

Hydrogel Thin-Film Magnetic Millirobots with Programmable Deformation and Shape Retention for Biomedical Applications

Weifeng Hu, Shirong Zheng, Seokjae Kim, Jiyun Nan, Tae-Il Kang, Minghui Nan, Junhyeok Lee, Taeksu Lee, Jong-Oh Park, Ki Joon Heo,* and Eunpyo Choi*

Soft millirobots capable of controlled deformation and rapid responsiveness are gaining prominence in biomedical applications, particularly for precise drug delivery. This study presents a novel triple-layered hydrogel thin-film magnetic millirobot (HTFMR), constructed from two non-toxic Parafilm layers and a biodegradable hydrogel infused with silica-coated NdFeB magnetic particles. The HTFMR exhibits complex shape-controllability and shape-retention properties, making it highly effective for cargo transport. Under programmed magnetic fields, its four-arm configuration enables efficient and secure grasping and cargo release at designated locations, with its shape-retention capability enhancing transport stability. In its four-claw configuration, the robot adheres firmly to surfaces, enabling targeted drug delivery to specific regions such as the gastric and intestinal mucosa. Cytotoxicity evaluations confirm the HTFMR's excellent biocompatibility. Furthermore, the robot is visible under X-ray imaging, and its precise movement and shape control within an electromagnetic actuation system demonstrate reliable performance under biologically simulated conditions.

While soft robots offer promising maneuverability and rapid responsiveness, their practical use within biological systems remains limited. A major constraint is that most are fabricated from elastomer-based materials,^[15–30] which often exhibit poor biocompatibility. Additionally, many designs incorporate conventional magnetic particles,^[16,17,31–35] which are inherently toxic, further hindering in vivo applications.

Numerous studies have developed thin-film robots with shape-retention capabilities that can grasp objects—such as small tomatoes—under external actuation, even maintaining their shape and the object's stability without continuous external stimulus.^[36,37] However, these shape-retaining robots typically require the combined use of multiple control fields, such as electromagnetic fields alongside photothermal effects, humidity, or ultrasound.^[36–39] In these systems, the magnetic field does not dominate the deformation process,

resulting in slow and indirect actuation. For instance, when relying on heating or humidity, the robot's response is delayed until environmental parameters—such as temperature or humidity—reach specific thresholds. The necessity for multiple, finely-tuned control fields makes it challenging to implement these systems in the confined and dynamic spaces typical of biological environments. This severely limits their practical utility for in vivo applications. Therefore, there is an urgent need for thin-film robots that offer fast responsiveness, simple and efficient control, and high performance under constrained conditions.

To address these challenges, this study introduces a hydrogel thin-film magnetic millirobot (HTFMR) composed of a biocompatible hydrogel embedded with silica-coated magnetic particles,^[40] encapsulated between two layers of parafilm (Figure 1a). The HTFMR exhibits not only high flexibility and rapid responsiveness to magnetic fields but also supports multiple shape-programming states and retains its deformed shape once actuated (Figure 1b). Unlike many existing systems that rely on multiple external stimuli,^[36,37] the HTFMR achieves this functionality—including repeatable shape control—using only a single magnetic control source. This advantage arises from the plastic deformation of the outer parafilm layers and the

1. Introduction

Soft millirobots, owing to their small size, flexible maneuverability, and excellent adaptability within biological environments, have found wide applications in the biomedical field. These include targeted drug delivery,^[1–3] capsule endoscopy,^[4–7] medical patches,^[8–12] and hyperthermia therapy,^[13,14] among others.

W. Hu, S. Kim, K. J. Heo
School of Mechanical Engineering
Chonnam National University
77 Yongbong-ro, Buk-gu, Gwangju 61186, Republic of Korea
E-mail: k.heo@jnu.ac.kr

W. Hu, S. Zheng, S. Kim, J. Nan, T.-I. Kang, M. Nan, J. Lee, T. Lee, J.-O. Park
Korea Institute of Medical Microrobotics
43-26 Cheomdangwagi-ro, Buk-gu, Gwangju 61011, Republic of Korea
E. Choi
Department of Mechanical Engineering
Sogang University
35, Baekbeom-ro, Mapo-gu, Seoul 04107, Republic of Korea
E-mail: echoi@sogang.ac.kr

The ORCID identification number(s) for the author(s) of this article can be found under <https://doi.org/10.1002/adfm.202510751>

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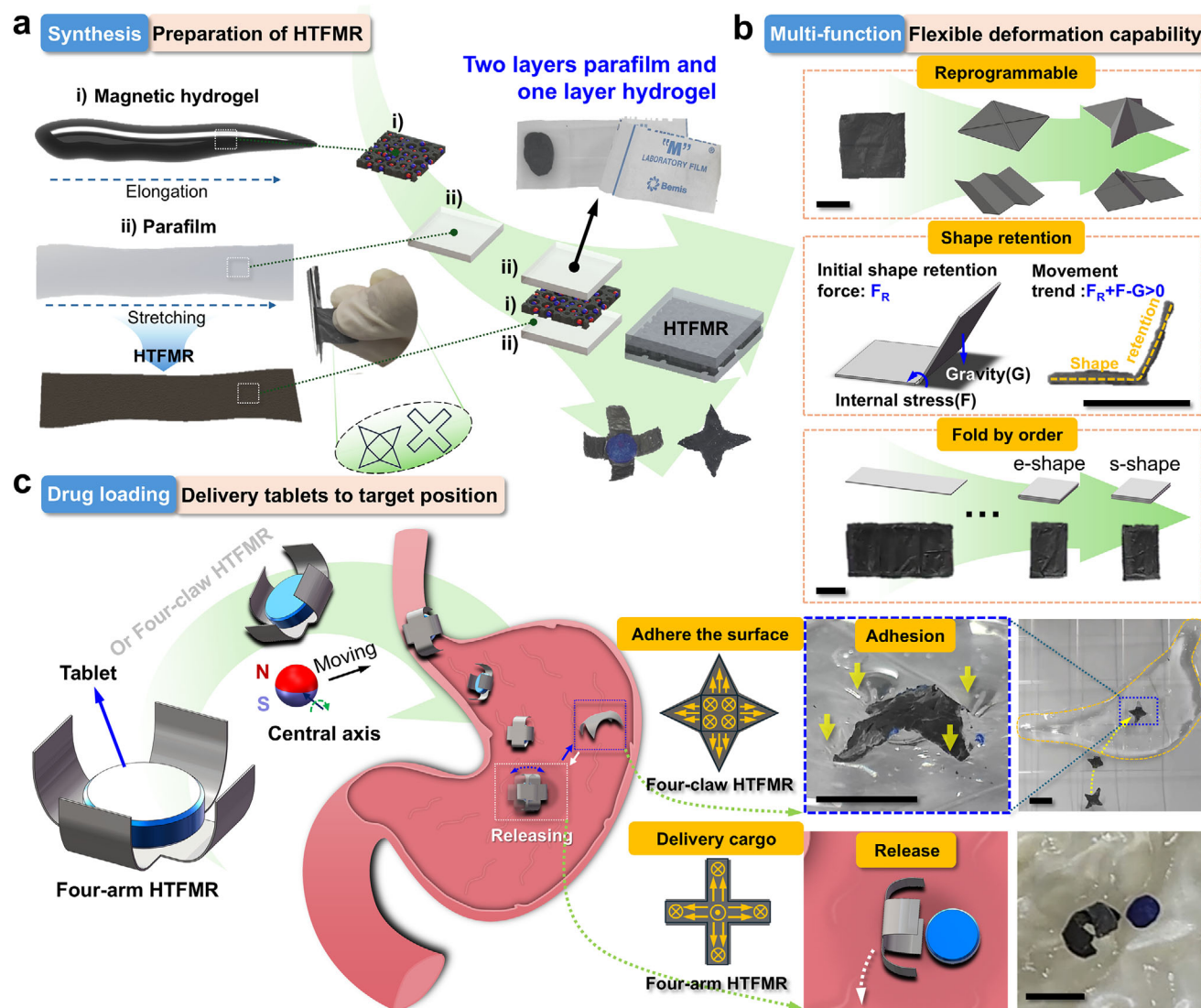


Figure 1. Schematic Illustration of HTFMR. a) Both the magnetic hydrogel and parafilm exhibit excellent ductility, demonstrating that HTFMRs can be fabricated into a variety of shapes. b,c) The reprogrammable, shape-retaining, flexible control, and cargo delivery and release functionalities of HTFMRs. (All scale bars are 10 mm).

material properties of the internal hydrogel during the shaping process, as discussed in Section 2.1. Cytotoxicity tests confirmed that the magnetic hydrogel is non-toxic to Raw264.7 cells. Moreover, Parafilm—used as the encapsulating material—is both non-toxic and biocompatible, with its safety and applicability supported by previous studies.^[41,42] Collectively, these properties confirm the HTFMR's in vivo safety. Compared with previously reported thin-film robots, the HTFMR offers improved performance and functionality (Tables S1 and S2, Supporting Information).^[3,8,9,36,37,43–46]

To explore the biomedical potential of HTFMRs, various geometries were fabricated and evaluated. A square-shaped HTFMR demonstrated the reprogrammability of internal magnetic particles, allowing it to adopt diverse target shapes under magnetic fields. A two-segment HTFMR validated its shape-

retention ability, while a three-segment variant showcased improved mobility and flexibility. Additionally, a four-arm HTFMR successfully performed controlled cargo delivery and release under an electromagnetic actuation (EMA) system. A four-claw HTFMR exhibited precise locomotion toward target sites and strong surface adhesion via magnetic force (Figure 1c). Therefore, this study explores a hydrogel thin-film magnetic millirobot (HTFMR) that integrates multi-modal actuation, post-actuation shape retention, and magnetic reprogrammability. These features not only ensure stable cargo retention during transport and enable directional adhesion for applications such as gastrointestinal drug delivery and mucosal patch deployment, but also provide the adaptability needed to perform sequential, multifunctional tasks—underscoring the HTFMR's strong potential for future clinical translation.

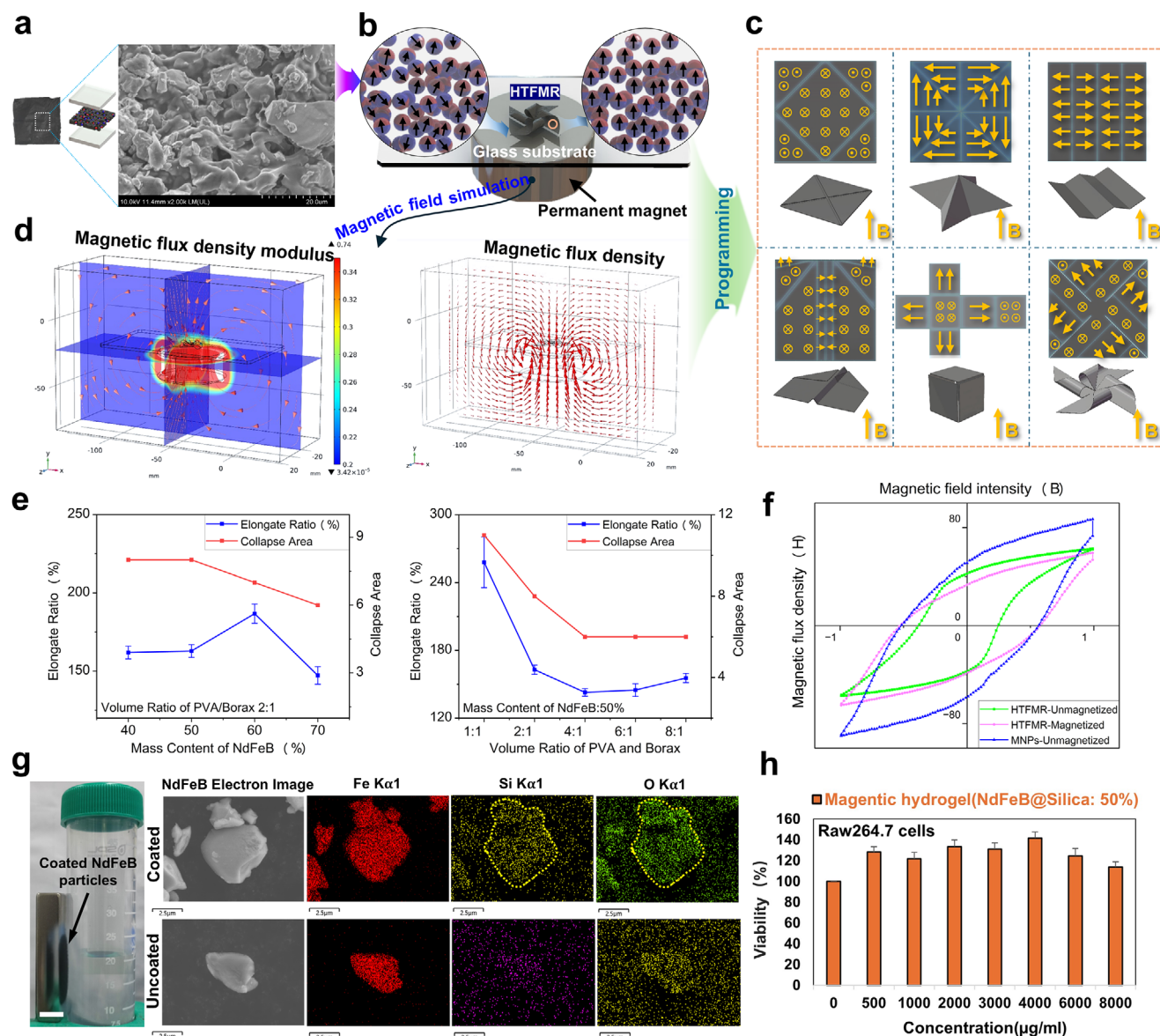


Figure 2. Magnetic field programming, hydrogel characterization, and cytotoxicity evaluation of HTFMRs. a, b) Programming of the HTFMR induces a transition in the arrangement of magnetic particles from a disordered to an ordered state. c) Schematic representation of the programmed magnetic field directions of internal magnetic particles in HTFMRs of different shapes. The orange arrows and the symbols “x” and “.” indicate the magnetization directions of the internal particles within the HTFMRs. d) Simulation of the magnetic field effects of permanent magnets using COMSOL Multiphysics. e) Collapse properties (after standing for 30 min) and elongation ratios exhibited by magnetic hydrogel with different internal compositions. f) VSM analysis comparing the magnetic properties of unmagnetized NdFeB particles, HTFMR, and magnetized HTFMR. g) Morphology and component analysis of coated and uncoated particles via SEM and EDX. Scale bar is 10 μm. h) Cytotoxicity assessment of magnetic hydrogel (NdFeB coated with silica: 50%) using Raw264.7 cells. Error bars represent one standard deviation, with $n = 3$ independent experiments.

2. Result and Discussion

2.1. Characterization of the Magnetic Hydrogel Inside the HTFMR

Figure S1 (Supporting Information) illustrates the fabrication process of the HTFMR. The magnetic hydrogel is synthesized via a crosslinking reaction and initially exhibits non-Newtonian fluid properties. Before encapsulated in parafilm, the internal structure of the magnetic hydrogel is examined using scanning elec-

tron microscopy (SEM), which revealed that the magnetic particles are tightly integrated within the hydrogel matrix (Figure 2a). In parallel, EDX was conducted to analyze the elemental distribution of Fe and Nd within the magnetic hydrogel. As shown in Figure S2 (Supporting Information), the results confirm a uniform dispersion of magnetic particles throughout the hydrogel network. The hydrogel matrix in our HTFMR system exhibits non-Newtonian, fluid-like behavior, allowing the embedded magnetic particles to remain unfixed and physically reorient under external magnetic torque. As a result, even after initial

magnetization, these particles can be reprogrammed by rotating their orientation—rather than reversing their polarity—using a strong external magnetic field (≈ 400 mT) during the programming process (Figure 2b). This process requires no heating or melting of the surrounding matrix to facilitate particle rotation, nor does it require exceeding the Curie temperature to reset the particles to a paramagnetic state for reprogramming (as shown in Table S3, Supporting Information). During subsequent actuation, a much weaker magnetic field (50–150 mT) is sufficient to drive deformation without triggering unwanted reprogramming, confirming the system's reliable programmability and magnetic responsiveness. This feature grants the HTFMR reprogrammable magnetic properties, allowing it to exhibit various deformation behaviors. By reprogramming the internal magnetic orientation and applying different external magnetic fields, the HTFMR can adopt diverse shapes. Based on this flexibility, we envision a single HTFMR capable of performing multiple functions within a single clinical procedure, thereby minimizing repeated invasiveness to the patient and enhancing adaptability in clinical workflows. Figure 2c shows how the magnetic pole orientation of internal particles corresponds to specific programmed shapes of the HTFMR, enabling rapid and targeted deformation in response to external stimuli. To further demonstrate the effect of magnetic field direction on HTFMR behavior, COMSOL Multiphysics simulations are used to model the magnetic field distribution of a permanent magnet and visualize the magnetic field lines interacting with the HTFMR (Figure 2d).

To evaluate the impact of magnetic particle concentration and the polyvinyl alcohol (PVA) to borax reaction ratio on the hydrogel's performance, two experimental series were conducted: 1) The mass of magnetic particles was held constant while varying the PVA-to-borax volume ratio. 2) The PVA-to-borax ratio was kept fixed while the magnetic particle content was altered. The maximum elongation ratio under a uniform external magnetic field was measured using top-down imaging. Additionally, the collapse ratio was assessed by allowing hydrogels (with equal volume but different compositions) to rest for 30 min in 14 concentric regions. The collapsed area was then analyzed to calculate the collapse rate (Figure S3, Supporting Information).

As the NdFeB particle content increased, the collapse ratio decreased—likely due to reduced water content within the hydrogel. Simultaneously, the elongation ratio increased steadily within the 40–60% NdFeB content range. However, at 70% content, a decrease in elongation was observed (Figure 2e), attributed to higher internal viscosity caused by decreased relative water content, which hinders deformation. Conversely, increasing the PVA-to-borax ratio significantly reduced both the collapse and elongation ratios. This is attributed to denser crosslinking, which produces a more compact hydrogel structure with lower porosity. These findings align with previous studies.^[47,48] A higher elongation ratio implies enhanced flexibility and responsiveness of the HTFMR, while a lower collapse ratio supports internal structural stability. Based on these results, the optimal formulation was determined to be 60% magnetic particle mass content and a PVA-to-borax volume ratio of 2:1.

The magnetic properties of unmagnetized particles, the unmagnetized HTFMR, and the magnetized HTFMR were analyzed using a vibrating sample magnetometer (VSM). All samples exhibited hard magnetic behavior, with the magnetized

HTFMR displaying higher coercivity than its unmagnetized counterpart (Figure 2f). This hard magnetic character ensures reliable retention of magnetic alignment during manipulation.

To improve biocompatibility for potential biomedical applications, NdFeB particles were coated with silicon dioxide. SEM imaging and energy-dispersive X-ray (EDX) spectroscopy confirm successful coating, as Si and O signals are detected on coated particle surfaces but not on uncoated ones (Figure 2g). Cytotoxicity testing using Raw264.7 cells demonstrated that magnetic hydrogel formulated with 50% silica-coated NdFeB particles exhibited no toxicity, even at concentrations as high as $8000 \mu\text{g mL}^{-1}$ (Figure 2h).

To further optimize HTFMR manipulation, the magnetic properties of the permanent magnet were characterized through COMSOL Multiphysics simulations (Figure S4, Supporting Information). Input parameters included the diameter (15 mm) and magnetic field strength (670 mT) of the spherical magnet (Figure S4b, Supporting Information). These simulations provided data on magnetic field strength and flux density in the vicinity of the HTFMR. The field strength within 15 mm from the magnet's surface gradually decreased from 600 to 150 mT (Figure S4c, Supporting Information). Experimental validation with a Teslometer showed slightly lower real-world values, with a steep decline observed within a 0–20 mm range. Consequently, an optimal operating distance of ≈ 10 mm—where field strength ranges from 50 to 150 mT—is recommended for effective HTFMR control. Distances shorter than this may lead to undesired reorientation of magnetic particles due to the hydrogel's internal fluidity, while longer distances may reduce responsiveness to magnetic actuation.

2.2. Preparation of the Differently Shaped HTFMRs

As shown in Figure 3a, the hydrogel thin film can be tailored into various target shapes for different application scenarios. When paired with precise programming of external magnetic fields, the HTFMR demonstrates highly versatile deformation capabilities. This enables the exploration of a wide range of functional applications. Through controlled magnetic field orientation, the HTFMR can reliably transform into diverse forms, making it suitable for tasks that demand adaptable structures.

The folding ability of the HTFMR was demonstrated through the construction of complex configurations—including an origami crane, cube, mimosa leaf mimic, and windmill (Figure 3b,c, Video S1, Supporting Information)—with the crane-folding task specifically designed to showcase multi-step programmed deformations inspired by origami choreography. To evaluate both response speed and retention of internal magnetic particle orientation after magnetization, the windmill-folding and unfolding cycle was repeated over 80 times. The fastest recorded folding time was 1 s, although response speed gradually decreased with repeated cycling. After 80 cycles, partial folding failure was observed, indicating mechanical fatigue. Nevertheless, the experiment highlights the HTFMR's rapid responsiveness and impressive mechanical stability under repeated actuation (Figure 3d; Video S1, Supporting Information).

The reprogrammable nature of the HTFMR was further validated by demonstrating deformation into multiple distinct

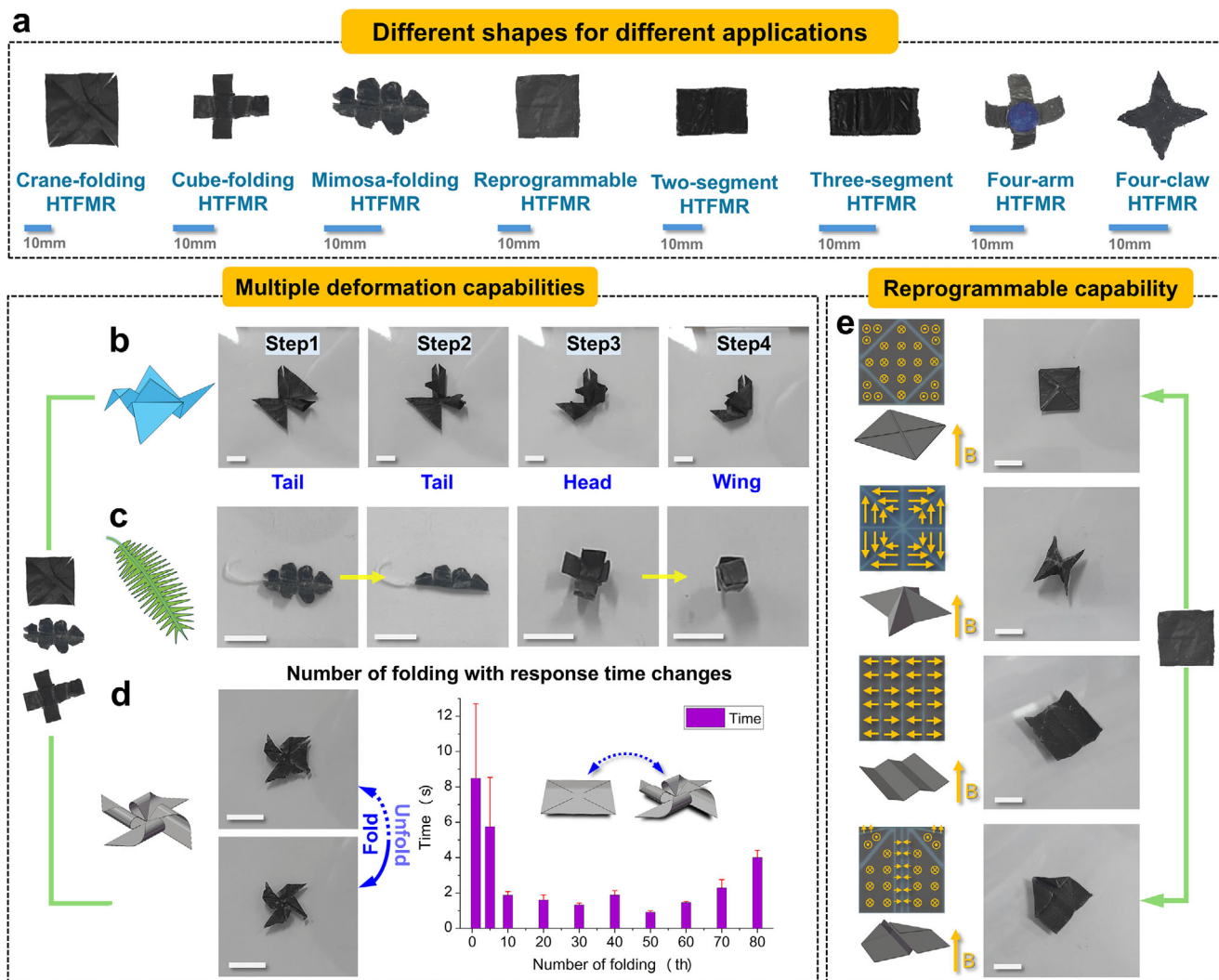


Figure 3. Programmable shape morphing and reprogrammable capabilities of HTFMRs. a) Different shapes of HTFMRs exhibit varying capabilities. b,c) Folding capabilities of HTFMRs to attain a paper crane, a mimosa, and a cube shape. d) The correlation between the frequency of complete unfold-to-fold cycles of the windmill and the resultant response time. Error bars represent one standard deviation, with $n = 3$ independent experiments. e) The same HTFMR demonstrates the ability to be reprogrammed to exhibit various final target shapes. (All scale bars are 10 mm).

shapes using a single device. By reorienting the internal magnetic particles through external magnetic field programming, the same HTFMR was successfully reconfigured into various shapes (Figure 3e; Video S1, Supporting Information). This confirms its capability for magnetic reprogramming and structural versatility. This feature holds strong potential for future clinical applications. For example, enabling a single robot to first travel long distances using one magnetization profile (e.g., rolling or undulating), and then perform mechanical functions such as gripping or expansion with a different profile. This multifunctional capability helps avoid task compromises and reduces the need for multiple device insertions, thereby minimizing procedural invasiveness and reducing patient burden. In addition, a two-segment HTFMR exhibited effective shape retention, maintaining its target configuration even after external magnetic fields were removed. This characteristic eliminates the need for continuous magnetic input, making it well-suited for in situ biomedical

applications. The three-segment HTFMR demonstrated exceptional flexibility and mobility, capable of executing various motion patterns, including crawling, rolling (on either long or short sides), and folding into “s” and “e” shapes.

Building upon these capabilities, two advanced configurations were explored: the four-arm HTFMR for cargo delivery and the four-claw HTFMR for medical patch deployment. Both designs exemplify the multifunctionality of the HTFMR platform and its potential for complex tasks in biomedical and soft robotics applications.

2.3. Shape-Retention Capability

To evaluate the shape-retention and deformation capabilities of the HTFMR, a two-segment configuration was fabricated. Both experimental tests and COMSOL Multiphysics simulations were

conducted to investigate the relationship between the robot's deformation behavior and the direction of the applied external magnetic field. Results show that to deform the HTFMR from a flat state to a 90° angle between the two segments, a minimum magnetic field strength of 90 mT is required (Figure 4a). During the simulation, when the magnetic field direction remains constant, the magnetic force acting on the HTFMR decreases in the latter stages of deformation due to the diminishing cosine component, as defined in Equation 1:

$$F = \nabla (m \cdot B) \cos \theta \quad (1)$$

where F represents the magnetic force, ∇ is the gradient operator, m is the magnetic moment of the material, B represents the magnetic field, and θ is the angle formed between the two HTFMR segments. When folding the HTFMR from 90° to a fully folded state, only 40 mT of magnetic field strength is necessary. A similar trend is observed in this phase—the response weakens as the folding angle increases (Figure 4a).

In practical applications using a permanent magnet, the direction of the magnetic field varies in real time. This variation means that the magnitude of deformation is not the sole determinant of the force applied to the HTFMR. Other influencing factors include the adhesion between the robot and the contact surface, as well as any surface irregularities. To remain within effective actuation parameters, the magnetic field strength should lie between 40 and 90 mT, requiring a magnet-to-robot distance of at least 10 mm (Figure S4, Supporting Information). In EMA system control, magnetic field strength can be calibrated more precisely, typically ranging from 50 to 80 mT. Within these parameters, the HTFMR is capable of smooth folding, unfolding, and shape deformation (Figure 4b; Video S2, Supporting Information).

To assess the HTFMR's ability to retain its shape after deformation, a permanent magnet was used to fold the two-segment HTFMR into five different configurations. Once the desired angle was achieved, the magnetic field was held for 20 s and then removed. A camera recorded the resulting angles, and measurements were repeated after 5 min. Each test was performed in triplicate, and angles were analyzed using AutoCAD 2018 to calculate the rate of angle change across trials. Results showed that the rebound angle decreased as the initial angle increased, and that angle variation also diminished over repeated trials (Figure S5a,b, Supporting Information).

The data suggest that internal stress within the HTFMR resists deformation, while gravitational force contributes additional influence. When the HTFMR is folded to angles below 90°, significant rebound occurs due to compression and buildup of internal stress at the hinge region. As the folding angle approaches 90°, this rebound becomes more pronounced. However, when the deformation angle exceeds 90°, gravity and internal stress begin to act in opposite directions. In this range, gravity helps counter internal stress, resulting in smaller rebound angles (Figure S5c, Supporting Information).

To further minimize rebound, a stress-relief method was developed, as shown in Figure 4c. The HTFMR is first fully folded and held under magnetic actuation, allowing internal stress to reach equilibrium in this large-angle state. After this preconditioning, the desired angle is set precisely. Experimental data confirm that

with this method, the rebound is limited to within 2° over a 5-min period, regardless of whether the angle is between 0–90° or 90–180°, demonstrating excellent shape retention.

Extended 24-h observation revealed that across the 90°–180° range, during the first hour, internal rebound stress caused noticeable angle changes. After 2 h, the internal stress gradually subsided, and gravity began to influence the system more significantly, leading to a compensatory reverse rebound. Ultimately, the total angular variation from the initial to final state was limited to just 2.32°. In contrast, across the 0°–90° range (Figure S6, Supporting Information), smaller variations (–2.78°) were observed initially, likely due to partial offsetting of gravity by reverse stress. In the later phase, deformation continued in the gravitational direction with reduced fluctuations (–4.42°).

To further demonstrate shape reversibility, a two-segment HTFMR was actuated to three predefined positions (Figure S7, Supporting Information). After removing the magnetic field, angular changes of 4.66°, 2.81°, and 5.68° were recorded at positions 1, 2, and 3, respectively. Clearly confirming its capability for reversible and programmable shape morphing without manual or thermal input. These results provide a strong foundation for further development of HTFMRs in precision-controlled biomedical applications, including targeted drug delivery, programmable cargo release, microrobotic transport, and the placement of medical patches.

2.4. Multi-Mode Motion Capability

A three-segment HTFMR with robust deformation performance and precise controllability was successfully fabricated, exhibiting a naturally elongated shape. Upon programming via an external magnetic field, two vertical internal creases uniformly divide the HTFMR into three segments. The magnetic particle orientations within each segment, as illustrated in Figure 5a, tend to align with the direction of the applied magnetic field. As a result, the HTFMR can easily undergo tri-folding in response to the magnetic field, effectively minimizing its footprint.

The transformation from a fully expanded state to an e-shaped configuration requires only four operational steps, while unfolding it back to its original shape requires an additional four steps. Building on the e-folded form, the HTFMR can be further folded into an s-shaped configuration in seven steps, with unfolding completed in three steps (Figure 5a; Video S3, Supporting Information). To further demonstrate the HTFMR's shape retention after magnetic actuation and its reconfigurability into diverse forms, one end of the HTFMR was fixed and subjected to magnetic field control. The HTFMR successfully folded into e- and s-shapes and subsequently returned to its flat configuration under magnetic guidance (Figure 5b-i,ii). Upon removal of the external magnetic field, the flat shape remained stable without noticeable deformation. Moreover, the HTFMR was reconfigured into alternative shapes such as a suspended S shape, a crawling posture, and a triangular form, all of which retained their configurations even in the absence of a magnetic field (Figure 5b-iii; Video S3, Supporting Information).

Experimental results confirm that the HTFMR can perform multiple locomotion modes, including both rolling and crawling. When manipulated by a rotating and shifting external

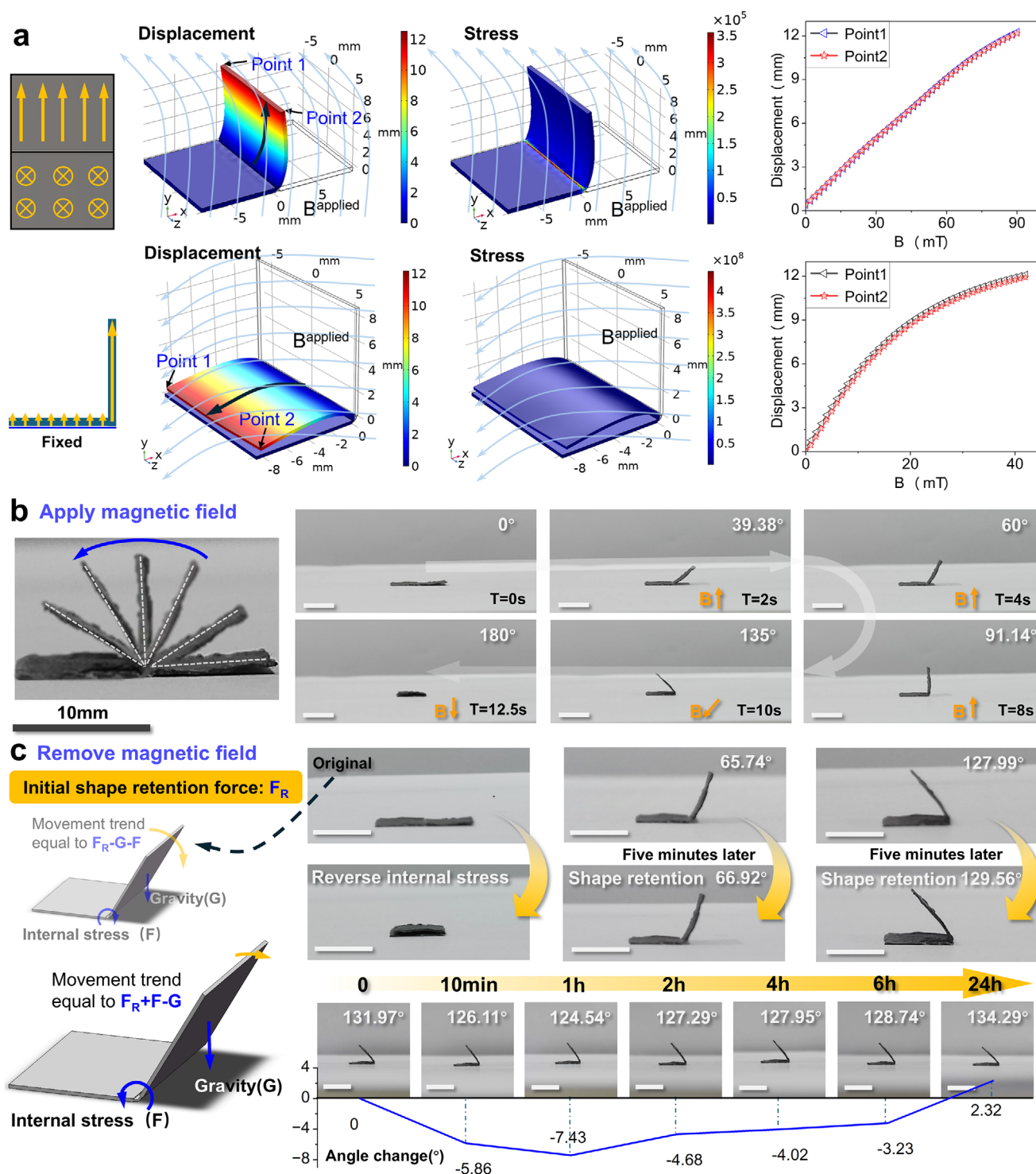


Figure 4. The two-segmented HTFMR. a) COMSOL Multiphysics simulations showing the displacement at the two terminal points at the top of the two-segment HTFMR. The light-colored arrow indicates the direction of the magnetic field. b) The complete transition from a fully expanded state to a fully folded state controlled by an external magnetic field. c) Improved shape retention capability of the two-segment HTFMR after internal stress reversal, with continuous recording over 24 h. (All scale bars are 10 mm).

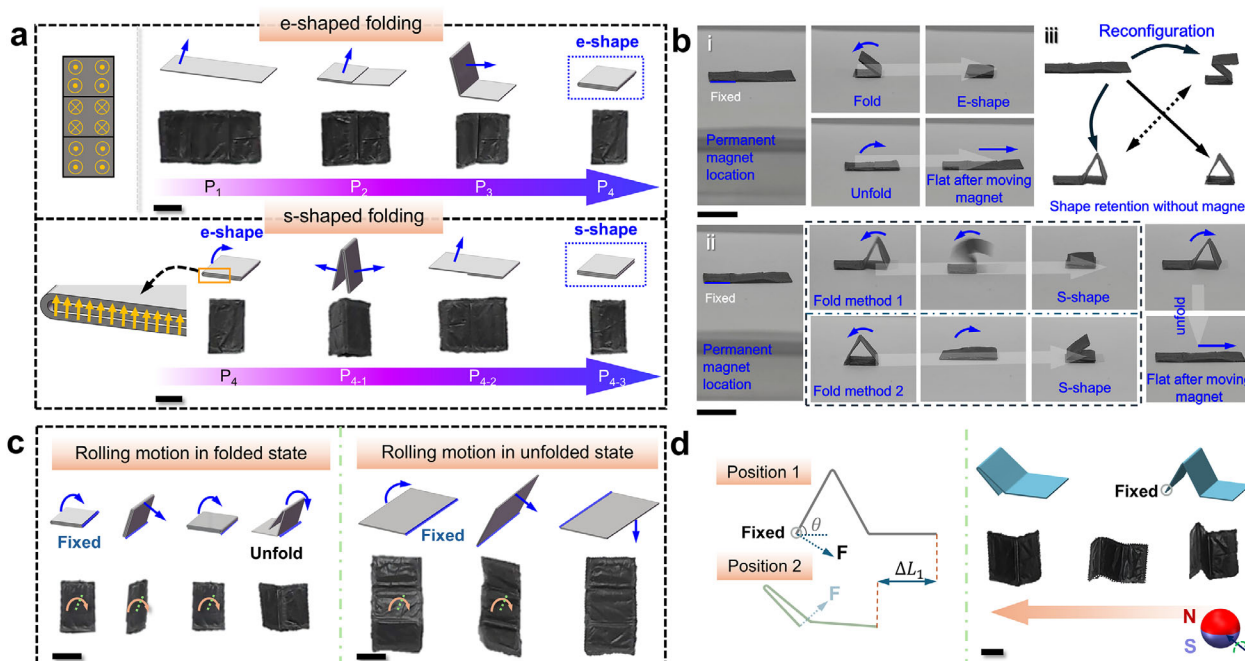


Figure 5. The three-segmented HTFMR. a) Two folding methods of the three-segmented HTFMR: e-shaped and s-shaped folding. “P1...P4” indicates positions in order. The small blue arrows represent the force on the robot from the external magnetic field. b) (i, ii) The three-segmented HTFMR recovers to a flat configuration after folding into E- and S-shapes, and maintains this flat shape even after the external magnetic field is removed. (iii) Reconfigurability of the three-segmented HTFMR: suspended S-shape, crawling posture, and triangular configuration. c) Two different rolling methods of HTFMR in the folded and unfolded states. d) Crawling motion characteristics of a three-segmented HTFMR. (All scale bars are 10 mm).

magnetic field, the HTFMR is capable of rotating around its edge and demonstrating high mobility. Specifically, when folded into an e-shape, it can roll along its short edge; when fully expanded, it rolls along its long edge. These modes enable swift and versatile movement across different configurations (Figure 5c; Video S3, Supporting Information).

In addition to rolling, the HTFMR exhibits crawling locomotion, mimicking the movement of an insect. During forward crawling, the tail segment anchors while the head extends forward, reducing its angle relative to the substrate. The head then stabilizes its position, and the external magnetic field reorients it to increase the angle with the horizontal plane. Simultaneously, the tail moves forward by a distance L , driven by the contractile force generated at the head. Repeating this cycle enables continuous forward motion (Figure 5d; Video S3, Supporting Information).

When a directional change is required, the HTFMR undergoes a six-step folding and flipping sequence, enabling it to turn around and proceed in the opposite direction (Figure S8 and Video S3, Supporting Information). This demonstrates the HTFMR's excellent manipulative adaptability and lays a strong foundation for future studies focused on the control and evolution of more complex shapes.

2.5. Multi-Task Capability in Multi-Scenario Application

Compared to earlier robotic designs, the four-arm HTFMR and four-claw HTFMR demonstrate enhanced end-effector functionality, significantly increasing their versatility in grasping and re-

leasing objects. This advancement stems from the HTFMR's shape-retention capabilities, which enable it to depart from traditional soft robotics and adapt to a wide range of application scenarios. As shown in Figure S9 (Supporting Information), when employed as a delivery robot, the four-arm HTFMR is mounted on a substrate containing cargo. Influenced by a magnetic field generated by a permanent magnet beneath the substrate, the HTFMR rolls forward and releases the cargo at the target location (Figure S9a and Video S4, Supporting Information). The shape-retention property ensures stable transportation, reducing the risk of cargo dropout while allowing for precise and controlled release. To further understand its magnetic actuation behavior, COMSOL simulations were performed to visualize the displacement of HTFMR arm tips under magnetic fields applied in two directions, along with displacement–field strength curves at selected points (Figure S9b, Supporting Information). In a grasping scenario, the four-arm HTFMR weighing 148.5 mg is attached to a movable shaft and tested for gripping capacity with gradually increasing payloads, as shown in Figure S10a (Supporting Information). When exposed to an external magnetic field, its four arms can be selectively opened and closed. Initially, the arms open to approach the cargo; reversing the field direction enables secure grasping. Once the cargo, placed on a substrate surface, is grasped, it remains securely held without requiring continuous magnetic input. Reversing the field again enables a quick release, the payload detached when attempting to grip a 474 mg object. Therefore, the maximum gripping capacity was recorded at 460 mg, which is more than three times the HTFMR's own weight (Figure S10b and Video S4, Supporting Information).

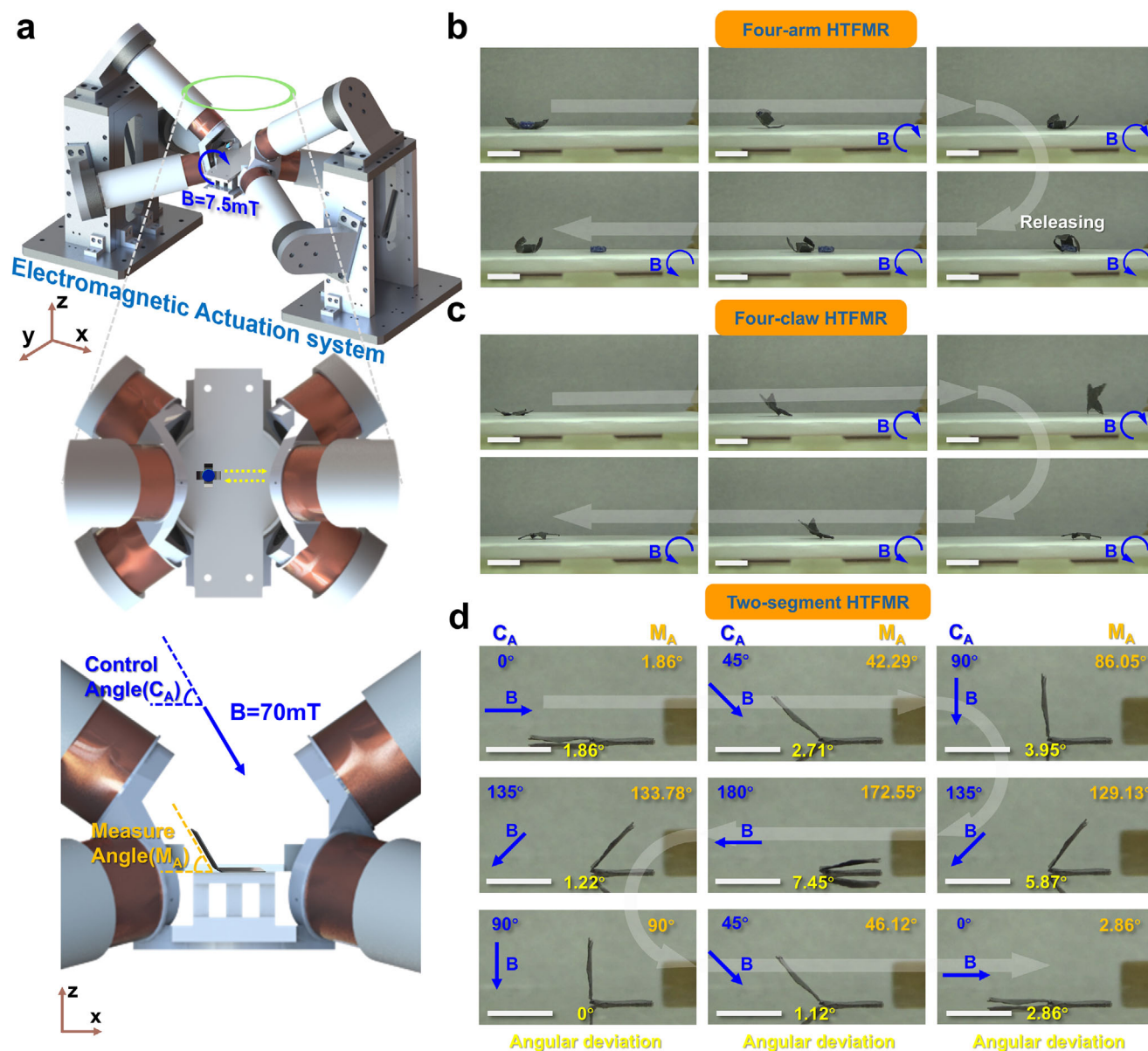


Figure 6. Demonstration of HTFMR controllability under an electromagnetic actuation system. a) Schematic illustration of the EMA system and its workspace. b) The rolling motion and internal cargo release of the four-arm HTFMR under the rotating magnetic field generated by the EMA system. c) Reciprocal rolling movements of the four-claw HTFMR within the workspace. d) Periodic angular positioning of the two-segment HTFMR under system control, with a comparison between the controlled and measured angles. (All scale bars are 10 mm).

For the four-claw HTFMR, the magnetic pole orientation at the tips of the claws is perpendicular to that of the main body. As a result, when exposed to a continuous uniform magnetic field, the claws align vertically relative to the body (Figure S11 and Video S4, Supporting Information). This vertical alignment is particularly effective on soft, mucosal surfaces. Once adhered, the claws penetrate the surface under magnetic influence, achieving robust anchoring. To illustrate this behavior, Figure S11b (Supporting Information) shows the sequential rolling of the HTFMR from P1 to P5 under an external field. Upon arrival, the initially flat body morphs into a raised four-claw structure. COMSOL simulations further depict the tip displacement during claw closure

and the corresponding displacement–field strength relationship at selected points (Figure S11c, Supporting Information).

The HTFMRs also demonstrate excellent controllability within the electromagnetic actuation (EMA) system, which uses six coils to generate a tunable, uniform magnetic field across the workspace. The direction of this field can be precisely adjusted along the x, y, and z axes (Figure 6a).

When placed into this workspace, the four-arm HTFMR and four-claw HTFMR are manipulated simultaneously. A rotating magnetic field applied on the xz-plane enables rolling locomotion of the four-claw HTFMR while allowing the four-arm HTFMR to transport and release cargo with high precision (Figure 6b,c).

Additionally, the four claws of the HTFMR can transition from an expanded to a closed state under a uniform magnetic field, further demonstrating its adaptability (Video S5, Supporting Information).

To further evaluate precision control, a two-segment HTFMR was tested for angle regulation. The HTFMR was subjected to cyclic motion—folding from 0° to 180° and unfolding back—using incremental and decremental steps of 10°, 30°, and 45°. By comparing the programmed input angles (Figure 6d; Video S6, Supporting Information) with those measured through image analysis using AutoCAD 2018, the accuracy of the HTFMR's response was assessed. Results show that the average angular deviation within the 0°–90° range is smaller than that in the 90°–180° range, indicating that larger deformations generate stronger rebound forces, which affect control precision. Furthermore, the angular deviation during unfolding is noticeably smaller than during the initial folding. This observation reinforces the beneficial role of stress reversal and rebound cancellation—discussed in Section 2.3—in improving angle control.

In parallel, a separate experiment was conducted to evaluate the HTFMR's precision under repeated low-field actuation and to verify that the internal magnetic particles were not unintentionally reprogrammed. The two-segment HTFMR was cyclically actuated between 30° and 120° using the EMA system, during which 100 angle values were recorded (Figure S12, Supporting Information). Each angle was extracted and analyzed frame by frame using AutoCAD 2018. The results indicated that the HTFMR consistently achieved angles close to the programmed targets of 30° and 120°, with precision improving over time. These findings confirm that the internal magnetic particles remained stable throughout actuation, underscoring the HTFMR's excellent reliability and controllability under repeated magnetic stimulation. Looking ahead, the demonstrated feasibility, efficiency, and precision of EMA-driven HTFMR manipulation confirm the system's reliability and stability for complex biomedical applications.

Furthermore, the current system offers limited positioning accuracy, and future efforts will focus on refining the magnetic control platform. Since the precision of magnetic actuation is closely tied to the control hardware, our present six-coil setup provides uniform magnetic fields and gradients but lacks the spatial resolution required for fine control. Looking ahead, integrating advanced multi-coil electromagnetic systems—such as Helmholtz–Maxwell arrays or clinically validated EM navigation platforms—holds strong potential to improve actuation smoothness, enable finer step resolution, and enhance position stability, thereby advancing the HTFMR's applicability in complex biomedical environments.

2.6. Potential of Biological Application

To investigate the potential of HTFMR in biological environments, a phantom experiment was conducted. This experiment aimed to assess the four-claw HTFMR's adhesion capability on a gastric model surface, as well as the cargo transport and release functions of the four-arm HTFMR—both controlled via an external magnetic field.

In the experiment, the four-claw HTFMR was guided forward by continuously rolling under the influence of a rotating permanent magnet. Upon reaching the designated site, the magnet was moved closer to enhance the magnetic force acting on the HTFMR (Figure 7a; Video S7, Supporting Information). This increased force enabled the four sharp claws to clamp onto and anchor the HTFMR securely to the folded surface of the gastric model. To release the HTFMR, the magnetic field direction was reversed to eliminate the clamping effect, and continued rotation of the magnet successfully expelled the HTFMR from the model. This experiment clearly demonstrates the working mechanism and controllability of the HTFMR, highlighting its potential utility in biomedical applications. Although the relatively rigid surface of the gastric model limited the display of shape retention, it effectively demonstrated the robot's ability to traverse uneven gastric surfaces.

To assess the gripping capability of the four-claw HTFMR for attachment to gastrointestinal mucosa under external magnetic field control, a dedicated experiment was performed. A 10GRAMS load cell was employed to measure the blocking force while the HTFMR, mounted on a shaft, was actuated under an external magnetic field (maximum ≈ 200 mT). The blocking force generated by a single claw was ≈ 11.63 mN (Figure S13a, Supporting Information). A larger, long segment HTFMR produced a force of ≈ 18.13 mN, as shown in Figure S13b (Supporting Information). Additionally, the device exhibited flexible and repeatable actuation behavior (Figure S13c, Supporting Information), confirming its stable mechanical performance under magnetic stimulation.

Microscopic analysis revealed that the tip area of a single claw was ≈ 0.3 mm $\times$ 0.14 mm (Figure S14, Supporting Information). Based on the measured gripping force, the corresponding tip pressure was calculated to be ≈ 0.277 MPa, which is close to the penetration threshold reported in previous studies.^[8] Although this pressure could be further enhanced through optimization, actual tissue penetration at the centimeter scale may lead to undesired clinical consequences. Thus, rather than aiming for penetration, these results indicate that the HTFMR possesses sufficient gripping force for reliable mucosal attachment without compromising tissue integrity.

Notably, as previously shown on a soft hydrogel substrate (Figure 1c), the claws physically embed into the surface under magnetic actuation, demonstrating stable adhesion enabled by its shape-retaining capability. Looking ahead, to further improve its clinical applicability and achieve more reliable tissue adhesion, anchoring performance could be enhanced by introducing microtextured surfaces to increase mechanical grip or applying a bioadhesive coating (e.g., on the parafilm layer) that remains inactive until it contacts moist tissue, thereby providing stronger and more selective adhesion for *in vivo* use.

Meanwhile, the four-arm HTFMR demonstrated its ability to stably retain internal cargo during movement. Its shape-retention properties allowed it to roll under magnetic field guidance without dropping the cargo. Upon reaching the target area, rapid alternation of the permanent magnet's position from left to right triggered a shaking motion, which dislodged and released the cargo. Similar to the four-claw HTFMR, it could then be expelled from the gastric model through continued magnetic manipulation (Figure 7b; Video S7, Supporting Information).

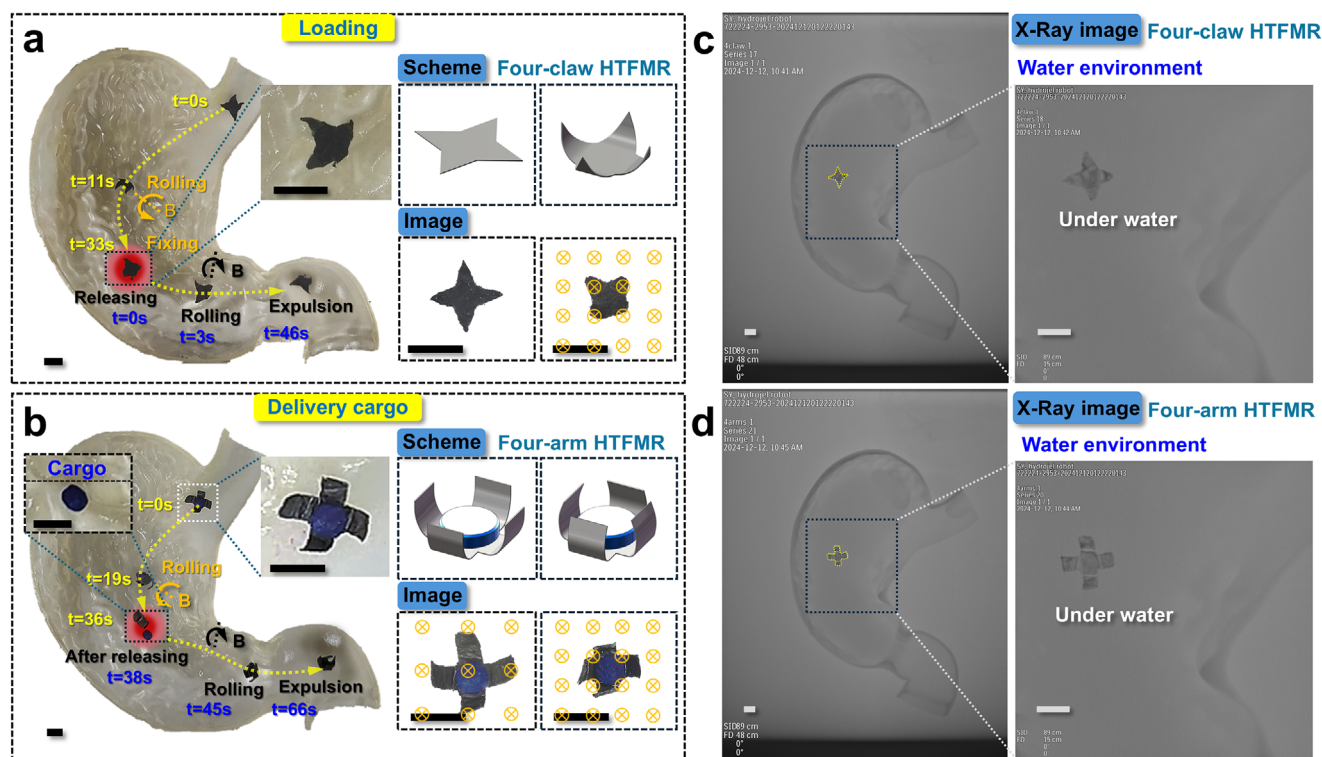


Figure 7. Motion of two different morphological HTFMRs within a stomach model: The four-claw HTFMR and the four-arm HTFMR. a) The rolling motion, attachment to the surface, and expulsion of the four-claw HTFMR within a stomach model, with schematic illustrations of the magnetic fields in different deformation states. b) The cargo delivery and active release capabilities of the four-arm HTFMR, with schematic illustrations of magnetic field density in two different states. The orange “x” symbols indicate the magnetization direction, and wider spacing implies weaker magnetic field intensity. c,d) X-ray imaging of the four-arm and four-claw HTFMRs within a gastric model. (All scale bars are 10 mm).

To further ensure operational reliability in gastric environments, the HTFMR's chemical stability was tested under mildly acidic conditions (pH 4.3). Samples were immersed in the solution for 48 h and showed no observable surface degradation under microscopic inspection, indicating strong acid resistance (Figure S15, Supporting Information).

In practical biomedical settings, the precise localization of microbots such as the HTFMR is essential, necessitating both accurate magnetic actuation and reliable imaging compatibility. To evaluate this, both four-arm and four-claw HTFMRs were assessed under clinical imaging conditions. The HTFMRs exhibited clear visibility under X-ray in both dry (Figure S16, Supporting Information) and aqueous environments (Figure 7c,d), with well-defined contours that enabled reliable identification of their position and shape. These imaging characteristics support real-time feedback and closed-loop control using modalities such as X-ray fluoroscopy, ultrasound, or MRI—strategies widely adopted in magnetic catheter navigation and capsule robotics. Together, they underscore the HTFMR's compatibility with existing clinical infrastructure and its potential for precise, image-guided deployment.

Building upon these attributes—including magnetic actuation, post-actuation shape retention, and high imaging contrast—we envision a practical application scenario in which the HTFMR is introduced into the gastrointestinal tract and passively transported via natural peristalsis. During this transit, the

cargo is securely held by the HTFMR's stable shape-retention capability. Once the target site is reached, X-ray imaging combined with external magnetic field control allows precise positioning, after which a brief magnetic signal can trigger controlled release. This approach minimizes the need for continuous monitoring or complex real-time actuation, enhancing operational robustness and tolerance to interruptions. Similarly, when used as an implantable anchoring patch, this imaging-guided, shape-programmable strategy could enable accurate and reliable placement, highlighting the HTFMR's promise for untethered biomedical applications.

3. Conclusion

This study presents a hydrogel thin-film magnetic millirobot (HTFMR) with outstanding shape-retention properties, enabling rapid and precise responses to external magnetic field control. Its programmable and repeatable deformation behavior supports multi-state folding, greatly enhancing its functional versatility. The HTFMR also demonstrates a wide range of agile motion modes—including self-folding, rolling, and crawling—allowing it to adapt dynamically to different tasks and environments. Experimental validation confirmed the HTFMR's ability to perform accurate cargo transport, controlled release, and stable adhesion to various substrate surfaces. These capabilities point to promising applications in biomedical scenarios.

Looking ahead, HTFMRs show strong potential for integration into targeted drug delivery systems, such as implantable or attachable medical patches. The successful application of an electromagnetic actuation (EMA) system further supports the feasibility of achieving precise, real-time control in vivo. Moreover, the HTFMR's visibility under X-ray imaging strengthens its candidacy for clinical deployment. Future research will focus on optimizing its use in drug delivery, improving the fabrication process, and further miniaturizing the HTFMR to better meet the demands of biomedical and clinical applications.

4. Experimental Section

Materials: Poly(vinyl alcohol) (PVA) with a molecular weight of 89,000–98,000, ammonium hydroxide solution (28%), tetraethyl orthosilicate (98%), and sodium tetraborate were purchased from Sigma-Aldrich. A 4-inch roll of Parafilm was also sourced from Sigma-Aldrich. NdFeB powder (MQP-15-7, average diameter 5 μm) was obtained from Magnequench, Inc. All materials were used in their original state without further purification.

Preparation of the HTFMR: To synthesize the magnetic hydrogel, 1 g of PVA was dissolved in 20 mL of deionized water and stirred at 400 rpm on a magnetic heating stirrer at 100 $^{\circ}\text{C}$ for 4 h. The resulting PVA solution (2 mL) was then transferred to a stirring container, where 3 g of NdFeB powder was added. The mixture was sonicated while stirring for an additional 10 min. Meanwhile, 1 g of sodium tetraborate was dissolved in 20 mL of deionized water, and 1 mL of this solution was added to the container. The two solutions were combined in a 2:1 volume ratio and stirred vigorously until a magnetic slime formed. For formulations with different internal compositions, the same procedure was followed, adjusting the amounts of NdFeB powder and the PVA to borax ratio as required. Initially, the material exhibited non-Newtonian behavior and lacked a defined shape. It was then encapsulated in two layers of Parafilm, stretched, and cut into the desired shape using scissors or a knife. However, due to the toxicity of NdFeB particles, the magnetic slime was not biocompatible. To address this, the NdFeB microparticles were coated with SiO_2 using the Stöber method, as described in previous research.^[40] The procedure followed the method outlined in the referenced paper.^[43]

Characterization of the HTFMR: Surface topography and elemental composition were observed using scanning electron microscopy (SEM) (SU-8010, HITACHI, Japan), coupled with energy-dispersive X-ray (EDX) analysis. The main elements present on the surface of the NdFeB particles were analyzed after coating with SiO_2 via the Stöber method. The magnetic hydrogel was dried using a freezer (OPERON ADVANTECH, FDCF-12003, Korea) at -110°C before imaging. The magnetization curves of the HTFMR and its major components were measured using a vibrating sample magnetometer (VSM) (Lake Shore Cryotronics 7404, Westerville, OH, USA). The imaging performance of the HTFMR in an aqueous environment was tested by placing the HTFMR in a gastric model and imaging it using X-ray (PHILIPS, Azurion 7M20, Netherlands).

In Vitro Cell Culture and Viability Test: The Raw264.7 macrophage cells were obtained from the American Type Culture Collection (ATCC, USA). These cells were maintained in a DMEM culture medium (Corning, USA), supplemented with 10% FBS and 1% penicillin-streptomycin at 37 $^{\circ}\text{C}$ and 5% CO_2 . Cell viability was verified using the AlamarBlue assay. Briefly, Raw264.7 cells were seeded at a density of 2×10^4 cells per well in 96-well plates for 24 h. The medium was then discarded, and 100 μL of fresh medium containing different concentrations of magnetic hydrogel (NdFeB@SiO_2 : 50%) was added to the cells. The cells were co-cultured with the samples for another 24 h. After washing, 10% diluted reagent in the medium was added to the 96-well plates and incubated for 2 h. The supernatants were analyzed by measuring absorbance with a microplate reader. A Live/Dead cell viability assay was also performed to confirm the results.

Magnetic-Actuation Systems: (1) When manually programming and controlling the HTFMR, a permanent magnet fixed at the end of a rod-shaped body was used, enabling flexible manipulation and rotational movements. The simulated magnetic field strength of this permanent magnet, as calculated with COMSOL Multiphysics, and the experimentally measured magnetic field strength, obtained using a Teslameter (FW Bell 8030, Acton, MA, USA). (2) The EMA system, which consists of six electromagnetic coils, was used for 3D manipulation of the HTFMR. Six power supplies (MX15, AMETEK, USA) controlled the coil currents. The maximum values achieved for the magnetic field, magnetic gradient, and current were 70 mT, 1.5 T m^{-1} , and 5.2 A, respectively, with control provided by a LabVIEW program.

Simulation and Computation: Finite element simulations of the HTFMR's shape-changing behavior were performed using COMSOL Multiphysics. Gravity was accounted for in the finite element models. The materials in the simulation were assumed to be incompressible and followed the neo-Hookean material model. The residual magnetic flux density, Young's modulus, and Poisson's ratio of the HTFMRs were set to 0.0876 T, 84.5 kPa, and 0.5, respectively.^[32,49] The distribution of magnetic moment density in the HTFMRs was discretized and simplified.

To calculate the residual magnetic flux density of the HTFMR, the magnetization at zero external magnetic field ($M_r = 33.37 \text{ emu g}^{-1}$) was obtained from the VSM measurements. In the calculation of ρ_{HTFMR} , the contribution of the outer film layer was neglected, resulting in $\rho_{\text{HTFMR}} = 2.09 \text{ g cm}^{-3}$. The density of the HTFMR (ρ_{HTFMR}), the magnetization (M_r), and the residual magnetic flux density (Br) was calculated using the following equation:

$$\rho_{\text{HTFMR}} = \frac{\rho_1 V_1 + m_2}{V_1 + V_2} \quad (2)$$

$$M_r (\text{A m}^{-1}) = M_r (\text{emu g}^{-1}) \times \rho \times 10^3 \quad (3)$$

$$B_r = \mu_0 M_r \quad (4)$$

where ρ_1 is the density of NdFeB, V_1 is the volume of NdFeB calculated from its mass, m_2 is the mass of the hydrogel, and V_2 is the volume of the hydrogel. μ_0 is the permeability of free space ($4\pi \times 10^{-7} \text{ H m}^{-1}$).

The magnetic flux density in the simulation domain is expressed by:

$$B = \mu_0 \mu_{\text{rec}} H + B_r \quad (5)$$

where B is the magnetic flux density (T), μ_0 is the permeability of free space. μ_{rec} is the relative permeability of the material, and H is the magnetic field strength (A m^{-1}).

The magnetic force exerted on the HTFMR is given by:

$$F = \int_{\partial\Omega} n T dS \quad (6)$$

where F is the total force acting on the magnetic object. $\partial\Omega$ is the boundary surface of the object, n is the outward unit normal vector on the boundary surface, T is the stress tensor, and dS is the differential surface element.

Statistics and Data Analyses: A Student's t-test was used to compare experimental data. Data were presented as mean $\pm$ standard deviation (SD). The sample size (n) for each analysis was specified in the corresponding figure legends or table captions. All statistical analyses and figure generation were conducted using Origin 2024.

Supporting Information

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

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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

hydrogel millirobot, magnetic actuation, reprogrammable deformation, shape retention

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