurdles when using low viscosity electrorheological fluids. With EMREs in soft robotics, sedimentation and homogeneous redistribution of magnetic particles are also significant practical difficulties.

Magnetic-based Shape Memory Polymers in Robotic 4D Printing

There has been a recent surge in the use of SMPs in the manufacturing of soft robotics. Heat-responsive SMPs [30, 31] have been the focus of the bulk of studies, although this is not always ideal since it is difficult to induce local controlled deformation via the thermal environment. As a result, it is critical to have SMPs that are acceptably sensitive to stimulus and have more local controllability. There has been more recent research on the creation of magnetic-responsive SMPs in 4D printing of soft robots [7, 32].

With its capacity to regulate signal direction and strength, as well as its simplicity in penetration into polymers, magnetic stimulation was a good choice for polymer and hydrogel-based soft robotics [33]. Medicinal robots, such as crawling and swimming ones, have profited from such remote and high-frequency control capabilities [34].

In biomedical applications, such as catheters, an SMP-based magnetically actuated variable stiffness soft robot [34] with simultaneous magnetic actuation and photothermal heating is practical [35]. In order to lower the modulus of SMP and enable actuation with applied magnetic fields, photothermal heating triggered by an LED was employed to heat the magnetic particles implanted in the material. In order to make the magnetic SMP sheet, thermoplastic polyurethane polymers were used. The LED and magnetic field are utilised to regulate the stiffness of SMP. In the absence of a magnetic field, the actuator is reshaped into its final state by simply turning the light back on.

To generate motion in polymer-based soft robots, magnetic fillers are often patterned onto an elastomeric layer. Applying a magnetic field aligns the fillers resulting in a variety of actuation modes, including bending and twisting. Despite the current application of magnetic soft robots, there are certain drawbacks to their use since the coils they use to generate external magnetic fields are heavy and demand a lot of power, making them burdensome in a small area.

Since magnetic micro and nanoparticles may be used to create new types of reconfigurable soft robots using a variety of manufacturing techniques, including casting, electrospinning, laser cutting, and 3D printing, this has opened the door to a new class of 4D printing magnetic-based soft robots [34, 36-38]. Polylactic acid (PLA) was 3D-printed as a magnetic responsive soft actuator with the addition of Fe_3O_4 nanoparticles [38]. The prepared ink was UV cured after extrusion from the printer nozzle. The developed soft robots showed a reversible remote actuation to the alternating magnetic field. Magnetic SMP soft robots also have a locking mechanism that allows for rapid and easy system reconfiguration (Fig. 1.c). To do so, two different types of filler particles were integrated to induce actuation and local heating independently [39].

Magnetic-based Hydrogels in Robotic 4D Printing

Another class of soft and elastic materials is hydrogels, which are hydrophilic polymers capable of absorbing large amounts of water. New materials for flexible polymer printing include hydrogels. Hydrogels may be used to make soft robotics devices provided their mechanical properties allow for actuation without rupturing the structures [40].

One of the hydrogel's most notable features is that it may be used in a variety of water-based applications. Because of their high viscosity and the range of printing methods available, hydrogels are well-suited for application in the 4D printing of soft robotics [41, 42]. Hydrogels that respond to stimuli like electrical and magnetic stimuli are increasingly used in biomedical soft robotics because their reversible shape-shifting may be swiftly activated. As a result of their high water content, hydrogels are useful for a wide range of biomedical applications, including drug delivery [43, 44] and tissue engineering [45] soft robots.

Hydrogel-based soft robots use volumetric changes in swelling and shrinkage as their primary actuation mechanism. Multiple methods exist, including bi-material [46] and multi-material [47, 48] composite, materials patterning [49], and functionally graded crosslinks [50] to achieve desired movements such as bending and twists. The polarity and amplitude of the external magnetic field control the actuation direction and bending deflection of the hydrogel in which magnetic nanoparticles (MNPs), such as Fe_2O_3 , Fe_3O_4 , and CoFe_2O_4 , are disseminated into the precursor hydrogel [40, 51]. Yet, in order to avoid particle agglomeration, the dispersion medium for nanoparticles must be sufficiently viscous [52]. Soft robots of this sort have found use in the pharmaceutical and medication delivery industry, where controlled and non-invasive release of medicinal dosages is possible (Fig. 1.b).

Microparticles of NdFeB alloy and fumed silica nanoparticles were mixed with a silicone rubber matrix for the preparation of auxetic type soft robotic hydrogel. To increase the printability of the ink via shear thinning as well as shape fidelity after extrusion the fumed silica was introduced as a rheological modifier. The prepared elastomer matrix was magnetized under the impulse magnetic field in the curation stage [7]. This study demonstrated the

application of magnetic responsive hydrogel soft robots with fast-moving metamaterial characteristics in pharmaceutical applications.

Direct 3D printing has lately been used to manufacture complex form magnetic-based robots with unique properties such as functionally graded and variable stiffness, which can be controlled remotely [7, 52-54].

An octopus soft robot was developed by dispersing ferromagnetic Fe_3O_4 nanoparticles in polyacrylamide (PAAm) [55] demonstrating a quick response to the magnetic stimulus (Fig. 1.c). Encapsulating Fe_3O_4 nanoparticles inside an alginate ionic hydrogel created a hydrogel soft actuator that could be manufactured in 4D [56]. The new actuator has been tested in aqueous soft robotics applications. Yet in practical applications, hydrogels' mechanical strength is the most important consideration. With nanocomposites inclusion, the mechanical strength of pure composites with electrical and magnetic fields was improved [57].

A 3D printing ink was created based on a combination of alginate, methylcellulose, polyacrylic acid (PAA), and Fe_3O_4 MNPs with the capability of developing convolutional geometry while being responsive to magnetic field [52]. The study has improved the rheological properties of the ink for direct 3D printing. Also, it was demonstrated that the infill pattern made via 3D printing led to a controlled bending of magnetic responsive soft hydrogel actuators.

Discussions of Smart Magnetic-based Robotic Materials in 4D Printing

MR elastomers differ from MR liquids in that they can transform morphology considerably in addition to their capacity to adjust viscosity [58]. A variety of novel properties, including 3D printability [59] and lower range viscosity, anisotropy for directional actuation [60], surface-modified magnetic particles for improved dispersion [61], novel polymer matrix for reduced cytotoxicity [62], and fiber reinforcement of MR elastomers for matrix enhancement and magnetic particle alignment [63] are investigated in MREs. These materials will enable previously unimaginable material properties tailored with additive manufacturing. Peristaltic pumps [64] and soft grippers [63] are only some of the recent examples of MREs [76]. All these applications will be further investigated using 4D printing of magnetic soft actuators used in soft robotics.

EMREs have been determined to be the least studied of all the electric and magnetic-responsive polymer soft robots evaluated in this research. As a result, further research into the uses of electrorheological fluids in soft robotics should focus on the size, dispersion, and kind of magnetic particles, as well as the optimal viscosity.

Using MREs, small-scale soft robots made of hard magnetic particles encapsulated in silicone were constructed functioning under very low Reynolds circumstances [5, 65, 66]. Untethered soft robotics have considerably profited from these features. The various additive manufacturing, including extrusion [7, 67] and voxel-level drop-on-demand 3D printing [68] were used to construct MREs soft robots.

4D-printed magnetic-based soft robots have issues due to their large magnetic field generator and significant energy consumption (Table 1). To lower the requisite field amplitude, small magnetic soft robots are preferable [69]. It is also critical that greater attention be paid to developing thin membranes with a low mass density that can deform rapidly at high speeds in these kinds of robots [70]. A combination of different stimuli, including electrical, pneumatic,

and magnetic [71, 72] response principles has been used to produce modern soft robots. As a consequence of the repulsive magnetic field, the soft robots have shown a significantly enhanced output stroke when activated at the resonance frequency.

When compared to MREs, magnetic SMP soft robots provide greater design freedom since they can be tuned for different glass transition temperature (T_g) values and conductive particle concentrations. Despite this, the locking mechanism of magnetic SMP soft robots is still sluggish. Developing autonomously navigating soft robots constructed of magnetic SMPs might be a promising new research area in the future [73, 74].

Further research on hydrogel soft actuators that are magnetic-based might be the future direction in the 4D printing of soft robots. Pioneering research on a freestanding liquid crystalline actuator manufactured from poly(acrylic acid) blocks to stabilize the newly created Fe_3O_4 magnetic microparticles was undertaken [75]. Hydro-swelling led to the formation of free-standing liquid crystalline ferrogels because the PAA absorbed a substantial quantity of water while the liquid crystalline domains remained dehydrated and undamaged. In the presence of a magnetic field, the produced ferrogel sample underwent rapid and reversible bending activation.

Hydrogel soft robots, especially in medical applications, have undergone a huge amount of study and accomplishment owing to their biocompatibility, biodegradability, transparency, and compliance. When it comes to fast and high force throughput in soft robotics, hydrogels have yet to demonstrate their full potential. Synthesis hydrogels that can be produced via 4D printing and generate more force are projected to increase their functioning in the present research areas.

The extrusion-based additive manufacturing of soft materials is a suitable way to print custom hydrogels and commercial silicones. Extrusion-based procedures enable a larger range of materials to be printed with high elongation percentages and heat tolerance, unlike photopolymerization processes and resin-based 3D printing approaches. It has been possible to produce heterogeneous composites with property variation by extruding polymers containing magnetically oriented additive particles [67].

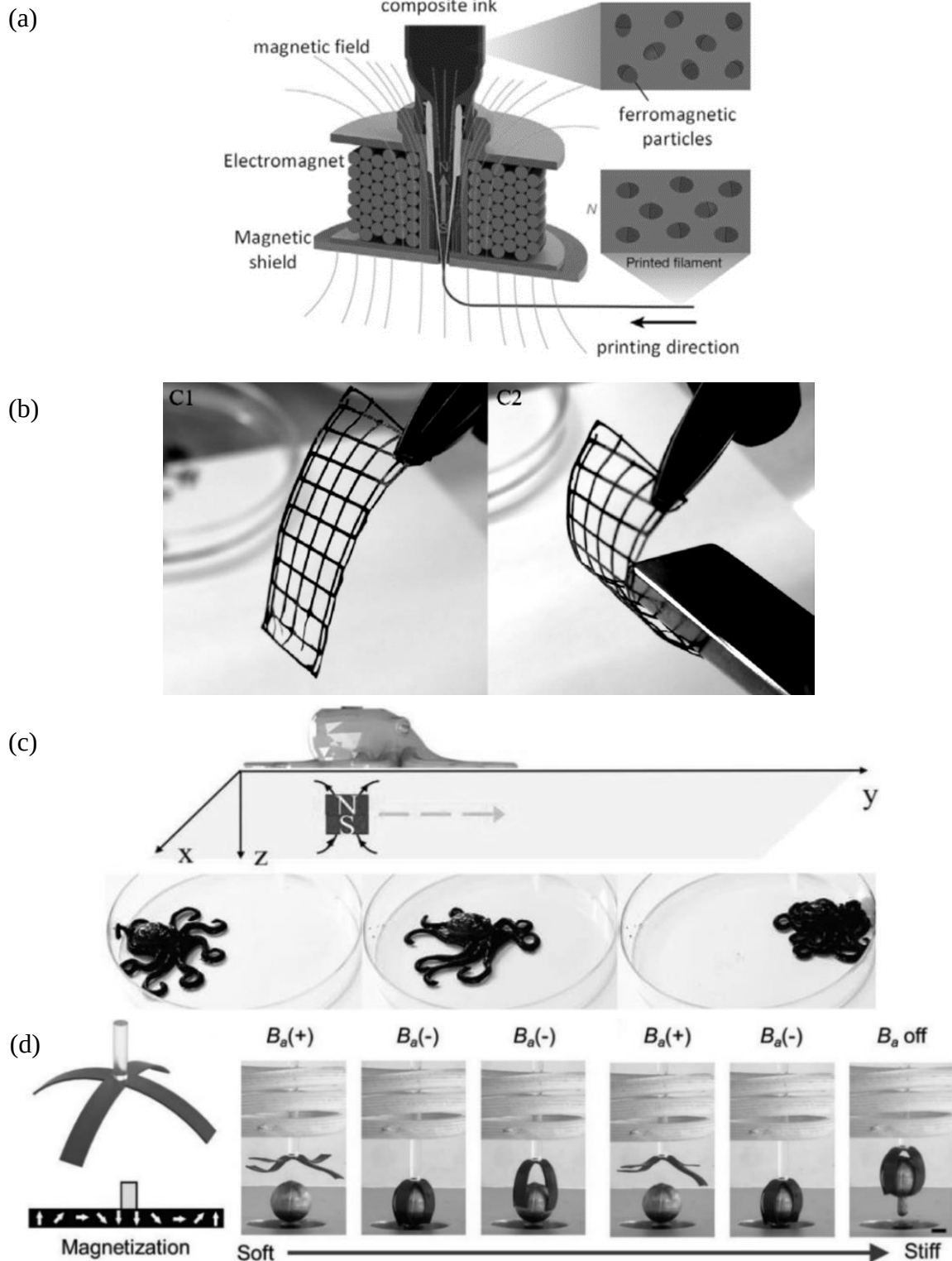


Fig. 1. (a) 4D printing of soft robots via magnetization (Reproduced with permission [7] Copyright 2018, Springer Nature) (b) the soft neodymium magnet hydrogel responsivity to the magnetic field that can be folded and twisted (Reproduced with permission [52] Copyright 2022, Elsevier) (c) A 4D-printed magnetic-responsive octopus hydrogel soft robot (Reproduced with permission [55] Copyright, 2022 Wiley). (d) A magnetic shape-locking SMP 4D-printed with and without locking mechanisms (left and right) (Reproduced with permission [39] Copyright, 2022 Wiley).

Table 1. Magnetic-based 4D printed-soft robots.

	MREs	EMREs	Magnetic SMPs	Magnetic Hydrogels
Response time	Low	Medium-High	Medium-High	High
Efficiency	Medium-High	Medium	Medium	Low-Medium
Strain	Low	Medium	Medium	High
Stress	Low-Medium	Medium	High	Low
3D/4D printing	[17, 67, 68, 70, 76]	NA	[38]	[7, 32, 50, 52, 53, 55]
Non-3D printing	[7, 14, 21]	[25, 27, 28]	[34, 39]	[69, 75]
Advantages	Remotely controlled	Little hysteresis	Easy process Locking	Low temperature Low operating voltage
Challenges	Low force External field interference	Sedimentation Redispersion	High voltage	Low strength

References

- Whitesides, G.M., *Soft robotics*. Angewandte Chemie International Edition, 2018. **57**(16): p. 4258-4273.
- Zolfagharian, A., et al., *Control-based 4D printing: Adaptive 4D-printed systems*. Applied Sciences, 2020. **10**(9): p. 3020.
- Zolfagharian, A., et al., *Evolution of 3D printed soft actuators*. Sensors and Actuators A: Physical, 2016. **250**: p. 258-272.
- Yarali, E., et al., *Magneto-/electro-responsive polymers toward manufacturing, characterization, and biomedical/soft robotic applications*. Applied Materials Today, 2022. **26**: p. 101306.
- Zhang, J. and E. Diller, *Untethered miniature soft robots: Modeling and design of a millimeter-scale swimming magnetic sheet*. Soft robotics, 2018. **5**(6): p. 761-776.
- Xu, L., T.C. Shyu, and N.A. Kotov, *Origami and kirigami nanocomposites*. Acs Nano, 2017. **11**(8): p. 7587-7599.
- Kim, Y., et al., *Printing ferromagnetic domains for untethered fast-transforming soft materials*. Nature, 2018. **558**(7709): p. 274-279.
- Chung, H.J., A.M. Parsons, and L. Zheng, *Magnetically controlled soft robotics utilizing elastomers and gels in actuation: A review*. Advanced Intelligent Systems, 2021. **3**(3): p. 2000186.
- Li, Y., et al., *Magnetic hydrogels and their potential biomedical applications*. Advanced Functional Materials, 2013. **23**(6): p. 660-672.
- Zhao, W., et al., *In situ synthesis of magnetic field-responsive hemicellulose hydrogels for drug delivery*. Biomacromolecules, 2015. **16**(8): p. 2522-2528.
- Evans, B.A., et al., *A highly tunable silicone-based magnetic elastomer with nanoscale homogeneity*. Journal of magnetism and magnetic materials, 2012. **324**(4): p. 501-507.
- Majidi, C. and R.J. Wood, *Tunable elastic stiffness with microconfined magnetorheological domains at low magnetic field*. Applied Physics Letters, 2010. **97**(16): p. 164104.
- Bastola, A., V. Hoang, and L. Li, *A novel hybrid magnetorheological elastomer developed by 3D printing*. Materials & Design, 2017. **114**: p. 391-397.

14. Li, X., et al., *A magneto-active soft gripper with adaptive and controllable motion*. Smart Materials and Structures, 2020.
15. Bastola, A.K., et al., *Recent progress of magnetorheological elastomers: a review*. Smart Materials and Structures, 2020.
16. Banerjee, H. and H. Ren, *Electromagnetically responsive soft-flexible robots and sensors for biomedical applications and impending challenges*, in *Electromagnetic Actuation and Sensing in Medical Robotics*. 2018, Springer. p. 43-72.
17. Xu, T., et al., *Millimeter-scale flexible robots with programmable three-dimensional magnetization and motions*. Science Robotics, 2019. **4**(29).
18. Ruddy, C., E. Ahearne, and G. Byrne, *A review of magnetorheological elastomers: properties and applications*. Advanced Manufacturing Science (AMS) Research. http://www.ucd.ie/mecheng/ams/news_items/Cillian%20Ruddy.pdf Accessed, 2012. **20**.
19. Zhang, X., et al., *Analysis and fabrication of patterned magnetorheological elastomers*. Smart Materials and Structures, 2008. **17**(4): p. 045001.
20. Fuhrer, R., et al., *Soft Iron/Silicon Composite Tubes for Magnetic Peristaltic Pumping: Frequency-Dependent Pressure and Volume Flow*. Advanced Functional Materials, 2013. **23**(31): p. 3845-3849.
21. Lee, M., et al., *Characterization of a magneto-active membrane actuator comprising hard magnetic particles with varying crosslinking degrees*. Materials & Design, 2020. **195**: p. 108921.
22. Tian, T. and M. Nakano, *Fabrication and characterisation of anisotropic magnetorheological elastomer with 45 iron particle alignment at various silicone oil concentrations*. Journal of Intelligent Material Systems and Structures, 2018. **29**(2): p. 151-159.
23. Wang, H., et al. *A wireless inductive sensing technology for soft pneumatic actuators using magnetorheological elastomers*. in *2019 2nd IEEE International Conference on Soft Robotics (RoboSoft)*. 2019. IEEE.
24. Yun, G., et al., *Liquid metal-filled magnetorheological elastomer with positive piezoconductivity*. Nature communications, 2019. **10**(1): p. 1-9.
25. Park, J.-M., et al., *Actuation of electrochemical, electro-magnetic, and electro-active actuators for carbon nanofiber and Ni nanowire reinforced polymer composites*. Composites Part B: Engineering, 2008. **39**(7-8): p. 1161-1169.
26. Zhu, M., et al., *Transient responses of magnetorheological elastomer and isolator under shear mode*. Smart Materials and Structures, 2019. **28**(4): p. 044002.
27. Bastola, A.K., L. Li, and M. Paudel, *A hybrid magnetorheological elastomer developed by encapsulation of magnetorheological fluid*. Journal of materials science, 2018. **53**(9): p. 7004-7016.
28. Sadeghi, A., L. Beccai, and B. Mazzolai. *Innovative soft robots based on electro-rheological fluids*. in *2012 IEEE/RSJ International Conference on Intelligent Robots and Systems*. 2012. IEEE.
29. Jackson, J.A., et al., *Field responsive mechanical metamaterials*. Science advances, 2018. **4**(12): p. eaau6419.
30. Bodaghi, M., A. Damanpack, and W. Liao, *Self-expanding/shrinking structures by 4D printing*. Smart Materials and Structures, 2016. **25**(10): p. 105034.
31. Zolfagharian, A., et al., *Pattern-driven 4D printing*. Sensors and Actuators A: Physical, 2018. **274**: p. 231-243.
32. Shao, L.-H., et al., *4D printing composite with electrically controlled local deformation*. Extreme Mechanics Letters, 2020: p. 100793.
33. Lum, G.Z., et al., *Shape-programmable magnetic soft matter*. Proceedings of the National Academy of Sciences, 2016. **113**(41): p. E6007-E6015.
34. Liu, J.A.-C., et al., *Photothermally and magnetically controlled reconfiguration of polymer composites for soft robotics*. Science advances, 2019. **5**(8): p. eaaw2897.

35. Chautems, C., et al. *A variable stiffness catheter controlled with an external magnetic field*. in *2017 IEEE/RSJ International Conference on Intelligent Robots and Systems (IROS)*. 2017. IEEE.
36. Zhang, F., et al., *Remote, fast actuation of programmable multiple shape memory composites by magnetic fields*. *Journal of Materials Chemistry C*, 2015. **3**(43): p. 11290-11293.
37. Mohr, R., et al., *Initiation of shape-memory effect by inductive heating of magnetic nanoparticles in thermoplastic polymers*. *Proceedings of the National Academy of Sciences*, 2006. **103**(10): p. 3540-3545.
38. Wei, H., et al., *Direct-write fabrication of 4D active shape-changing structures based on a shape memory polymer and its nanocomposite*. *ACS applied materials & interfaces*, 2017. **9**(1): p. 876-883.
39. Ze, Q., et al., *Magnetic shape memory polymers with integrated multifunctional shape manipulation*. *Advanced Materials*, 2020. **32**(4): p. 1906657.
40. Zolfagharian, A., et al. *3D printed hydrogel soft actuators*. in *2016 IEEE Region 10 Conference (TENCON)*. 2016. IEEE.
41. Shiblee, M.N.I., et al., *4D printing of shape-memory hydrogels for soft-robotic functions*. *Advanced Materials Technologies*, 2019. **4**(8): p. 1900071.
42. Zolfagharian, A., et al., *Polyelectrolyte soft actuators: 3D printed chitosan and cast gelatin*. *3D Printing and Additive Manufacturing*, 2018. **5**(2): p. 138-150.
43. Dong, Y., et al., *4D Printed Hydrogels: Fabrication, Materials, and Applications*. *Advanced Materials Technologies*, 2020: p. 2000034.
44. Shi, Q., et al., *Bioactuators based on stimulus-responsive hydrogels and their emerging biomedical applications*. *NPG Asia Materials*, 2019. **11**(1): p. 1-21.
45. Champeau, M., et al., *4D Printing of Hydrogels: A Review*. *Advanced Functional Materials*, 2020: p. 1910606.
46. Zolfagharian, A., et al., *Topology-optimized 4D printing of a soft actuator*. *Acta Mechanica Solida Sinica*, 2019: p. 1-13.
47. Zhai, Y., et al., *Printing Multi-Material Organic Haptic Actuators*. *Advanced Materials*, 2020: p. 2002541.
48. Zolfagharian, A., et al., *Effects of Topology Optimization in Multimaterial 3D Bioprinting of Soft Actuators*. *International Journal of Bioprinting*, 2020. **6**(2).
49. Erb, R.M., et al., *Self-shaping composites with programmable bioinspired microstructures*. *Nature communications*, 2013. **4**(1): p. 1-8.
50. Lin, C., et al., *4D-Printed Biodegradable and Remotely Controllable Shape Memory Occlusion Devices*. *Advanced Functional Materials*, 2019. **29**(51): p. 1906569.
51. Gao, W., et al., *Magnetic driving flowerlike soft platform: Biomimetic fabrication and external regulation*. *ACS applied materials & interfaces*, 2016. **8**(22): p. 14182-14189.
52. Podstawczyk, D., et al., *3D printed stimuli-responsive magnetic nanoparticle embedded alginate-methylcellulose hydrogel actuators*. *Additive Manufacturing*, 2020: p. 101275.
53. Kim, D., et al., *Untethered gripper-type hydrogel microrobot actuated by electric field and magnetic field*. *Smart Materials and Structures*, 2020.
54. Kim, Y., et al., *Ferromagnetic soft continuum robots*. *Science Robotics*, 2019. **4**(33).
55. Chen, Z., et al., *3D Printing of Multifunctional Hydrogels*. *Advanced Functional Materials*, 2019. **29**(20): p. 1900971.
56. McCracken, J.M., et al., *Ionic Hydrogels with Biomimetic 4D-Printed Mechanical Gradients: Models for Soft-Bodied Aquatic Organisms*. *Advanced Functional Materials*, 2019. **29**(28): p. 1806723.
57. Lee, J.H., et al., *Highly Tough, Biocompatible, and Magneto-Responsive Fe₃O₄/Laponite/PDMAAm Nanocomposite Hydrogels*. *Scientific reports*, 2019. **9**(1): p. 1-13.
58. Sun, S., et al., *Development of an isolator working with magnetorheological elastomers and fluids*. *Mechanical Systems and Signal Processing*, 2017. **83**: p. 371-384.

59. Qi, S., et al., *Versatile magnetorheological plastomer with 3D printability, switchable mechanics, shape memory, and self-healing capacity*. Composites Science and Technology, 2019. **183**: p. 107817.
60. Zhang, J., et al., *The magneto-mechanical properties of off-axis anisotropic magnetorheological elastomers*. Composites Science and Technology, 2020. **191**: p. 108079.
61. Zhou, Y., et al., *The fabrication and properties of magnetorheological elastomers employing bio-inspired dopamine modified carbonyl iron particles*. Smart Materials and Structures, 2020. **29**(5): p. 055005.
62. Cvek, M., et al., *Poly (2-oxazoline)-based magnetic hydrogels: Synthesis, performance and cytotoxicity*. Colloids and Surfaces B: Biointerfaces, 2020. **190**: p. 110912.
63. Shu, Q., et al., *High performance magnetorheological elastomers strengthened by perpendicularly interacted flax fiber and carbonyl iron chains*. Smart Materials and Structures, 2020. **29**(2): p. 025010.
64. Wu, C., et al., *Smart magnetorheological elastomer peristaltic pump*. Journal of Intelligent Material Systems and Structures, 2019. **30**(7): p. 1084-1093.
65. Wu, C., et al., *Numerical study of millimeter-scale magnetorheological elastomer robot for undulatory swimming*. Journal of Physics D: Applied Physics, 2020. **53**(23): p. 235402.
66. Hu, W., et al., *Small-scale soft-bodied robot with multimodal locomotion*. Nature, 2018. **554**(7690): p. 81-85.
67. Qi, S., et al., *3D printed shape-programmable magneto-active soft matter for biomimetic applications*. Composites Science and Technology, 2020. **188**: p. 107973.
68. Sundaram, S., et al., *Topology optimization and 3D printing of multimaterial magnetic actuators and displays*. Science advances, 2019. **5**(7): p. eaaw1160.
69. Gouda, S.R., et al., *Biodegradable Untethered Magnetic Hydrogel Milli-Grippers*. Advanced Functional Materials, 2020: p. 2004975.
70. Zhu, P., et al., *4D printing of complex structures with a fast response time to magnetic stimulus*. ACS applied materials & interfaces, 2018. **10**(42): p. 36435-36442.
71. Cao, C., X. Gao, and A. Conn, *A compliantly coupled dielectric elastomer actuator using magnetic repulsion*. Applied Physics Letters, 2019. **114**(1): p. 011904.
72. Liu, L., et al., *A biologically inspired artificial muscle based on fiber-reinforced and electropneumatic dielectric elastomers*. Smart Materials and Structures, 2017. **26**(8): p. 085018.
73. Ji, X., et al., *An autonomous untethered fast soft robotic insect driven by low-voltage dielectric elastomer actuators*. Science Robotics, 2019. **4**(37).
74. El-Atab, N., et al., *Soft Actuators for Soft Robotic Applications: A Review*. Advanced Intelligent Systems, 2020. **2**(10): p. 2000128.
75. Zhou, Y., et al., *Hierarchically structured free-standing hydrogels with liquid crystalline domains and magnetic nanoparticles as dual physical cross-linkers*. Journal of the American Chemical Society, 2012. **134**(3): p. 1630-1641.
76. Alapan, Y., et al., *Reprogrammable shape morphing of magnetic soft machines*. Science advances, 2020. **6**(38): p. eabc6414.