Magnetic Arthropod Millirobots Fabricated by 3D-Printed Hydrogels

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Magnetically driven small-scale soft robots are promising for applications in biomedicine, due to their fast, programmable deformation, and remote, untethered actuation to accomplish complicated tasks. Although diverse materials and designs have been proposed for magnetic soft robots with programmable shape transformation, it is still challenging to produce strong actuation by a small magnetic field. Inspired by arthropod species, magnetic soft millirobots with joint structures by 3D printing hydrogels have been developed. The joints can turn the bending deformation into the folding deformation, with the jointed region deforming locally. Different from homogeneous bending deformation, such local deformation allows larger motions of robots and reduces the overall energy consumption at the same time. Through experiments and numerical simulations, it is shown that the magnetic arthropod millirobots are capable of performing multimodal locomotion and programmed shape transformation, such as move, flip, catch, carry, and release. Finally, ex vivo experiments of removing a foreign object from porcine organs (e.g., aorta, stomach, and intestine) are presented to demonstrate the potential surgery application of magnetic arthropod millirobots.

1. Introduction

Magnetomechanical small-scale soft robots have wide applications in minimally invasive medicine,[1] targeted cargo delivery,[2] and biomedical engineering,[3] because of their prominent ability in swift locomotion, fast response, and remote control.[3] Recently, hard-magnetic soft robots have realized complex deformations.[5] The interactions between the external magnetic field and the programmed magnetic domains of the materials induce body torques, leading to the deformation of the structure to align the magnetization with the external magnetic field.[5] Bending is a basic deformation mode for these shape-transforming structures. However, the homogeneous bending of the whole structure is not energy efficient, with every volume element deforming and storing energy.[7] A design strategy is needed to improve the performance of the magnetic soft robots.

For actuation, generating a large folding angle locally is always easier and more energy efficient than homogeneous bending. A successful example can be found in nature. Arthropod (e.g., crab, shrimp) refers to an invertebrate animal with an exoskeleton, a segmented body, and pairs of jointed appendages.[8] Compared to mollusks (e.g., snail, octopus), arthropods have multiple jointed limbs to perform complex movements. For instance, a snail moves its body by squirming (Figure 1a), while a crab moves faster through the movement of the limbs and can further grab objects with claws (Figure 1b). The crab claw functions through the exquisite structure of the limb joint: the driving force is generated by muscle contraction and transmitted through the joint to open or close the claw (Figure 1c).[8b] The joints connect skeleton systems and enable a large folding angle. Joints are also ubiquitous in more advanced mammals, and are widely used in hard robots nowadays,[9] It would be of great interest to introduce joints into the design of magnetic soft robots.

In this article, we have developed a series of bioinspired magnetic arthropod millirobots. The basic deforming element is a magnetic beam with a joint. The joint can turn the bending deformation into folding deformation, with the joint region deforming locally. As shown in Figure 1d, the jointed beam deforms into a V-shape (folding mode) instead of a U-shape (bending mode) for the beam without a joint. Obviously, the strain energy is stored more intensively and locally around the sharp angle in the V-shaped sample. Figure 1e indicates that the folding mode is more energy efficient than the bending mode since less strain energy is required for the folding mode to achieve the same bending angle as the bending mode.

Toward biomedical applications, hydrogels are preferable as the matrix material of magnetic robots for their good...
A few attempts have been made to achieve shape transformation of magnetic hydrogels (Table S1, Supporting Information). However, most magnetic hydrogels are embedded with soft magnetic nanoparticles (e.g., Fe$_3$O$_4$), and actuated by magnetic force or magnetothermal effect.[10] The shape transformations are either too simple or too slow. Recent works have shown that the magnetic torque generated by hard magnetic nanoparticles (NdFeB) is favorable for the rapid and complex shape transformation of magnetic soft robots[4k,5b]. Compared with other actuation strategies (e.g., pH, humidity, and temperature), magnetic torque-induced shape transformation is fast and reversible, which is especially preferable for in vivo operation of magnetic soft robots. With careful design of both their structure and magnetization, deformation of magnetic hydrogels can be programmed.

Here, we fabricate the magnetic arthropod millirobots using magnetic hydrogels printed by direct ink writing. The ink is composed of microgel-reinforced polyacrylamide (PAAm) hydrogel
precursor, nanoclay, and neodymium iron boron (NdFeB) microparticles (Figure 1f). We use the template-assisted magnetization strategy to program the magnetic domains of the millirobots.\[11\]

Under external magnetic fields, the millirobots are actuated to show programmed deformations. We have carried out numerical simulation to compare folding with bending quantitatively, and to investigate the effect of the joint geometry on the deformation of the structure. Various shape-changing structures have been realized with different magnetization profiles and distribution of joints. We have designed a magnetic arthropod millirobot that can accomplish complex tasks, such as move, flip, catch, carry, and release. We further conduct imaging-assisted foreign body removal experiments in porcine organs (e.g., aorta, stomach, and intestine) ex vivo, demonstrating that the magnetic arthropod millirobots can operate in harsh environments.

2. Results and Discussion

We fabricate magnetic soft structures with 3D printed magnetic hydrogels. The magnetic hydrogel is composed of acrylamide (AAm, monomer), N,N’-Methylenebis(acrylamide) (MBAA, crosslinker), 2,2’-Azobis(2-methylpropionitrile) (AIBN, thermal initiator), microgel powder, nanoclay (rheology modifier), and NdFeB microparticles (up to 50% of the total weight of AAm and deionized water). Hard-magnetic filler (NdFeB) is embedded into the hydrogel matrix to realize programmable complex deformation in response to the external magnetic field, because magnetized NdFeB microparticles can retain strong remanence after the magnetic field is removed.\[12\] The microgel is used to improve the mechanical properties of magnetic hydrogels due to the double-network topology.\[11\] To print magnetic hydrogels with direct ink writing (DIW), we add nanoclay (Laponite) to modify the rheology of the precursor. Both the microgels and nanoclays can increase the viscosity of the precursor, and prevent the NdFeB microparticles from settling down to the bottom of the container (Figure S1, Supporting Information). The ink is printable under appropriate concentrations of microgel and nanoclay, while the NdFeB content (0–50 wt% relative to the total weight of AAm and water) seems to hardly influence the ink printability (Figure S2, Supporting Information). The magnetic hysteresis loops of the magnetic hydrogel and NdFeB are compared in Figure S3, Supporting Information. The magnetic hydrogels show a larger hysteresis and higher coercivity than NdFeB microparticle, which may ascribe to that the entanglement of the polymer network hinders the movement of the magnetic particles. The higher coercivity of the magnetic hydrogel can allow a larger actuating magnetic field without disturbing the programmed magnetization of the material. More detailed information about the material preparation and printing procedure can be found in our previous study.\[11\]

We investigate the deformation of a jointed cantilever beam as a basic element. We differentiate the deformation mode of two kinds of beams: folding for the jointed beam and bending for the beam without joints. We fix one end of the cantilever beam, activate it with a magnetic field, and measure the bending angle of the beam. Figure 2a shows that the jointed beam has a larger bending angle than the beam without joint under the same applied magnetic field. This is because the jointed region has a smaller thickness and a lower bending stiffness, hence a larger deflection is induced under the same applied magnetic field. To predict the deformation of the beams, we have conducted finite element (FE) simulation based on recent theoretical works.\[6,14\] The results show that our simulation matches well with experiments. We plot the energy difference between the folding mode and bending mode. The strain energy for the folding mode is about half of the bending mode at the same bending angle (Figure 2b). The simulated deformed shapes of the beams with bending and folding mode are shown in Figure 2c,d, respectively. The details for the theory and simulation are in Supporting Information.

In the following, we quantitatively analyze the effect of the joint geometry on the deformation of the beams. The jointed beam always demonstrates the feasibility of the jointed structure to encode information about the material preparation and printing procedure.

In Figure 3a we attempt to extend the jointed beam to more complex structures and realize more advanced functions. The soft structures in Figure 3a are prepared with microgel (2 wt%), nanoclay (2 wt%), and NdFeB (20 wt%). The approach of template-assisted magnetization is used to program the magnetization profiles of the printed structures on demand.\[11\] The printed structure is deformed to a designed configuration, and then magnetized under a 1.6 T impulse magnetic field (Figure S4, Supporting Information). The local magnetization direction is denoted by green arrows in Figure 3. A mild magnetic field (<100 mT) is used to actuate the shape change of the structure, without disturbing the programmed magnetization. Magnetic torques will generate at the location where the magnetization is not aligned with the applied magnetic field, and force the body to change its shape.\[6,13\]

We prepare a strip with four joints and place it on the working plane with an arched shape. The arch collapses when applying a magnetic field perpendicular to the plane (Figure 3a). This example demonstrates the feasibility of the jointed structure to encode...
alternating magnetization profile into a small planar body, which is difficult for a structure without joints. Figure 3b shows a four-arm structure deforming into a “stool” shape under an external magnetic field. Another four-arm structure is illustrated in Figure 3c, where each arm has two segmented magnetic profiles with opposite magnetization directions. The structure deforms into a “box” shape, with two joints at each arm folding simultaneously. An origami “bird” hovers its wings in a time-varying manner.
We further design a six-armed structure with multiple modes of locomotion (Figure 4). The structure has a hexagonal main body with a diagonal length of 8 mm, and six triangular arms with 6 mm length from vertex to bottom edge. The magnetization profile of the millirobot is shown in the central scheme of Figure 4a, where the magnetization is downward for the arms and upward for the main body. A gradient magnetic field generated by a permanent magnet is used for the actuation and control of the millirobot. The millirobot has three configurations under different conditions: freestanding configuration when $B^\text{app}$ is zero, flat configuration when $B^\text{app}$ increases to 100 mT along the downward direction, and moving/catching configuration when $B^\text{app}$ increases to 100 mT along the upward direction (Figure 4a). The freestanding configuration is not flat, because the as-fabricated materials have residual deformation caused by the fabrication process. By switching the amplitude and direction of the applied magnetic field, we can control the millirobot to complete a series of tasks, including lie down, 180° flipping, move to the desired position, and catch and carry a ceramic ball (Figure 4b). Movie S2. Supporting Information, demonstrates the whole process that the six-armed millirobot flips side, approaches the ball, catches it, makes zig-zag movement with the ball, and finally releases the ball.

Next, we attempt to demonstrate a potential application of the six-armed arthropod millirobot in biomedicine: removal of foreign body. Since most in vivo surgeries cannot be observed directly, the medical imaging modalities are vital for communicating the operator and the millirobot with real-time control and feedback. Several medical imaging strategies can be used to track the magnetic millirobots (e.g., magnetic resonance imaging, X-ray imaging, and ultrasonography\cite{16}). For convenience, here we use an endoscope to provide optical images and guide the operation of the millirobots. The other medical imaging modalities may be more appropriate for applications where no camera vision is available.

We perform foreign body removal experiments using the six-armed millirobot in porcine organs ex vivo. The process can be divided into four movements: placement of the millirobot, magnetic navigation to the foreign body, catch and carry, and release (Figure 5a). The components used are shown in Figure 5b. A curly lead wire is used as an example of a foreign body, since it is not magnetic and will not be attracted by the magnet. A handheld endoscope is employed to capture the internal image and guide the operation of the millirobot. Before the foreign body experiment, we test the biocompatibility of the magnetic hydrogel in a live/dead assay of mouse embryonic fibroblasts. The result shows a 90% relative cell viability after 24 h culture (Figure 5c), indicating an acceptable level of toxicity of the surface of the magnetic hydrogel. Since NdFeB particles are toxic\cite{16} and PAAm hydrogels are nonbiodegradable, the robot needs to be retrieved via magnetic navigation after the experiment.

We demonstrate the use of the millirobot for foreign body removal in a porcine aorta (Figure 5d and Movie S3, Supporting Information). The millirobot is placed at the opening of the organ, moves toward the foreign body under the guidance of the magnet, deforms itself to catch the foreign body, and relocates the foreign body to the desired position. The process is completed within 10 s. The same procedure is performed in a porcine stomach (Figure 5e and Movie S3, Supporting Information).
Information), which is filled with water to provide sufficient operating space. The swelling of hydrogels can cause the decrease of mechanical properties (Figure S6, Supporting Information) and the loss of maneuverability of the millirobots. Thus, only short surgeries are appropriate with the millirobots to avoid excessive swelling of the hydrogels. The inner surface of the porcine stomach is undulating, with folds and creases. The millirobot can still easily grab the foreign body, and smoothly pass the folds and creases, and finally get out. The foreign body removal is also well demonstrated in a porcine intestine filled with water (Figure 5f and Movie S3, Supporting Information). Since the aforementioned experiments of foreign body removal are completed within a few minutes, no obvious swelling of the hydrogel is observed.

Though the magnetic arthropod millirobots have shown great capabilities of moving and grabbing inside porcine organs ex vivo, further study is needed towards in vivo applications. Considering the toxicity of NdFeB microparticles, a functional coating on the microparticles may be applied. Moreover, the used PAAm hydrogel matrix can be replaced by more biocompatible PEG- or PVA-based hydrogels. It is hoped that more functionalized hydrogels with enhanced biocompatibility and 3D-printability can be synthesized to fabricate magnetic millirobots with jointed structures.

### 3. Conclusions

We have proposed magnetic arthropod soft millirobots fabricated by 3D-printed hydrogels. Finite element simulations have been conducted on the deformation of jointed magnetic structures to guide the design of magnetic arthropod millirobots. The joints can induce folding of the beam, which is more energy efficient than homogeneous bending. We design a six-armed arthropod millirobot with multimodal locomotion for foreign body removal. It is noticed that the demonstration of millirobot for foreign body removal...
removal is still very preliminary, without any in vivo validation. More study is needed for the evaluation of the utility of the millirobot. We believe that the design concept of 3D-printed structures with joints can be extended to more magnetic soft robots.

4. Experimental Section

**Materials:** 2-Acrylamido-2-methyl-1-propanesulfonic acid sodium salt (NaAMPS; Sigma-Aldrich 655821), N,N′-Methylenebis(acrylamide) (MBAA; Aladdin E124733), Acrylamide (AAm; Aladdin A108467), 2,2′-Azobis(2-methylpropionitrile) (AIBN; Macklin A800354), α-ketoglutaric
Preparation of the Magnetic Hydrogel Ink: We followed the previous recipe to prepare microgels of PNaAMPS hydrogel. The hydrogel precursor was composed of deionized (DI) water, AAm (4 mol%), MBAA (0.001 mol% relative to AAm), and nanoclay (2 wt% relative to the total weight of AAm and DI water). The mixture was blended by magnetic stirring for 1–2 h to get a homogeneous solution. Then, we added microgels into the aforementioned solution at various concentrations (1–4 wt% relative to the total weight of AAm and DI water). The precursor with microgels was mixed using a planetary mixer (Thinxy ARE-300) at 2000 RPM for 2 min, and was stored at 2–8 °C for 2 days to attain full swelling of microgels. We further added NdFeB as magnetic particles (0–50 wt% relative to the total weight of AAm and DI water), and AIBN as the thermal initiator (0.5 wt% relative to the total weight of AAm and DI water) to obtain the ink of magnetic hydrogels. The ink was mixed with the planetary mixer at 2000 RPM for 2 min, and degassed for 30 s again.

Bending and Folding of Magnetic Hydrogel Beam: A custom-built six-coiled electromagnetic coil system (Hunan PaiSheng Technology Co., LTD, China) was used to actuate the beam (Figure S5, Supporting Information). The applied field direction was perpendicular to the magnetization direction of the material. For experimental results, the magnetic field strength was measured with a Gaussmeter and the bending angle was measured from the photo captured by a video camera (Cannon CI 890). The bending angle was defined as the angle between the horizontal line and the line connecting two ends of the beam. For simulation results shown in Figure 2, the gravitational force was calculated, and shear modulus and the residual magnetic flux density of the material were set to be 150 kPa and 3 mT, respectively.

3D Printing of the Magnetic Hydrogel: The DIW printer was converted from a MakerBot ReplicatorTM2X and an air pump system (OTS-800-30L and 3 mT, respectively.

Magnetical Characterization: We measured the hysteresis loop of the pure NdFeB particles and magnetic hydrogel (NdFeB content, 20 wt%) by a vibrating sample magnetometer (MicroMag 3900) under the maximum magnetic field of 16 000 Oe.

Magnetization and Actuation of the Magnetic Millirobots: We constrained the as-printed magnetic hydrogels into designed shapes with acrylic molds, and then magnetized the hydrogels under a large magnetic field of 1.6 T generated by a parallel-pole dipole electromagnet (Beijing S&T Tech. Inc. China, see Figure S5, Supporting Information). Shape transformations, the functions of the millirobot such as cargo grasping, transportation, and release were performed under a gradient magnetic field generated by a permanent magnet (Φ 50 mm × 30 mm). The magnetic field measured at the center of the magnet surface was 410 mT. The motion of the magnetic millirobots was recorded by a video camera.

In Vitro Biocompatibility Tests: Biocompatibility tests were conducted using magnetic hydrogel-conditioned medium for cell culture by National Natural Science Foundation of China (Nos. 12172272, 11702208, and 11820101001) and the National Key R&D Program of China (2017YFE0119800).

Conflict of Interest
The authors declare no conflict of interest.

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

Keywords
3D printing, arthropod, hydrogel, magnetic soft robots, shape change

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

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