Mechanically-Guided 4D Printing of Magnetoresponsive Soft Materials across Different Length Scale

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Untethered magnetoresponsive soft materials that can transform between various complex 3D shapes are of growing interests in diverse areas such as soft robotics, flexible electronics, and biomedical engineering. Previous approaches are difficult to precisely encode continuous 3D magnetization profiles in small-scale 3D structures. Herein, a novel method to produce magnetoresponsive soft materials for shape-morphing systems across different length scale is reported. The magnetization profiles are programmed by the mechanically-guided 4D printing with high precision and time-cost efficiency. A theoretical model and model-based simulation method are developed to quantitatively predict shape-morphing of printed structures under applied magnetic fields, and guide the design of complex 3D shapes. A broad set of structures with complex transformations are fabricated to illustrate the capability of the methods. Diverse typical functional applications from centimeter- to millimeter-scale, including soft crawling robots, flexible grippers, bionic butterfly, and multistate magnetic switch, are further demonstrated.

1. Introduction

Shape-morphing systems responsive to heat \cite{1,2} solvent, \cite{3} pH, \cite{4} light, \cite{5}–\cite{7} electric, \cite{8} and magnetic fields \cite{9–12} are of great interests in recent years. They have widespread applications across different areas such as soft robotics, \cite{13–18} flexible electronics, \cite{9,19} metamaterials, \cite{17,20} and biomedical engineering. \cite{21} Among them, responsive soft materials composed of hard magnetic particles, \cite{20,22,23} especially small-scale materials, \cite{15,24,25} have shown its great potentiality. The magnetoresponsive materials can be manipulated quickly, reversibly, safely, and remotely under applied magnetic field without pneumatic or electrical tethers. \cite{12}

Magnetoresponsive properties of the soft materials stem from well-designed magnetic anisotropy and structural geometry. Different techniques have been used to encode the magnetic anisotropy \cite{9,15,17,20,22,24,26–32} (Table S1, Supporting Information). Many of previous programming approaches make magnetization after fabrication, which mostly rely on template- or other tool-assisted \cite{15,22,32} and laser-based magnetization. \cite{29,30} Template-assisted programming is hardly to be precisely and digitally controlled due to intricate template geometries. Laser-based methods are difficult to encode 3D structures, restricting shape-programming freedom. To overcome these limitations, other programming approaches are introduced by directly printing ferromagnetic domains in soft materials with 3D intricate geometries, \cite{9,28,33} offering great freedom for fabrication. In their work, the hard magnetic particles are magnetized before printing. The magnetization profiles are encoded during printing. Discrete 3D magnetization profiles are generated in the 3D structures. The different magnetic domains are printed separately, and usually physically attached to each other. Interfaces between the magnetic domains may lower down the mechanical properties of the structures.

Previous researches have paved a critical foundation for the fabrication of magnetoresponsive soft materials. However, the ability to precisely encode continuous 3D magnetization profiles in 3D structures across different length scale without any template or mold has not been shown. Especially, creating small-scale materials with programmed magnetic anisotropy is challenging, which is significant for applications in microfactories \cite{13,24} bioengineering, \cite{14} and healthcare. \cite{25} To date, small-scale magnetic materials below centimeter size are mostly 2D sheets \cite{15,24}. The difficulty and cost rise remarkably with decreased size, increased magnetization, and structure complexity, possessing a limitation on further applications.

Meanwhile, 4D printing has led to an era of creation of advanced materials with tunable functionality, \cite{8} such as shape memory polymers (SMPs), \cite{2,21,35,36} liquid crystal elastomers (LCEs), \cite{17} hydrogels, \cite{18,38} ceramics, \cite{39} and other composites. \cite{24,32,40} Herein, we report a novel method to precisely program the magnetic anisotropy in 3D structures from centimeter- to millimeter-scale. Direct ink writing (DIW) technique is used to print structures with intricate 3D geometries. Programming of the magnetization profiles is realized by the mechanically-guided 4D printing.
Comparison of our method with major existing strategies to encode hard magnetic particles is shown in Table S1, Supporting Information. Using our method, a broad set of structures are fabricated with complex magnetization profiles across different length scale. We further present a model-based simulation method and typical applications of the magnetic materials from centimeter-to millimeter-scale.

2. Results and Discussion

Our composite magnetic ink for 3D printing (Figure 1a) is composed of a polydimethylsiloxane (PDMS) matrix containing cross-linking agent and catalyst. The fumed silica nanoparticles (Figure S1a, Supporting Information) are added as a rheological modifier to make the ink have the shear thinning and shear yielding behaviors required for DIW (Figure S2, Supporting Information). These properties ensure that the composite ink can be smoothly squeezed out of a nozzle through an external air pressure and maintain its shape during the printing process.[43] Magnetizable NdFeB particles (Figure S1b, Supporting Information) are added to make the composites have the magnetic response. After being printed, the soft composites embedded with nonmagnetized NdFeB particles are cured in a vacuum oven. The NdFeB particles will become magnetic with the magnetic pole direction consistent with the pulsed magnetic field when the materials are magnetized (Figure 1a). Scanning electron microscope (SEM) characterization demonstrates that distribution of the NdFeB particles within the printed objects is uniform (Figure S3, Supporting Information).

4D printing of magnetoresponsive soft materials is realized by a DIW-assembly-magnetization method (Figure 1b). First, an elastomer substrate and the nonmagnetized soft material were printed by DIW. The substrate was stretched by a homemade stretch device. Then the nonmagnetized soft material was attached to the prestretched substrate via a series of well-designed joints. Positions of the joints can be accurately controlled by 3D printing. 4D-printed structure with complex 3D shape can be obtained when the prestrain in the substrate is released.[42,43] The resultant deformed structure of the nonmagnetized soft material is determined by the prestrain in the substrate, positions of the printed joints, and the microstructures of the nonmagnetized soft material. Afterward, the 4D-printed structure was magnetized to saturation under an impulse field (about 3.25 T). The magnetized soft material would revert to its original shape when the joints connecting the substrate and the magnetized material are removed (Figure 1b). Using this approach, a nonuniform and continuously varying 3D magnetization profile was generated in the 3D structures. The hard magnetic particles embedded in the soft composite would induce magnetic moments and create stresses under applied magnetic field, leading to a macroscopic deformation behavior of the magnetized soft materials (Figure 1b). To demonstrate the ability of our method for fabricating magnetoresponsive soft materials, experimental results of an example are shown in Figure 1c. The magnetized elastic sheet as produced rapidly transformed into a curved shape from a flat configuration in 0.2 s under an applied uniform magnetic field (about 200 mT). The transformation is reversible and can be repeated on demand (Movie S1, Supporting Information).

To explore the influence of magnetic particles content on the magnetoresponsive performance of the magnetized soft material, samples were printed with different mass fractions of NdFeB particles. The magnetic moment density of the magnetized samples varies linearly from 20.77 to 49.07 kA m⁻³ as the NdFeB particle contents in the composite ink increase from 17.2 to 38.5 wt% (Figure 1d and Table S3, Supporting Information). To achieve better magnetoresponsive performance, content of the NdFeB particles in the composite ink is chosen as 38.5 wt%. Tensile tests were performed to evaluate the mechanical properties of the magnetized soft material, which can be stretched to more than three times of its initial length (Figure 1e). Excellent stretchability may enable the soft material to be used in some extreme conditions and environments.

In the model we concern, the residual magnetization density of the saturated magnetized materials is assumed to be a constant because the applied actuating field is much smaller than the coercivity of the embedded NdFeB particles (Figure S5, Supporting Information). Meanwhile, we also assume that the presence of the magnetic particles will not perturb the applied magnetic field as the permeability of the fully saturated hard magnetic particles and polymer matrix is approximately the same as that of air.[44,45] Constitutive law of the ideal hard magnetic soft material proposed by Zhao’s group[46,47] is adopted here, which gives the Cauchy stress tensor induced by the applied magnetic field as \( \sigma_{\text{magnetic}} = -J^{-1}B \otimes FM \). Here, \( J, B, F, M \) denote the volumetric Jacobian, applied external magnetic field, deformation gradient tensor, and magnetization vector in the stress-free configuration, respectively. \( \otimes \) refers to the dyadic product. The magnetization vector \( M \) in the stress-free configuration can be programmed by 4D printing, which can be calculated as \( M = F_0^{-1}M_0 \) (Figure S6, Supporting Information). The formula has considered influences of both the deformation and rotation on the magnetization vector in the stress-free configuration. Here, \( F_0 \) and \( M_0 = m[0 \ 0 \ 1] \) are the gradient tensor and magnetization vector in the 4D-printed configuration. Orientation of \( M_0 \) is same with the magnetization direction. The saturated magnetization \( m \) can be tuned by the mass fraction of the magnetic particles (Figure 1d). In our model, finite element simulations using the commercial software ABAQUS were first performed to explore the shape-morphing process occurred during 4D printing. The deformation gradient tensor of each element was then obtained. Using the commercial software MATLAB, the magnetization vector \( M \) for each element can be calculated. Afterward, the magnetization vector and the magnetic Cauchy stress tensor were implemented as a user-defined element subroutine in ABAQUS to simulate shape evolution of the magnetized soft materials under applied magnetic fields.

Based on the model, we can give quantitatively predictions of the shape evolution of the magnetoresponsive soft materials during 4D printing and magnetic actuation. To validate the applicability of our model, shape-morphing structures including bending and twisting configurations were designed and fabricated (Figure 2a–f). During 4D printing, the flat sheets and serpentine ribbons would experience buckling and twisting due to the compressive forces induced by the release of the prestrain in
the substrate. After being magnetized and removing the joints, the elastic sheets and serpentine ribbons could have magnetic response. Under applied magnetic fields, the elastic sheets and serpentine ribbons would transform into the wavy shapes and conical helices (Figure 2a–f and Movie S2, Supporting Information). The buckling wavelength of the elastic sheets can be controlled by the positions of joints. Wavy structures with uniform (Figure 2b) and gradient wavelength (Figure 2c) were

![Figure 1. 4D printing of magnetoresponsive soft materials via a DIW-assembly-magnetization method. a) Schematic of DIW and composition of the composite ink. b) Schematic of key steps for DIW-morphing-magnetization method. Magnetoresponsive soft materials with nonuniform, programmed and continuously varying 3D magnetization profiles can be fabricated using this method. Both the substrate and the nonmagnetized structure are used after being cured. Vectors M and B refer to the directions of magnetization and applied magnetic field. c) Experimental results of a magnetized elastic sheet as produced by our method. The flat magnetized sheet can reversibly transform into a curved shape in 0.2 s under applied magnetic field. The prestrain in substrate is about 10%. d) Effect of the mass fraction of magnetic particles in the ink on the magnetization of magnetized soft materials. e) Nominal tensile stress–strain curves of the magnetized soft materials with magnetic particle content being 38.5 wt% (A1, A2), 29.4 wt% (B1, B2), and 17.2 wt% (C1, C2). Magnetic soft materials demonstrated in (c) was printed with composite ink containing 38.5 wt% of NdFeB particles using a nozzle with a diameter of 410 μm. Substrate was printed with the SE1700 ink using a nozzle with a diameter of 250 μm. In this work, actuation of magnetized soft materials was performed by applied magnetic fields of about 200 mT generated by a permanent magnet. Geometric dimensions of the 3D-printed magnetic materials demonstrated in (c) are shown in Figure S4a, Supporting Information.](image-url)
Figure 2. A model to quantitatively predict shape evolution of magnetoresponsive soft materials under applied magnetic fields. (a–c) Finite element simulations and experimental results of an elastic sheet with a single (a), uniform (b), and gradient (c) wavelength designed by 4D printing and actuated by applied magnetic field. In (a–c), the prestrain of substrate along x axis is about 10%. (d–f) Finite element simulations and experimental results of conical helices with right-hand (d), left-hand (e), and mutative (f) chirality designed by 4D printing and actuated by applied magnetic field. The prestrain of substrate along x axis is about 40% in (d–f). (g) Quantitative comparison of the response configurations for an elastic sheet with uniform wavelength given by theoretical analysis, finite element simulations and experiments. All the demonstrated magnetic soft materials were printed with the composite ink containing 38.5 wt% of NdFeB particles using a nozzle with a diameter of 410 μm. The substrates were printed with the SE1700 ink using a nozzle with a diameter of 250 μm. Geometric dimensions of the 3D-printed magnetic materials are shown in Figure S4a (Supporting Information). Deformation of the buckled sheet under compressive field can be obtained as \[ w(x) = A \cos(kx), \] where \( A = \frac{2}{k} \sqrt{\frac{E \epsilon_{pre}}{E + h}}, \) and \( k = \frac{2\pi}{\lambda} \) are amplitude and wavenumber, and \( \lambda \) and \( \epsilon_{pre} \) are the buckling wavelength and prestrain in the substrate. Under hypothesis that the deformation is small, shape of the magnetized sheet under applied magnetic field can be obtained as \[ Z(x) = \frac{12mBx}{24mB + 3k^2E} \cos(kx). \] Here, \( B, E, \) and \( h \) are strength of the applied magnetic field, elastic modulus, and thickness of the elastic sheet, respectively. The equation reveals that in our method, response configuration of the magnetized sheet can be tuned by regulating the prestrain, positions of joints, saturated magnetization, and strength of the applied magnetic field. Comparisons of the response deformations given by theoretical analysis, simulations, and experiments are shown in Figure 2g, which shows good consistency. The relative error of the maximum deflection in Figure 2g between theoretical analysis and experiments is 3.65 ± 1.59%. Relative error of the maximum deflection between simulations and experiments is 2.18 ± 1.69%. The results demonstrate the capability of our model for quantitative predictions and precise encoding. Figure 2 shows that the model and simulations can be used to guide the design of magnetoresponsive soft materials with complex shape transformations.

A set of magnetoresponsive structures and their deformed shapes under applied magnetic field are shown in Figure 3 to demonstrate the versatility of our method. In Figure 3a, a bionic butterfly was designed by the model-based simulations and fabricated by 4D printing. The ferromagnetic domain in the butterfly was programmed by attaching it on a prestretched soft substrate. The butterfly would flap its wings under applied magnetic field (Movie S3, Supporting Information), as predicted by the model-based simulations, further validating the capability of
our model for guiding the design of responsive soft materials with complex shape transformations. Afterward, a classic topological structure, i.e., Miura-ori, which can be used as a basic unit to achieve origami structures with high complexity,[48] was designed and fabricated (Figure 3b). The Miura structure was printed with well-designed creases, which could lower local bending stiffness and enable facile folding deformation. The magnetization profile in the Miura was programmed by attaching it on a soft substrate with biaxial prestrains. Then the Miura structure would experience dynamic origami deformation in 0.6 s under applied magnetic field (Figure 3b and Movie S3, Supporting Information), which is consistent with the model-based prediction. It should be mentioned that the response speed of our magnetoresponsive structures is much faster than existing ones based on other stimulus-responsive materials such as liquid crystal elastomers[37] and shape memory polymers.[49] Our method would show its excellent time-cost efficiency when different response deformations are desired for a series of magnetic structures with same geometric design. In this case, large numbers of the magnetic structures can be printed with high efficiency in advance. Then each structure can be personalized programmed on demand. For example, as shown in Figure 3c,d, a cross-shaped structure and an annulus can exhibit disparate magnetoresponsive configurations under different designs by 4D printing. Equi-biaxial prestrain in substrate along x and y axes is 40%. Figure 3. 4D printing of various magnetoresponsive topological structures with complex shape transformations. a) A bionic butterfly designed by the model-based simulation and fabricated by 4D printing. Uniaxial prestrain in substrate along x axis is 50%. b) A Miura-ori structure designed by the model-based simulation and fabricated by 4D printing. The prestrains in substrate along x and y axes are 30% and 15%, respectively. c) A cross-shaped structure with distinct magnetoresponsive configurations under different designs by 4D printing. Equi-biaxial prestrain in substrate along x and y axes is 40%. d) An annulus with distinct magnetoresponsive configurations under different designs by 4D printing. Equi-biaxial prestrain in substrate along x and y axes is 30%. All of the demonstrated magnetic soft materials were printed with the composite ink containing 38.5 wt% of NdFeB particles using a nozzle with a diameter of 410 μm. The substrates were printed with the SE1700 ink using a nozzle with a diameter of 250 μm. Geometric dimensions of the 3D-printed magnetic materials are shown in Figure S4f–i, Supporting Information.
magnetic field when its four ends were attached on a pre-stretched substrate during 4D printing. Alternatively, if its midpoint was also tied, the magnetoresponsive configuration is a wavy structure. Similarly, the annulus could present undulating magnetoresponsive configurations with different wavenumbers. The results demonstrate the advantage of our method for manufacturing soft materials with complex magnetic responses.

In our method, adoption of the 4D printing technique enables us to produce small-scale magnetoresponsive materials with high encoding precision. Small-scale materials may have significant applications in biomedical engineering, such as construction of tissue scaffolds, drug delivery, and minimally invasive surgery.\textsuperscript{[14,15,25]} It is challenging to fabricate small-scale magnetoresponsive materials with complex construction. Using our method, a series of millimeter-scale structures with complex magnetoresponsive configurations (bending, twisting, jellyfish-like, and wavy structures) were printed and programmed, as shown in Figure 4 and Movie S4, Supporting Information. The printing resolution can reach 150 μm using the nozzle with the diameter being 110 μm (Figure S1f, Table S2, Supporting Information). Higher resolution may be realized by adopting finer nozzle and regulating the gas pressure and printing speed.\textsuperscript{[50]} Our method may find application on the fabrication and manipulation of a swarm of small-scale magnetic soft robots. Actuation performance of our 4D-printed magnetic materials can be evaluated by calculating the actuation rate and power density.

According to experimental and simulation data, the structures shown in Figure 2 and 4 deform up to strain levels from 0.09 to 0.25 within 0.1–0.6 s, providing a power density ranging from 2.3 to 103.1 kW m$^{-3}$. The actuation rate and power density of our 4D-printed magnetic materials are orders of magnitude greater than other untethered 3D-printed soft active materials such as hydrogels, liquid crystal elastomers, or shape memory polymers (Figure S8, Supporting Information). Speed and reversibility are challenges in conventional contactless 4D printing.\textsuperscript{[51]} The results demonstrate the advantage of our materials in realizing fast and reversible 4D printing.

The capability to create rapid, reversible, complex shape-morphing of magnetoresponsive materials ranging from centimeter- to millimeter-scale allows us to achieve diverse functions such as crawling robots, flexible grippers, biomimetic butterfly, and multistate magnetic switch, as shown in Figure 5. First, we designed an inchworm-like crawling robot whose locomotion is controlled by magnetic moments and differential friction forces (Figure 5a and Movie S5, Supporting Information). To achieve differential friction forces, a grid structure was printed and programmed to realize an inchworm-like magnetoresponsive configuration (Figure 5a). Distinguished contact area and angle would induce differential friction forces during the crawling locomotion when the magnetic field is applied. The deformed structure would crawl to the direction with low friction force when the external magnetic field is removed. Therefore, the

![Figure 4. 4D printing of small-scale magnetoresponsive materials. a) Small-scale structures with bending configuration under applied magnetic field. Uniaxial prestrain in substrate along x axis is 40%. b,c) Small-scale conical helixes with right-hand (b) and left-hand (c) chirality. Uniaxial prestrain in substrate along x axis is 40%. d) Small-scale jellyfish-like structures. Equi-biaxial prestrain in substrate along x and y axes is 40%. e) Small-scale wavy annulus. Equi-biaxial prestrain in substrate along x and y axis is 30%. All of the demonstrated magnetic soft materials were printed with the composite ink containing 38.5 wt% of NdFeB particles using a nozzle with a diameter of 250 μm. The substrates were printed with the SE 1700 ink using a nozzle with a diameter of 250 μm. Geometric dimensions of the 3D-printed magnetic materials are shown in Figure S4–n, Supporting Information.](image-url)
soft robot can achieve continuous crawling locomotion by repeatedly applying and removing the magnetic field (Figure 5a and Movie S5, Supporting Information). The capability to create small-scale responsive materials enables us to produce a millimeter-scale soft crawling robot that can also realize continuous crawling locomotion (Figure 5a and Movie S5, Supporting Information). The cross-shaped structure in Figure 3c with jellyfish-like responsive configuration can serve as a flexible magnetically controlled gripper, as shown in Figure 5b. The flexible gripper can rapidly grab and release a plastic pellet with a diameter of 20 mm by regulating the state of the applied magnetic field (Movie S6, Supporting Information). A marble (3.597 g) with a weight more than 10 times of the gripper (0.312 g) can also be grabbed (Figure S9 and Movie S6, Supporting Information). A millimeter-scale gripper that can precisely grab a small resin ball with a diameter of 1 mm is also produced here (Figure 5b and Movie S6, Supporting Information). Afterward, the bionic butterfly demonstrated in Figure 3a is shown to imitate the flight of some insects under circulating magnetic field (Figure 5c and Movie S7, Supporting Information). Finally, a multistate magnetic switch for electronics was fabricated to control the state of light-emitting diode (LED) lamps (Figure 5d). The magnetic switch is composed of a circuit board (Figure S10, Supporting Information) and a dual-wave magnetic structure. The state of the LED lamps can be accurately controlled via the applied magnetic field (Figure 5d and Movie S8, Supporting Information). The results suggest that this dual-wave structure may be used as a basic unit to form more complex control systems.

3. Conclusion

In summary, mechanically-guided 4D printing of magnetoresponsive soft materials for shape-morphing systems ranging from centimeter- to millimeter-scale is reported. The soft materials as produced can be responsive to both mechanical stimuli and magnetic field. Our methods can be extended to other material systems such as shape memory polymers or hydrogels to create multiple responsive materials. Multifunctional magnetic soft machines could be produced by using biomaterials or by incorporating cells to extend applications in biomedical engineering. Other 4D printing methods including light- or heat-driven methods can also be adopted to achieve programmable magnetization profiles. It should be mentioned that our method does not rest with the technique of 3D printing. By means of other printing techniques with higher resolution such as inkjet printing, two-photon polymerization lithography, and projection microstereolithography, other functional designs can be realized.

Figure 5. Functional demonstrations of the magnetoresponsive soft materials. a) Inchworm-like soft crawling robots. b) Flexible grippers. c) A bionic butterfly. d) A multistate magnetic switch. Geometric dimensions of the 3D-printed magnetic structures are shown in Figure S4f,m,o–r, Supporting Information.
magnetoresponsive soft materials of microscale, even nanoscale, can be expected. Reconfiguration of magnetic soft materials can also be realized by introducing phase change polymers.\textsuperscript{[29-31]} Our methods provide a novel way to create remotely, rapidly and reversibly controlled responsive soft materials across different length scale, and would suggest more possibilities for applications in soft robotics, artificial muscles, flexible electronics and biomedical engineering.

4. Experimental Section

PDMS matrix in the magnetic ink system was prepared first by mixing two silicone-based materials—SE 1700 (Dow Corning Corp.) and Ecoflex-10 Part B (Smooth-on Inc.) in a 1:2 mass ratio. Second, fumed silica nanoparticles (5 wt\% with respect to Ecoflex-10 Part B) were added into the matrix to achieve desired rheological properties. After being preliminary blended by a glass rod for 1 min, the matrix was added with NdFeB particles with an average size of 5 μm (LW-BA 16-7A, Xinnuode, 100 wt\% with respect to Ecoflex-10 Part B) and the SE 1700 base catalyst (10 wt\% with respect to SE 1700 base). The composite ink was then mixed thoroughly at 1200 rpm for 3 min, followed by vacuum degassing at 1500 rpm for 7 min by a planetary mixer (ITT-3005, Integrity). Finally, the ink mixture was poured into a printing syringe and centrifuged at 4000 rpm for 3 min to remove gas bubbles before 3D printing. The final concentrations of the ink system were as follows: 19.23 wt\% SE 1700 base, 38.46 wt\% Ecoflex-10 Part B, 1.92 wt\% fumed silica nanoparticles, 38.46 wt\% NdFeB particles, and 1.92 wt\% SE 1700 catalyst. Ink system for substrates and joints is composed of SE 1700 (Dow Corning Corp.), which was prepared by mixing the base and catalyst in a 10:1 weight ratio. The mixture was manually blended by a glass rod for 5 min. Then the ink was poured into a printing syringe and centrifuged at a centrifuge at 4000 rpm for 5 min to remove gas bubbles.

Rheological properties of the inks were measured using a rotational rheometer (AR2000EX; TA Instruments) with a 20 mm diameter stainless steel plate at 25 °C. Viscometry measurements were performed via steady-state flow experiments within shear rates from 0.1 to 1000 s\textsuperscript{-1}. Oscillation experiments were conducted to measure shear storage moduli at a fixed frequency of 1 Hz with a sweep of stress (10–10000 Pa). All the inks were configured at 25 °C in the first half hour of the test and cryopreservation to prevent inks from cross-linking.

3D printing of the magnetic materials, substrate, and joints was conducted by using a commercial 3D bioprinter (BioX, Cellink Company). Ink in the syringe was extruded through a nozzle (diameter, 410 or 250 μm) with a controlled gas pressure (150–500 kpa) and printing speed (10–40 mm s\textsuperscript{-1}). The gas pressure and printing speed were adjusted according to the liquid rheological properties, the nozzle diameter, and the dimensions of the printed structures. The printing parameters for the magnetic materials and substrate are shown in Table S2, Supporting Information. The printed magnetic structures were solidified in a vacuum drying oven at 100 °C for 5 h. The printed nonmagnetic substrates were solidified in a vacuum drying oven at 75 °C for 3 h.

Dimensions of printed fibers were measured using an optical microscope (Primotech, Carl Zeiss AG). Shapes and dimensions of the NdFeB and fumed silica particles were characterized using the SEM (Sigma300, Carl Zeiss AG). Distribution of the magnetic particles within the printed objects was characterized using SEM and EDS. The samples were printed with a size of 4 × 4 × 0.4 mm and coated with a thin gold film. All characterizations were performed at room temperature.

Magnetization was conducted under an applied impulse field (\textasciitilde 3.25 T) generated by a commercial magnetizer (PF-2000; Tiansh Magnetoelectric Technology Corp.). The magnetic moments of the magnetic materials were characterized using a Physical Property Measurement System (PPMS-Dynacool 9; Quantum Design) at a sweep of external magnetic fields from \textasciitilde 30 000 to 30 000 Oe. To prepare specimens, a cuboid structure (dimension: 10 × 10 × 4 mm; nozzle diameter, 410 μm; center-to-center ligament spacing, about 550 μm) was printed and cut into cylinders using a 3 mm biopsy punch (Miltec Inc.) to fit into the sample holders of the machine. All experiments were performed at 25 °C. The magnetic moment densities of the specimens were calculated by dividing its volume into the specimen’s remanent magnetization.

Tension tests of the printed structures were performed to obtain the mechanical properties using an INSTRON 5982 compound material testing machine at a displacement rate of 50 mm min\textsuperscript{-1}. Dimension of the magnetic samples was 60 × 10 × 1.2 mm (nozzle diameter, 410 μm; center-to-center ligament spacing, 550 μm). Dimension of the substrate samples was 60 × 10 × 2 mm (nozzle diameter, 250 μm; center-to-center ligament spacing, about 500 μm). Nominal stress–strain curves were plotted for both materials. The shear moduli of magnetic materials and substrates were obtained by fitting experimental curves to the neo-Hookean model (Figure 1f and Figure S2d, Supporting Information), which were about 110 and 169 kPa, respectively. The breaking elongations of the magnetic materials and substrates were about 241% and 220%.

Supporting Information

Supporting Information is available from the Wiley Online Library or from the author.

Acknowledgements

Y.Z. acknowledges the support from the National Natural Science Foundation of China (grant no. 11902114), the Natural Science Foundation of Hunan Province (grant no. 2020JJ5032), and Fundamental Research Funds for the Central Universities (grant no. 531118010231); C.J. acknowledges the support from National Science Foundation for Distinguished Young Scholars (grant no. 51725502).

Conflict of Interest

The authors declare no conflict of interest.

Author Contributions

H.Z. and Y.H. contributed equally to this work. Y.Z.: conceived the idea and designed the study. H.Z.: performed the experiments, theoretical analysis, analyzed the data, and produced the figures and movies. Y.H.: performed the theoretical analysis and model-based simulations. Y.W.: assisted the ink preparation and characterization. Y.Z. and C.J.: supervised the research, oversaw all research phases, and provided guidance to the research. All authors contributed to writing and discussion of the manuscript.

Data Availability Statement

The data that support the findings of this study are available from the corresponding author upon reasonable request.

Keywords

magneto-responsive soft materials, mechanically-guided 4D printing, shape-morphing, small-scale

Received: July 13, 2021
Revised: September 1, 2021
Published online: October 14, 2021
