

MATERIALS SCIENCE

Supramolecular hydrogel actuators with reprogrammable magnetic orientation by locally mediated viscoelasticity and pinning force

Chuan Wei Zhang^{1,2†}, Xing Peng Hao^{1†}, Weifeng Zou³, Zhixin Zhu¹, Jia Yu Hu¹, Li Xin Hou¹, Si Rui Xu¹, Zhen Luo², Yichen Yan², Andrea Sarabia^{4,5}, Andrea Litwak^{4,5}, Shili Xu^{4,5,6}, Zhi Jian Wang^{7*}, Ximin He^{2*}, Qiang Zheng¹, Zi Liang Wu^{1*}

Magnetic soft actuators with reprogrammable deformations have gained substantial attention due to their adaptability for various applications. However, achieving precise and local reorientation of magnetic particles remains challenging. Here, we present a strategy to locally tailor the viscoelasticity of magnetic supramolecular hydrogels, facilitate reorientation of the embedded magnetic particles, and enable reprogrammable magnetoactuated deformation and locomotion of the composite gels. The magnetic hydrogels are facily prepared by mixing neodymium-iron-boron particles with an aqueous poly(acrylic acid-co-acrylamide) solution, which spontaneously forms supramolecular network with carboxylic-ferric ion coordinates as physical cross-links. This network enables dynamic control of viscoelasticity by localized laser heating, which reduces the pinning force of gel matrix and allows for reorientation of magnetic particles under a modest magnetic field. We demonstrate that the same hydrogel sheet can be reprogrammed to exhibit various complex deformations and locomotion. This versatile approach to developing magnetic hydrogels with adaptive responses offers exciting potential for soft robotics and biomedical devices.

INTRODUCTION

Magnetic soft materials have emerged as a fascinating platform in materials science (1–5), where their unique integration of mechanical flexibility and magnetic responsiveness enables unprecedented control over material behaviors and functions (6–8). Unlike conventional responsive materials, magnetic soft materials can undergo instantaneous and reversible transformations under magnetic fields, offering precise control over shape reconfiguration, directed motion, and programmed functions (9–11). Through strategic manipulation of magnetic field distributions and material architectures (12, 13), researchers can program these materials to mimic complex biological movements, leading to adaptive soft machines capable of real-time feedback and control (14, 15). The noncontact actuation mechanism of these materials (16, 17) proves particularly valuable in biomedical applications, where they serve as remotely controlled surgical tools (18) that enable minimally invasive procedures to minimize patient trauma. By incorporating additional functional components, these magnetic systems can be further optimized to respond to multiple environmental cues, creating multifunctional platforms that expand their potential applications (19–22).

Although incorporating magnetic particles into soft materials affords promising opportunities for responsive and multifunctional systems, controlling their spatial arrangement and orientation within soft matrices remains a critical challenge (23–27). This spatial control directly determines the material's magnetic responsiveness and functionality. Current approaches, while innovative, face substantial limitations. For instance, three-dimensional (3D) printing technology harnesses the interplay between printing paths and external magnetic fields, as demonstrated by Zhao and colleagues (28), enabling precise particle alignment and programmable magnetic properties during fabrication. However, this method struggles with postfabrication reconfigurability. Once the matrix is cured, the aligned particles experience a high pinning force, a force exerted by the surrounding polymer network due to rigid spatial constraints and strong interactions between the particles and the network, which restricts their movement and reorientation. Forcing their orientation risks compromising the material's structural integrity, limiting the development of truly adaptable systems. To address this issue, Sitti and colleagues (23) developed a strategy using localized laser heating to dynamically control magnetic properties by selectively heating regions above the Curie temperature of magnetic particles for remagnetization. However, for strong magnetic materials such as neodymium-iron-boron (NdFeB) with high Curie temperatures (typically exceeding 300°C), this approach faces substantial limitations. In contrast, Lin and co-workers (24) elegantly lowered the operating temperature using phase change polymers as the pockets of magnetic particles, where the pinning force decreases once the pocket polymer melts. However, this method is primarily applicable to elastomeric matrices and is not suitable for hydrogels, as the melted pocket material can flow and seep through the gel network. Strategies for achieving these reprogrammable magnetic responses in hydrogels are still lacking. This motivates the development of complementary strategies that operate at lower temperatures while maintaining the advantages of localized control.

¹Ministry of Education Key Laboratory of Macromolecular Synthesis and Functionalization, Department of Polymer Science and Engineering, Zhejiang University, Hangzhou 310058, China. ²Department of Materials Science and Engineering, University of California, Los Angeles, Los Angeles, CA 90095, USA. ³State Key Laboratory of Fluid Power and Mechatronic Systems, Zhejiang University, Hangzhou 310058, China. ⁴Department of Molecular and Medical Pharmacology, University of California, Los Angeles, Los Angeles, CA 90095, USA. ⁵Crump Institute for Molecular Imaging, University of California, Los Angeles, Los Angeles, CA 90095, USA. ⁶Jonsson Comprehensive Cancer Center, David Geffen School of Medicine, University of California, Los Angeles, Los Angeles, CA 90095, USA. ⁷Institute for Chemical Reaction Design and Discovery, Hokkaido University, Sapporo 001-0021, Japan.

*Corresponding author. Email: wangzj@icredd.hokudai.ac.jp (Z.J.W.); ximinhe@ucla.edu (X.H.); wuziliang@zju.edu.cn (Z.L.W.)

†These authors contributed equally to this work.

Inspired by magnetotactic bacteria, where magnetosomes readily align in magnetic fields due to the fluid-like cytoplasmic environment and maintain their orientation through protein-mediated interfaces (Fig. 1A), we explored supramolecular hydrogels (29–31) as dynamic matrices for spatial patterning of the magnetic particles (Fig. 1B). Supramolecular hydrogels form physically cross-linked networks through noncovalent interactions [e.g., hydrogen bonding (32, 33), electrostatic interactions (34), metal coordination (35), and hydrophobic associations (36, 37)], mimicking the dynamic internal environment of bacterial cells. Just as the fluid cytoplasm enables easy magnetosome alignment under weak magnetic fields while protein complexes stabilize the arranged structures, supramolecular hydrogels offer switchable viscoelasticity that can be tailored by external stimuli. Unlike covalently cross-linked hydrogels (38), supramolecular gels are particularly sensitive to temperature changes (29) due to their dynamic noncovalent bonds. At elevated temperatures, they usually exhibit a marked change in viscoelasticity characterized by reduced storage modulus and increased loss factor, resembling the fluid-like state of cellular cytoplasm. The decrease in storage modulus, arising from the dissociation of noncovalent interactions and the relaxation of polymeric network, reduces the pinning force on magnetic particles and lowers the required coercivity for particle rotation (Fig. 1C). Therefore, this biomimetic strategy should enable the localized reorientation of magnetic particles under relatively weak magnetic fields and at mild temperatures (well below degradation temperature of polymers and Curie temperatures of magnetic particles).

To validate this design, we select a metallosupramolecular hydrogel as the matrix for NdFeB magnetic particles. The composite hydrogel is formed by spontaneous acid decay of magnetic particles

and exposure of ferric ions to form carboxyl- Fe^{3+} coordination complexes as physical cross-links of poly(acrylic acid-*co*-acrylamide) [P(AAc-*co*-AAM)] chains (39, 40). The viscoelasticity of the supramolecular hydrogel depends on temperature, allowing reversible tuning of the pinning force and subsequent reorientation of the particles under a magnetic field. Leveraging the photothermal effect of magnetic particles, laser heating is achieved to afford localized modulation of the viscoelasticity, enabling reprogrammable orientation of the magnetic particles under mild conditions. Thus, obtained patterned hydrogels show reprogrammable morphing and motion behaviors by harnessing distinct magnetic responses in local regions with different magnetic particle orientation. Robotic function is also imparted to nonresponsive elastomer by adhesive integration with magnetic responsive supramolecular hydrogel. This strategy should be applicable to various supramolecular materials, facilitating the development of magnetic soft robots with versatile functions and applications.

RESULTS

Hydrolysis-assisted synthesis of magnetic hydrogels

NdFeB particle-containing P(AAc-*co*-AAM) hydrogels were prepared by a straightforward yet efficient strategy involving spontaneous acid decay of the magnetic particles and physical cross-linking of the copolymer chains, as illustrated in Fig. 2A. The procedure begins with the synthesis of a viscous copolymer solution through free radical copolymerization of acrylic acid (AAc; 0.14 M) and acrylamide (AAM; 1.26 M) in water. Then, NdFeB particles (0 to 30 vol %) are directly blended into the copolymer solution using a

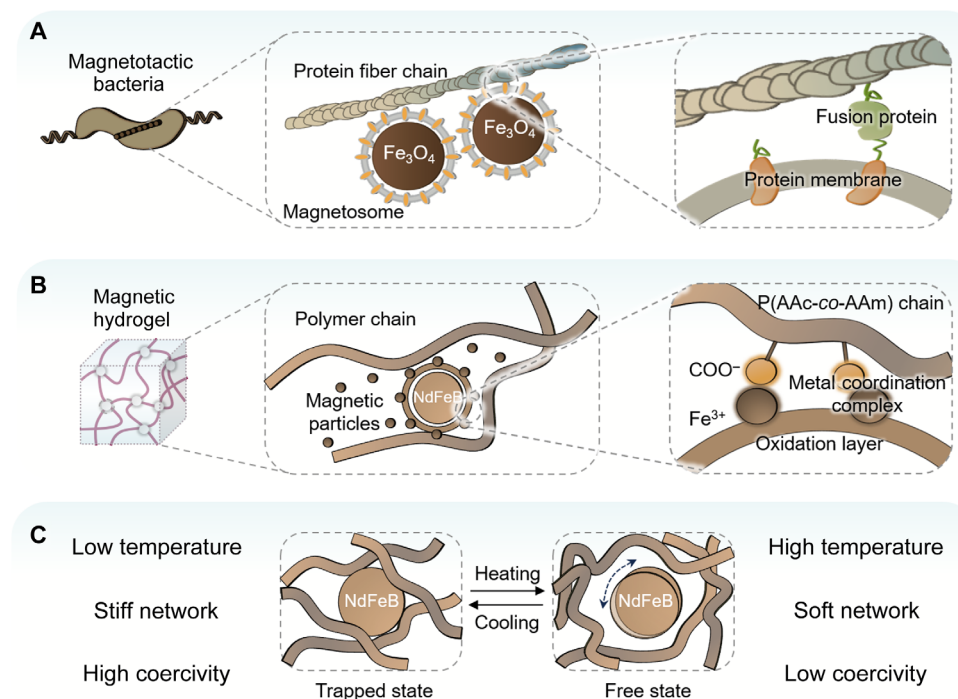


Fig. 1. Biomimetic design of magnetic supramolecular hydrogel with tunable viscoelasticity for controlled orientation of magnetic particles. (A) Magnetotactic bacteria with magnetosome chains assembled by protein-mediated organization of Fe₃O₄ nanoparticles. (B) Synthetic analog: a supramolecular hydrogel composed of NdFeB particles embedded in P(AAc-*co*-AAM) polymer network cross-linked by Fe³⁺-carboxylate coordination. (C) Reversible tuning of pinning force and magnetic particle reorientation under weak magnetic fields via temperature-dependent viscoelastic modulation of the gel matrix.

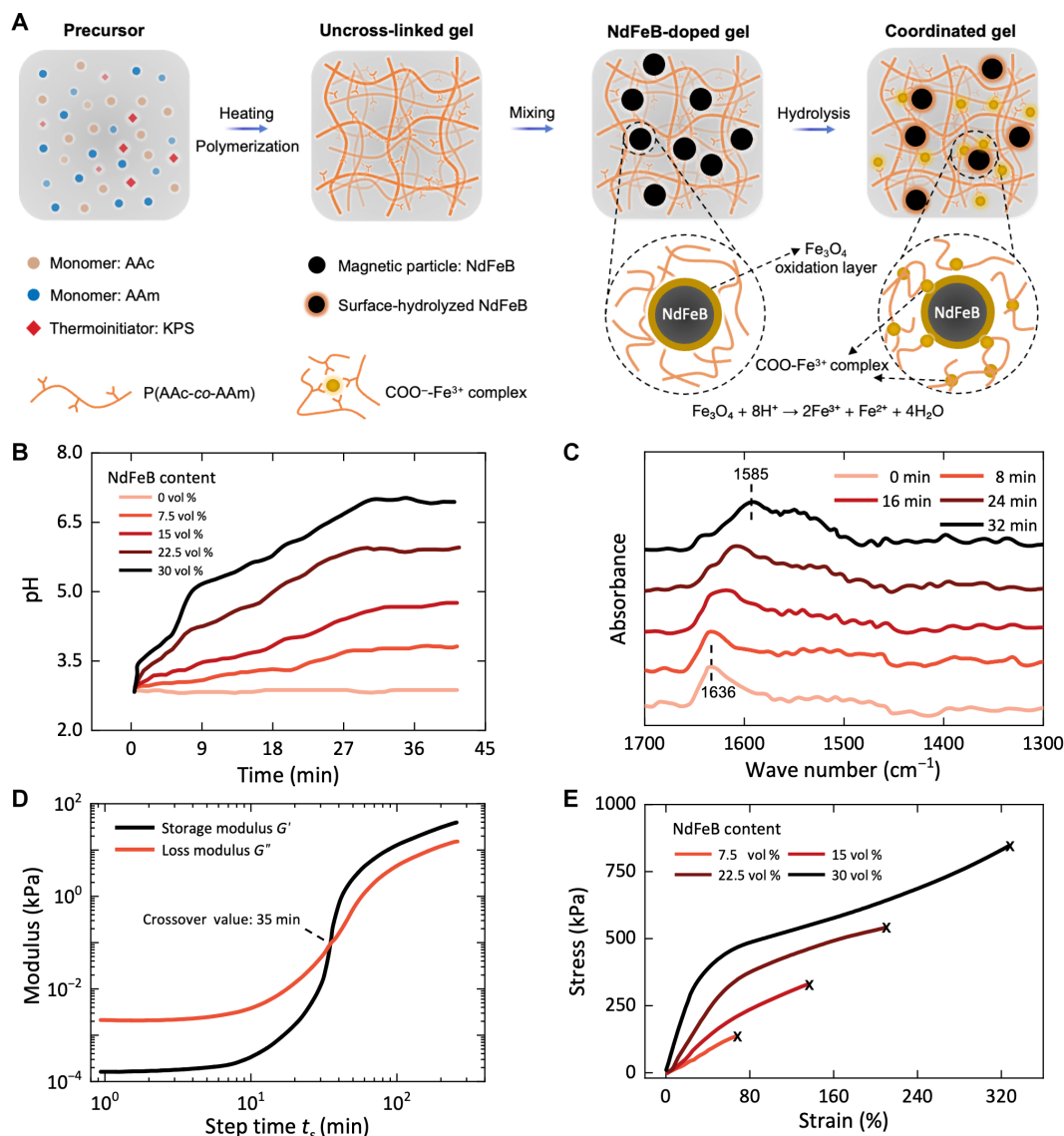


Fig. 2. Synthesis of magnetic hydrogels via spontaneous metal coordination cross-linking and their mechanical characteristics. (A) Schematic representation of the synthesis process. A viscous P(AAc-co-AAm) copolymer solution was obtained through free radical polymerization of AAc and AAm using potassium persulfate (KPS) as the initiator, followed by incorporation of NdFeB particles to form the supramolecular hydrogel. (B) Time-dependent pH profiles of the systems with different contents of NdFeB particles. (C) Infrared spectra of the system at various times after mixing the copolymer solution with 30 vol % of NdFeB particles. (D) Time sweep test of the copolymer solution after mixing with 30 vol % of NdFeB particles. (E) Tensile stress-strain curves of the magnetic hydrogels with different contents of NdFeB particles.

vortex mixer. The high viscosity of the solution effectively prevents particle sedimentation after mixing, ensuring homogeneous distribution of the particles during the subsequent gelation process. The physical cross-linking process is closely associated with the acid-triggered corrosion of the surface oxide layer of the NdFeB particles, which is predominantly composed of Fe₃O₄. The acidic environment provided by the abundant carboxylic acid groups in the copolymer solution promotes the hydrolysis of this oxide layer. Scanning electron microscopy (SEM) coupled with energy-dispersive x-ray spectroscopy of the NdFeB particles (fig. S1) confirms the uniform distribution of Fe and O elements on the particle surface, providing direct evidence for the presence of an oxide layer.

Upon mixing, the hydrolysis reaction at the particle surface releases Fe³⁺ ions, following the reaction: Fe₃O₄ + 8H⁺ → 2Fe³⁺ + Fe²⁺ + 4H₂O. These Fe³⁺ ions subsequently induce gelation by forming carboxyl-Fe³⁺ coordination complexes (fig. S2) (41, 42). This process is accompanied by a substantial increase in the pH of the suspension, as protons from the carboxylic acid groups are consumed during hydrolysis (Fig. 2B). If the pH of copolymer solution is initially adjusted using sodium hydroxide before mixing, then the Fe³⁺ ions cannot be effectively released, resulting in a much slower gelation process (fig. S3A). This confirms that acid-released Fe³⁺ ions, rather than surface-bound Fe₃O₄, are primarily responsible for cross-linking with the carboxyl groups. To further probe this

mechanism, we compared NdFeB particles with pure Fe_3O_4 particles (both at 30 vol %). As shown in fig. S3B, both systems exhibit similar evolution profiles of the storage modulus (G'), suggesting that a comparable coordination cross-linking mechanism is involved. This observation supports the hypothesis that Fe_3O_4 hydrolysis is the primary source of Fe^{3+} ions responsible for gel formation. To elucidate the state of Fe^{3+} ions during gelation, we conducted ultraviolet-visible spectroscopy following acid treatment of NdFeB particles in the absence of polymer, with subsequent removal of NdFeB particles by filtration. The supernatant exhibited characteristic absorption features at ~ 200 and ~ 290 nm (fig. S4), confirming the presence of Fe^{3+} ions in the aqueous phase. These findings support that Fe^{3+} ions are released from the particle surface into solution, where they coordinate with carboxyl groups to mediate physical cross-linking.

The hydrogel formation is governed by a hydrolysis-induced coordination process, which exhibits clear time-dependent behavior. Fourier transform infrared (FTIR) analysis (Fig. 2C) confirms the formation of coordination complexes between carboxyl groups and Fe^{3+} , indicated by the shift of the carboxyl stretching band from 1636 to 1595 cm^{-1} . In systems containing 30 vol % of NdFeB, the sol-gel transition occurs at ~ 35 min (Fig. 2D), while mechanical properties such as Young's modulus continue to increase until plateauing after ~ 60 min (fig. S5A). This evolution reflects the gradual establishment of a coordination network, with a higher cross-linking density anticipated near the NdFeB particles due to localized Fe^{3+} release.

As the particle concentration increased from 7.5 to 30 vol %, Young's modulus rose from 0.22 to 1.4 MPa, and the breaking strength

increased from 150 to 850 kPa (Fig. 2E and fig. S6), demonstrating a strong correlation between particle loading and mechanical robustness. A notable advantage of our approach is its operational simplicity. The entire supramolecular gel preparation occurs under ambient conditions without requiring special protection or handling procedures.

Tunable magnetic properties of the composite hydrogels

The magnetic properties of the supramolecular hydrogels at ambient temperature exhibit a strong dependence on the NdFeB particle content, as revealed by well-defined magnetic hysteresis curves (Fig. 3A). With an increase in NdFeB particle content from 0 to 30 vol %, the saturation magnetization (B_s) and remanence magnetization (B_r) increase proportionally (Fig. 3B). This linear increase indicates the uniform particle distribution throughout the hydrogel matrix and consistent magnetic performance across different concentrations. At 30 vol % loading of NdFeB, B_s reaches 176 emu/g and B_r reaches 121 emu/g, remarkably retaining 68.8% of pure NdFeB particles' magnetization ability. This high retention rate highlights the effectiveness of our coordination cross-linking strategy in preserving the intrinsic magnetic properties of NdFeB particles while successfully integrating them into the hydrogel matrix. Notably, the coercivity (H_c) of our magnetic hydrogel shows a marked increase from 112 to 632 mT with rising NdFeB content, suggesting a stronger magnetic domain pinning force due to the more densely cross-linked network.

The strength of the coordination network can also be modulated by adjusting the polymer concentration and the AAc content. As

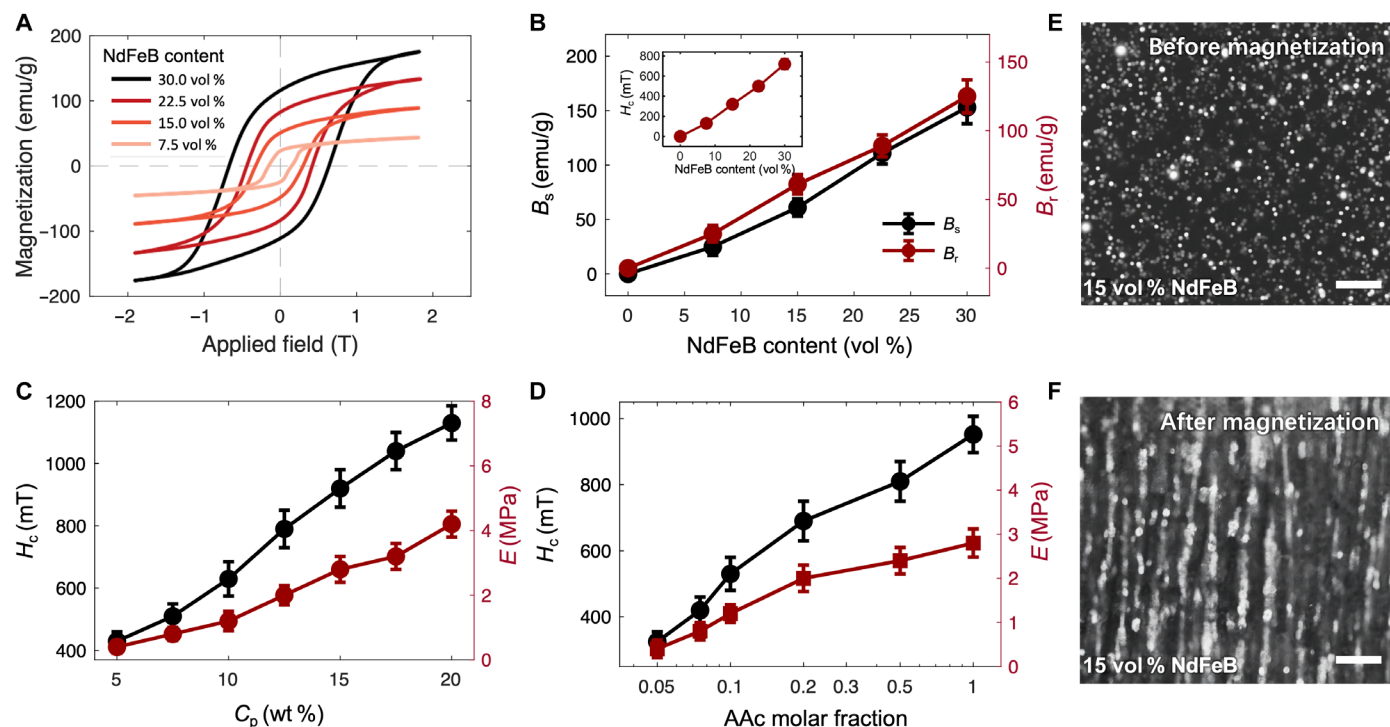


Fig. 3. Magnetic properties and structural organization of magnetic hydrogels. (A) Magnetic hysteresis loops of hydrogels with different NdFeB contents. (B) Saturation magnetization (B_s), remanent magnetization (B_r), and coercivity (H_c ; inset) measured for samples with varying NdFeB contents. (C) Coercivity (H_c) and Young's modulus (E) determined at different total polymer concentrations (C_p). (D) Coercivity and modulus measured at varying AAc molar fractions. (E and F) SEM images of hydrogels with 15 vol % of NdFeB before (E) and after (F) magnetization, with particle alignment observed under applied magnetic field. Scale bars, 30 μm .

shown in Fig. 3C, when the molar ratio of AAc is fixed at 0.1 and NdFeB content at 30 vol %, increasing the total polymer concentration from 5 to 20 wt % leads to a substantial rise in coercivity from 420 to 1130 mT, along with an increase in Young's modulus (E). This enhancement is attributed to the formation of a denser network at higher polymer concentrations. Similarly, as shown in Fig. 3D, when NdFeB content is fixed at 30 vol % and the total polymer concentration at 10.5 wt %, increasing the AAc molar ratio from 0.05 to 1.0 raises the coercivity from 325 to 970 mT. This trend is attributed to the more acidic environment and higher carboxyl group content, which promote greater ion release and coordination cross-linking, resulting in a denser network and stronger pinning effect. These results demonstrate that beyond varying NdFeB content, magnetic properties can be finely tuned through structural parameters of the polymer network. A time-dependent evolution of coercivity is also observed (fig. S5B), with values increasing rapidly from 150 to ~630 mT within the first 60 min before plateauing, indicating the parallel progression of network cross-linking and magnetic domain pinning. This behavior contrasts with other systems (43), where magnetic particles are embedded in matrices whose mechanical properties are largely independent of particle content, leading to coercivity that is unaffected by magnetic loading.

After magnetization, the magnetic particles, initially randomly distributed, aligned into chain-like structure along the direction of applied magnetic fields, as clearly visualized through SEM imaging (Fig. 3, E and F). This anisotropic domain formation significantly enhances the gel's mechanical strength both parallel and perpendicular to the direction of magnetic alignment due to magnetic field attraction (fig. S7). Furthermore, these magnetic properties maintain remarkable stability under mechanical deformation, as demonstrated by the preservation of remanent magnetization under various strain conditions (fig. S8A). Long-term cyclic testing further confirms the durability of the magnetic response, with consistent magnetic behavior maintained over extended periods of magnetization cycling (fig. S8B). The stability of the magnetic supramolecular hydrogel supports it to be repeatedly reprogrammed and actuated under an external magnetic field.

Laser heating-modulated viscoelasticity for magnetic domain patterning

To achieve magnetoinduced shape morphing, we should locally pattern the orientation of magnetic particles within the hydrogels, affording different regions with distinct responses to the magnetic field. If the viscoelasticity of the gel matrix is reduced to lower the pinning force on the magnetic particles, then the particles can be oriented under a modest magnetic field. By harnessing the photo-thermal effect of Fe_3O_4 layers on NdFeB particles (44, 45), we can apply 520-nm green laser to locally heat the composite gel, thereby modulating the viscoelasticity of the supramolecular hydrogel and reprogramming the localized orientation of magnetic particles. As illustrated in Fig. 4A, the irradiated regions have a higher temperature and lower pinning force for magnetic domain reorientation than the unirradiated regions. When exposed to a magnetic field with intensity exceeding the coercivity in the irradiated regions rather than the unirradiated regions, only the magnetic particles in the irradiated region become reoriented (fig. S9).

This strategy necessitates temperature-sensitive viscoelasticity and coercivity of the magnetic hydrogels, which perfectly aligns with the characteristics of the supramolecular hydrogels. As shown

in Fig. 4B, with an increase in temperature from 25° to 50°C, the complex viscosity of the supramolecular gel with 30 vol % of NdFeB particles decreases by approximately two orders of magnitude across the test window of frequencies, corresponding to reduced pinning force of the gel network on the magnetic particles. The influence of temperature on the magnetic properties of the gel is crucial for the reprogramming of magnetic domains. While the maximum magnetization remains stable at ~170 emu/g at different temperatures, the coercivity decreases substantially from 600 mT at 25°C to 53 mT at 55°C, reflecting reduced constraints against rotation of magnetic particles at elevated temperatures (Fig. 4C). The marked reduction in coercivity enables domain reorientation of locally heated regions under relatively weak applied fields. To ensure selective reorientation in the irradiated regions, we should set the external magnetic field for reprogramming between the coercivity of the heated and unheated regions. In this study, an 80-mT magnetic field is applied, which is between the coercivity of 600 mT at 25°C and the coercivity of 53 mT at 55°C, for reprogramming the magnetic domains of the composite gel upon laser irradiation. Consistent with this, the reprogramming efficiency shows a sharp increase with temperature, surpassing 90% at 55°C (Fig. 4D). This threshold confirms the existence of a reprogrammable temperature window, in which domain realignment can be achieved using commercially accessible magnetic fields.

This correlation between viscoelastic and magnetic properties is further elucidated through quantitative analysis. As shown in Fig. 4E, coercivity increases linearly with storage modulus, rising from 178 mT at 0.21 MPa to 724 mT at 1.61 MPa, confirming that network stiffness directly governs the magnetic pinning force. In contrast, Fig. 4F presents a nonmonotonic relationship between coercivity and loss factor. With increasing temperature, the loss factor initially rises from 0.34 to 0.79 and then decreases to 0.41. This trend is attributed to enhanced chain mobility at intermediate temperatures, followed by excessive network softening at higher temperatures, which reduces energy dissipation capability. The much stronger correlation between coercivity and storage modulus compared to that with loss factor highlights that the primary contribution to magnetic pinning arises from network stiffness rather than viscoelastic dissipation. These insights provide a mechanistic foundation for the rational design of reprogrammable magnetic hydrogels with clearly defined temperature windows for domain reorientation.

To evaluate the stability of reprogrammed magnetic properties of the gel, we examine the temporal evolution of normalized remanent magnetization (B_r/B_0) at different temperatures (Fig. 4G). At 55°C, only 43% of the magnetization remains after 1 hour, suggesting significantly enhanced particle mobility within a softened matrix. In contrast, at 25°C, the robust network effectively maintains magnetic alignment, preserving 87% of the initial magnetization. These results indicate that the programmed magnetic domain can be reliably maintained at low temperatures. The viscoelasticity and the coercivity of the magnetic gel exhibit consistent and reversible transitions between high and low temperatures (Fig. 4, H and I, and fig. S10), validating our strategy for domain reprogramming without compromising the material's structural integrity. In addition to reversibility, the system demonstrates excellent long-term stability under repeated use. As shown in fig. S11, after 30 thermal-magnetic reprogramming cycles, the hydrogel retains over 90% of its initial coercivity, with only a ~10% decrease in storage modulus and ~5% increase in water content. The maximum magnetization remains within 5%

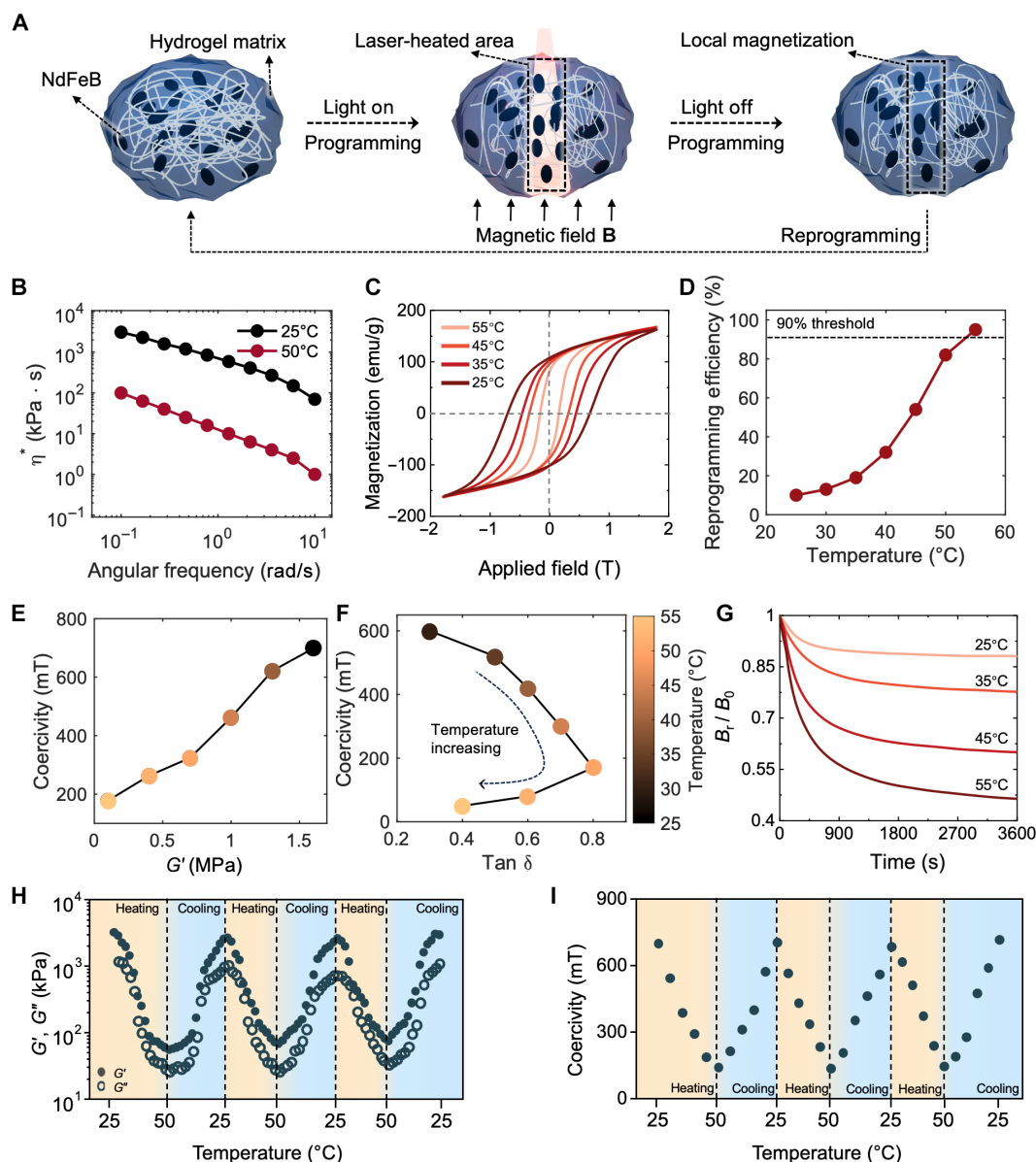


Fig. 4. Laser heating-modulated viscoelasticity and reprogramming of magnetic domains. (A) Schematic illustration of local programming through laser heating and magneto-orientation of magnetic particles: unheated hydrogel matrix (left), laser-heated area under an applied magnetic field (middle), and resultant programming of magnetic domains in the irradiated region (right). (B) Frequency dependence of complex viscosity (η^*) of the magnetic hydrogel with 30 vol % of NdFeB particles at 25°C and 50°C. (C) Magnetic hysteresis loops of the gel at different temperatures. (D) Temperature-dependent reprogramming efficiency of this system. (E) Correlation between storage modulus (G') and coercivity, demonstrating relationship between network stiffness and magnetic pinning. (F) Relationship between $\tan \delta$ and coercivity across temperature gradient. (G) Time-dependent normalized remanent magnetization B_r/B_0 curves of the gel at various temperatures. (H and I) Reversible changes in the storage modulus G' , loss modulus G'' (H), and magnetic coercivity (I) of the magnetic hydrogel during repeated thermal cycles between 25°C and 50°C.

of its original value. This long-term durability confirms that the supramolecular hydrogel preserves its functional integrity throughout repeated reconfiguration processes, making it suitable for practical applications requiring multiple programming cycles. The combination of localized laser heating and reversible viscoelasticity modulation enables unprecedented control over magnetic domain patterns, as well as programmed shape morphing and motions of the patterned gels under magnetic fields, which will be described in the following sections.

Reprogrammable magnetic patterns for morphing and locomotion of the gel

Patterned hydrogels with programmed orientations of magnetic particles at specific regions can be developed by controlling the laser-heated regions while applying directional magnetic fields for magnetic domain orientation. We demonstrate this capability by generating various magnetic patterns in an identical hydrogel, including basketball, botanical, and Chinese knot patterns (Fig. 5A). These magnetic patterns exhibit high stability at room temperature yet can be readily erased

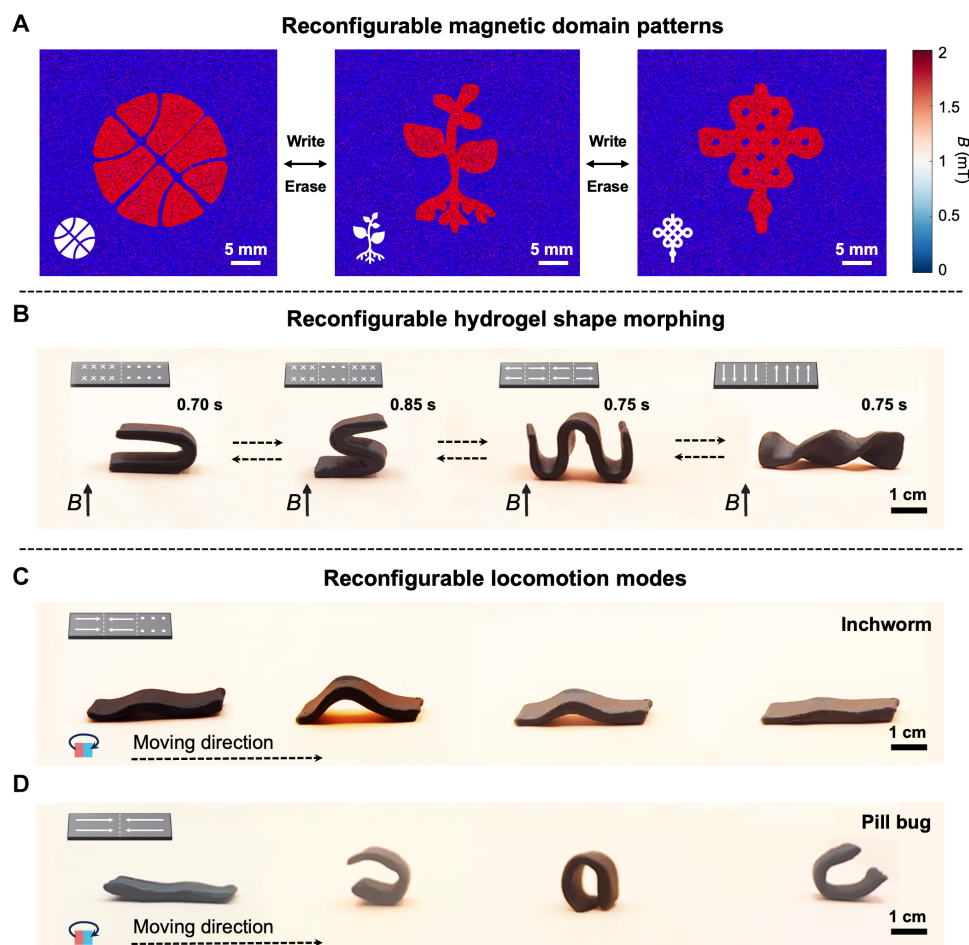


Fig. 5. Reprogrammable magnetic patterns and locomotion of a single hydrogel. (A) Magnetic flux density map showing reprogrammable magnetic patterns transforming from basketball to botanical and Chinese knot patterns on the same gel. (B) Magnetic-filed (80 mT) actuated shape morphing with distinct programmed magnetic patterns, transitioning between U-shape, S-shape, wave-shape, and helix configuration. (C and D) Distinct locomotion modes of the hydrogels with different magnetic patterns under a rotating magnetic field (2 Hz, 80 mT): continuous inchworm-like crawling (C) and pill bug-like rolling (D).

upon uniform heating and then reprogrammed into other patterns, highlighting the reprogrammability of the system. The hydrogel with the same appearance yet distinct magnetic patterns should be an ideal material for rewritable information storage and encryption (46, 47). We systematically examined hydrogels patterned with feature sizes of 2, 1, and 0.5 mm to evaluate the influence of spatial resolution on material properties (fig. S12). As the resolution increases, both Young's modulus and coercivity remain stable, indicating that magnetic pattern resolution has minimal influence on material properties at the tested scale. These findings suggest that magnetic pattern resolution can be leveraged as a design parameter to fine-tune actuator performance without compromising magnetic stability. Given that shape morphing and locomotion behaviors are closely tied to local mechanical and magnetic properties, control over pattern resolution offers a promising strategy for enhancing functionality.

Besides the display of patterned information, the composite hydrogel with programmed magnetic domains shows rapid shape morphing to form specific configurations under external magnetic fields. As shown in Fig. 5B and movies S1 and S2, the slender hydrogel sheet readily deforms into U-shaped, S-shaped, wave-like, and helical configurations under an 80-mT magnetic field, with response times

as low as 0.7 to 0.8 s for complete configuration changes (fig. S13A). After removing the magnetic field, the 3D configurations readily recover to the original flat shape. We should note that the different configurations are from the identical hydrogel under the same magnetic stimulation. This achievement is realized by reprogramming the magnetic patterns within the hydrogel sheet. When compared to other morphing hydrogels, one advantage of magnetic gels is the high morphing speed upon magnetic stimulations, which is often difficult to be realized for hydrogel-based soft actuators and robots.

Considering the instantaneous response to the applied magnetic field, the composite hydrogel with patterned magnetic domains can be effectively used for controlled locomotion with quantifiable performance metrics. Two representative locomotion modes demonstrate this capability. Under a rotating magnetic field (2 Hz, 80 mT), the gel sheet with three distinctly oriented magnetic domains exhibits inchworm-like crawling motion with a speed of 14.3 mm/s (Fig. 5C and movie S3). By strategically reprogramming the identical gel sheet to a different pattern with two distinct magnetic domain regions, we achieved continuous pill bug-like rolling motion with an enhanced speed of 21.3 mm/s (Fig. 5D and fig. S13B). Both locomotion modes maintain precise directional control through

adjustment of the rotating field orientation, with minimal degradation in performance after multiple actuation cycles.

Adhesive integration of magnetic gel with elastomer as functional soft device

Another advantage of this kind of magnetic supramolecular hydrogels is the strong adhesion to various elastomers, enabling the integration of responsive and nonresponsive components to devise soft machines. The integration should start from the gel synthesis to form topological interlocking between the gel and microstructured surface of the elastomer (Fig. 6A) (48, 49). For example, the fresh mixture of NdFeB particle and P(AAc-co-AAM) solution is coated on one surface of commercial polydimethylsiloxane (PDMS) sheet. The suspension wets and fills the micrometer- and nanometer-sized voids of the rough surface. The subsequent physical cross-linking produces a strong adhesion between the gel and elastomer. The interfacial adhesive energy between the two materials is examined by peeling tests at room temperature (Fig. 6B). It increases with the content of magnetic particles, reaching a value of $\sim 400 \text{ J/m}^2$. The influence of particle content on the adhesive is associated with the increased mechanical properties of the composite gel with aligned magnetic particle (fig. S14). This adhesive integration is suitable for other elastomers, generating strong interfacial bonding of the magnetic gel with the VHB and EcoFlex (Fig. 6C).

This robust integration enables the development of hybrid soft devices, affording the nonresponsive elastomer with response to magnetic field. We demonstrate the versatility of this combination by developing a magnetically steerable catheter system by integrating a slender PDMS sheet as the backbone with strategically coated

magnetic gels as the active segments (Fig. 6D). The soft device exhibits wirelessly controlled, multifunctional capabilities, including directional crawling, cargo trapping, and cargo transfer by manipulating the magnetic fields. Figure 6E is a conceptual demonstration of the integrated system capable of effectively navigating while manipulating and transporting the cargo. The PDMS elastomer is integrated at two locations with magnetic hydrogels containing oppositely oriented magnetic particles (vertical to the surface of the elastomer). First, a magnetic field is applied to drive the magnetic gels to move closer to the cargo. Then, by adjusting the magnetic field direction, one end of the elastomer bends to wrap around the cargo, which is subsequently attracted by the other magnetic gel located on the elastomer. This process enables the cargo to be securely grasped and transported away from its original position under a moving magnetic field. Furthermore, our magnetic hydrogel holds potential for in vivo applications, particularly in magnetically guided interventions and minimally invasive tracking. To assess its imaging capability, we embedded the integrated device in agarose gel and performed T2-weighted magnetic resonance imaging (MRI). The hydrogel exhibits clear signal contrast against the surrounding tissue-mimicking environment, allowing accurate spatial localization (fig. S15). This MRI visibility arises from the intrinsic hydrophilicity of the hydrogel matrix and the localized magnetic inhomogeneity introduced by embedded particles. Combined with its reprogrammable magnetic responsiveness and mechanical flexibility, the system offers a promising platform for soft biomedical devices that require noninvasive monitoring and controllable actuation.

Future designs could further integrate laser heating technology with the integrated system, enabling real-time reprogramming during

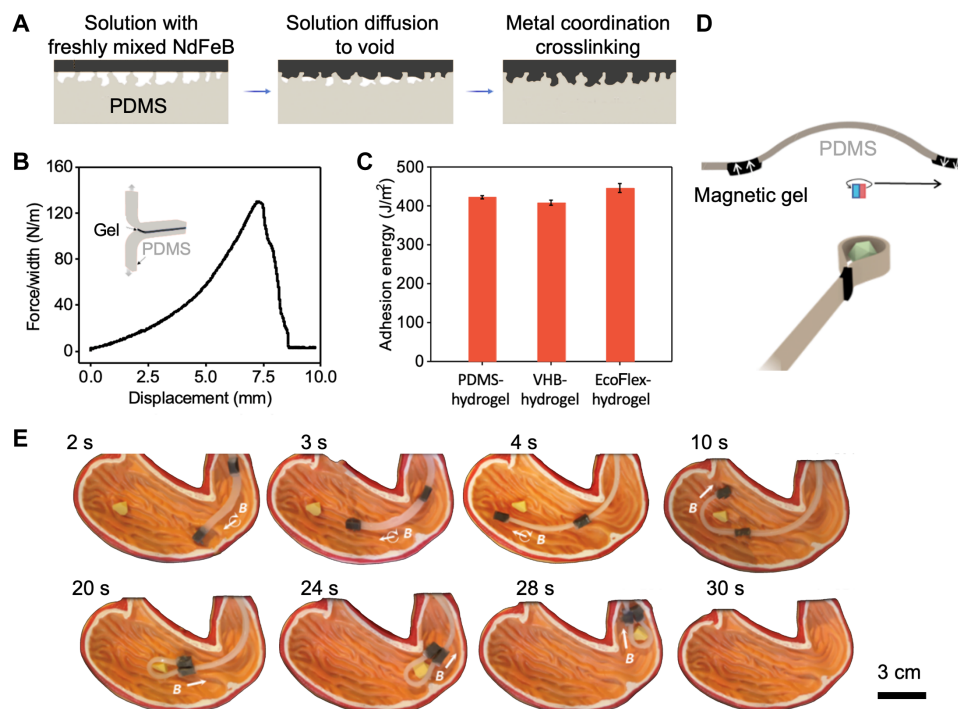


Fig. 6. Multifunctional soft device by integrating magnetic hydrogel with elastomer. (A) Illustration of adhesive integration of magnetic supramolecular gel to elastomer. (B) A representative peeling curve of the gel-PDMS sample with a sandwich-like structure. (C) Adhesive energy of the magnetic supramolecular gels to diverse elastomers. (D) Schematic design of magnetically steerable catheter with integrated hydrogel elements and a slender PDMS sheet for crawling and cargo manipulation. (E) Time-lapse sequences demonstrating the crawling locomotion, cargo trapping, and controlled cargo transfer using programmed magnetic fields.

operation. This would allow the device to dynamically adapt during navigation from confined channels to open spaces or seamlessly transition between grasping and transport phases, thereby enhancing operational flexibility in complex environments. This adaptability would be particularly valuable in minimally invasive medical applications, where spatial constraints limit the number of tools that can be simultaneously deployed. With this capability, a single device could perform multiple sequential tasks that would otherwise require several specialized instruments.

DISCUSSION

In summary, we have designed magnetic supramolecular hydrogels with reprogrammable patterning of magnetic domains by locally modulating the viscoelasticity of the gels. These gels are spontaneously formed after mixing the NdFeB particles with the P(AAc-co-AAm) solutions, in which the hydrolysis reaction of NdFeB particles leads to exposure and release of Fe^{3+} ions to form robust carboxyl- Fe^{3+} coordination complexes as physical cross-links of the copolymer chains. The metallosupramolecular gels have good mechanical performances and tunable magnetic properties. By harnessing the temperature-dependent viscoelasticity and, thus, the pinning force of the matrix, localized orientation of the magnetic particles under a modest magnetic field is realized by laser heating of the composite gel. Various patterns of magnetic domains are created in one hydrogel sheet by reversibly tuning the viscoelasticity and orientation of particles at laser-irradiated regions, enabling reversible and rapid shape morphing and motion upon magnetic stimulations. The magnetic response of the gel is further incorporated to nonresponsive elastomer to devise soft devices by adhesive integration, showing application potentials in soft robotics and biomedical devices. This work demonstrates that localized orientation of magnetic particles in hydrogels can be achieved by modulating the viscoelasticity and pinning force of the supramolecular network. Although this study focuses on the metallosupramolecular hydrogels containing NdFeB particles, the design strategy and working principle should be suitable to other soft materials with tunable viscoelasticity embedded with functional fillers responsive to external fields. This work opens possibilities for developing soft machines based on composite materials with programmable patterns of responsive particles and, thus, versatile functionalities.

MATERIALS AND METHODS

Materials

AAc (monomer), AAm (monomer), and potassium persulfate (KPS; initiator) were purchased from Aladdin Chemistry Co. Ltd. N,N,N',N' -tetramethylethylenediamine (TMEDA; reaction accelerator) was obtained from Sigma-Aldrich. Agarose gel (biological grade) was purchased from Thermo Fisher Scientific. NdFeB microparticles were purchased from Hongyu Technology Co. Ltd. VHB4905 (3M), PDMS (Dow Corning Sylgard 184), and EcoFlex (EcoFlex 00-30, Smooth-On) were received from Alibaba Co. Ltd. All reagents were used as received without further purification. Millipore deionized water (18.25 megohm-cm) was used throughout all the experiments.

Synthesis of NdFeB particle-containing P(AAc-co-AAm) hydrogels

Composite hydrogels were synthesized by mixing NdFeB magnetic microparticles with a viscous aqueous solution of P(AAc-co-AAm).

First, the P(AAc-co-AAm) solution was prepared via free radical polymerization. AAc (0.2 g, 1.0 wt %), AAm (1.8 g, 9.4 wt %), and KPS (0.02 g, 0.1 wt %) were dissolved in water (17.1 g, 89.5 wt %). The total polymer concentration was varied from 5 to 20 wt % to assess its effect on mechanical and magnetic properties, while the AAc molar ratio was also investigated in the range of 0.05 to 1.0. The precursor solution was degassed by bubbling argon for 20 min, and then prescribed amount of TMEDA was added to the solution, which was transferred to a reaction cell (table S1). The polymerization was completed at room temperature for 48 hours, producing a viscous solution of P(AAc-co-AAm). Then, NdFeB microparticles (0 to 30 vol %, relative to the volume of the solution) were added to the viscous solution. The mixture was homogenized using a vortex mixer for 2 min to ensure uniform dispersion. The resulting blend was poured into a nitrogen-flushed casting mold and heated to 50°C for 1 hour. During this process, the hydrolysis reaction between the oxidized surface of NdFeB particles and the carboxylic acid groups led to spontaneous gelation and formation of magnetic composite hydrogels.

Laser heating-assisted magnetic domain reorientation of the gel

Local viscoelasticity of the composite gel was modulated by laser heating at specific regions using a laser beam with a wavelength of 520 nm and a power intensity of 0.62 W/cm². Subsequent orientation of magnetic particles at the irradiated regions was realized under an external magnetic field with the strength exceeding the coercivity. Specifically, a magnetic hydrogel with 30 vol % of NdFeB particle was placed on a temperature-controlled stage of 25°C. The laser scanning speed was set to 1 mm/s; the scanning pattern was controlled by custom-built software. The hydrogel was exposed to the laser for 30 s to reduce the local viscoelasticity. During laser irradiation, an external magnetic field of 80 mT generated by an electromagnet was applied to orient the magnetic particles at the laser-heated regions. The direction of the external magnetic field was adjusted on demand to achieve the desired orientations. The gel sample was then cooled to room temperature under the applied magnetic field to fix the magnetic domain orientation.

To quantitatively assess the reprogramming capability, we define reprogramming efficiency as the ratio between magnetization achieved under a moderate magnetic field (150 mT) at elevated temperatures and maximum magnetization under a strong field (1.8 T). A reprogramming efficiency exceeding 90% is considered the threshold for successful magnetic domain reprogramming, as this level ensures complete reorientation of the magnetic particles in practical applications. This definition provides a standardized metric for evaluating temperature-dependent magnetic reprogramming performance.

Morphing and motion of patterned gels upon magnetic stimulations

Shape morphing of the magnetic gels was actuated using a custom-built electromagnetic system. Rectangular hydrogels containing 30 vol % of NdFeB particles (dimensions, 35 mm by 10 mm by 2 mm) were used to examine the morphing behaviors. An electromagnet equipped with 100-mm-diameter pole pieces generated uniform magnetic fields up to 200 mT across a 50-mm gap. The gel samples were suspended horizontally between the pole pieces using a nonmagnetic holder. Magnetic field direction was controlled through a programmable rotary stage with 0.1° angular resolution.

For reconfigurable shape demonstrations, the hydrogel sheets were first uniformly magnetized at 80 mT. Reprogramming orientation of local magnetic domain was achieved in the gel sheet using a 520-nm laser beam (spot size, 100 μm ; scanning speed, 1 mm/s) in the presence of a directional magnetic field. Shape morphing was realized by applying magnetic fields at specific orientations relative to the gel sample with programmed magnetic domains. The field strength was gradually increased from 0 to 200 mT while recording the deformation response. Time-lapse imaging captured the dynamic shape changes using a digital camera (60 frames/s). For cyclic testing, the gel sample underwent repeated actuation at 80 mT with alternating field orientations between 0° and 90°.

Locomotion of the magnetic gel was investigated by placing the gel on a horizontal glass surface lubricated with silicone oil to minimize the friction. For inchworm-like crawling, a rotating magnetic field (80 mT, 2 Hz) was applied perpendicular to the long axis of the slender hydrogel sheet. Pill bug-inspired rolling motion was achieved in the composite gel with patterned magnetic domains upon a rotating magnetic field (80 mT, 1 Hz). Directional control was assessed by varying the field orientation with 45° increments while maintaining constant strength of the magnetic field. All experiments of shape morphing and locomotion of the gels were performed at room temperature (25°C) unless specified.

Characterizations

Tensile tests were performed to characterize the mechanical properties of the hydrogels using a commercial tensile tester (3343, Instron). Dumbbell-shaped samples with a gauge width of 4 mm, a gauge length of 12 mm, and a thickness of 1 mm were cut from the hydrogel sheets and directly used for tensile tests with a stretch rate of 100 mm/min. Each hydrogel was examined by at least three parallel tests. The Young's modulus (E) was calculated from the slope of the stress-strain curve with a strain below 5%. Rheological measurements were conducted to monitor the sol-to-gel transition during the gel synthesis, as well as the modulated viscoelasticity of the gel at varying temperatures, using a DHR-2 rheometer (TA Instruments). The samples were transferred to the sample stage of the rheometer. Then, the top parallel plate with a diameter of 25 mm was lowered to achieve a 1-mm gap. The measurements were performed with the shear rate ranging from 1 to 1000 s^{-1} .

FTIR spectra of the gels were recorded with a Nicolet iS 10 FTIR spectrometer in the region of 4000 to 400 cm^{-1} with a resolution of 2 cm^{-1} . SEM observations of magnetic hydrogels were performed using a Hitachi S4800 field emission scanning electron microscope at an acceleration voltage of 3 kV. The samples were prepared by freeze-drying the hydrogels and then being fractured in a liquid nitrogen. The fractured surface was coated with a thin layer of gold to avoid charging effects.

Magnetization measurements were carried out using a Superconducting Quantum Interference Device (MPMS-XL-5). The magnetic hysteresis loops of NdFeB particles and composite gels were measured at room temperature with the maximum applied magnetic field of 2 T. Magnetization-mediated mechanical properties of the magnetic gels were accomplished by placing the sample in the center of an electromagnet GMH X080 (Magnet-Schultz) equipped with a programmable power supply (Keysight 2901A). The magnetic hydrogel was attracted to the electromagnet using a variable magnetic field induced by varying voltage between 0 and 50 V, corresponding to a maximum current of 2.21 A. The highest field

strength used on the pole of the electromagnet was 0.63 T. The magnetic flux density (B) of the magnetized gel was measured by a gauss meter (CH-1800, Beijing Cuihai Jiacheng Magnetic Technology Inc.), which was driven by a moving stage to measure the degree of B in different orientations. To evaluate MRI visibility, the hydrogel-integrated device was embedded in 5 wt % of agarose gel to mimic the tissue implantation environment. MRI was performed using a 1-T M2 system (Aspect Imaging) equipped with a 25-mm length-by-23-mm diameter radio frequency coil. T2-weighted images were acquired in the coronal plane with the following parameters: repetition time (TR) = 4000 ms, echo time (TE) = 62.9 ms, field of view = 30 mm by 30 mm, number of excitations (NEX) = 15, resolution = 0.25 mm by 0.25 mm by 1 mm, and excitation angle = 100°. Image visualization was conducted using AMIDE 1.0.5.

Interfacial bonding strength between the magnetic gels and the elastomers was examined by peeling tests at room temperature. In a typical adhesive integration of the two materials, 2 ml of fresh mixture of NdFeB particles and P(AAc-co-AAm) solution was coated on one surface of the elastomer (dimensions, 70 mm by 15 mm by 2 mm). After 2 min, the free surface of the mixture was gently pressed by another piece of elastomer. The sandwich-like sample was kept for 1 hour to allow the gelation process to form robust interfacial bonding. Peeling tests were performed at 180° mode, with a displacement rate of 100 mm/min. The peeling force and the displacement were recorded. The adhesion energy was calculated as the maximum force divided by the width of the sample.

Supplementary Materials

The PDF file includes:

Figs. S1 to S15

Table S1

Legends for movies S1 to S3

Other Supplementary Material for this manuscript includes the following:

Movies S1 to S3

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