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Reprogrammable Ferromagnetic Domains for Reconfigurable Soft Magnetic Actuators

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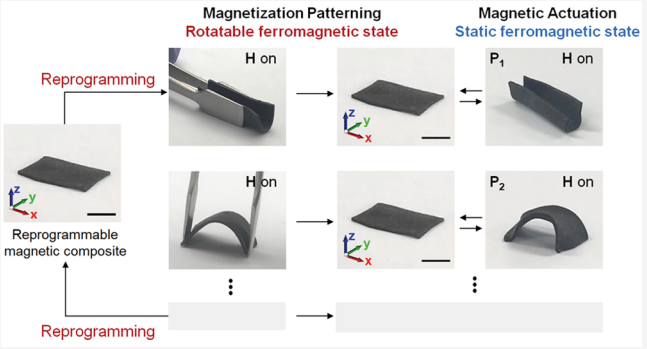
ABSTRACT: Soft magnetic materials have shown promise in diverse applications due to their fast response, remote actuation, and large penetration range for various conditions. Herein, a new soft magnetic composite material capable of reprogramming its magnetization profile without changing intrinsic magnetic properties of embedded magnetic particles or the molecular property of base material is reported. This composite contains magnetic microspheres in an elastomeric matrix, and the magnetic microspheres are composed of ferromagnetic microparticles encapsulated with oligomeric-PEG. By controlling the encapsulating polymer phase transition, the magnetization profiles of the magnetic composite can be rewritten by physically realigning the ferromagnetic particles. Diverse magnetic actuators with reprogrammable magnetization profiles are developed to demonstrate the complete reprogramming of complex magnetization profile.



KEYWORDS: *reprogrammable magnetization, soft magnetic composite, soft magnetic actuator, smart materials*

Soft active materials have evolved to achieve mechanical functionalities beyond those of their rigid counterparts for development of active components in a wide range of applications.[1−4] Among these materials, magnetic soft materials have become particularly attractive because they can be remotely controlled and operated rapidly in many different media, since magnetic fields penetrate a wide range of environments.[5−9] Actuation of magnetic soft material arises from spatiotemporal interactions between the applied magnetic field, environment, and programmed magnetization.[10−15] These magnetic soft materials are usually composed of hard magnetic particles or discrete magnets incorporated in a base material, where the desired magnetization pattern direction and magnitude can be imposed by orienting or aligning these magnetic fillers, which have intrinsic magnetic moments. However, magnetic fillers are usually physically confined in the soft matrix after fabrication is completed, and a limitation arises because actuation is constrained by the permanent programmed magnetization patterns. Thus, a soft magnetic composite with a facile remagnetization scheme could be used to make reconfigurable functions of soft devices for a wide range of applications. In this regard, remagnetization of a soft magnetic composite can be realized by changing both the physical arrangement of magnetic particles and the intrinsic magnetic properties of hard magnetic fillers.

Remagnetization in a hard ferromagnetic material can be realized by applying a strong magnetic field above its coercive field[10,16,17] or by increasing temperature to decrease the intrinsic coercivity,[18,19] at which point the magnetic domain structures in the magnetic particle should be altered. Remagnetization in hard magnetic material is still affected by its crystallographic anisotropy, and temperature changes to reduce coercivity may have undesirable effects on the base material. Thus, the remagnetization condition crucially depends on the intrinsic properties of incorporated magnetic particles.

With regard to the physical arrangement of magnetic particles, magnetic particles were embedded in smart polymers, such as shape memory polymer, thermally responsive hydrogel, liquid crystal elastomer, etc., as a base material to provide reconfigurable functions to the composite materials in several studies.[20−24] Their morphologies can be changed into arbitrary shapes or alternate between two shapes using other stimuli related to the base polymer molecular properties; nevertheless, subsequent magnetic actuation of the reshaped structure is still tied to its morphology and initial magnetization profile, because the relative positions and alignments of magnetic

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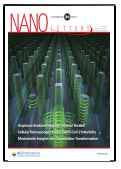

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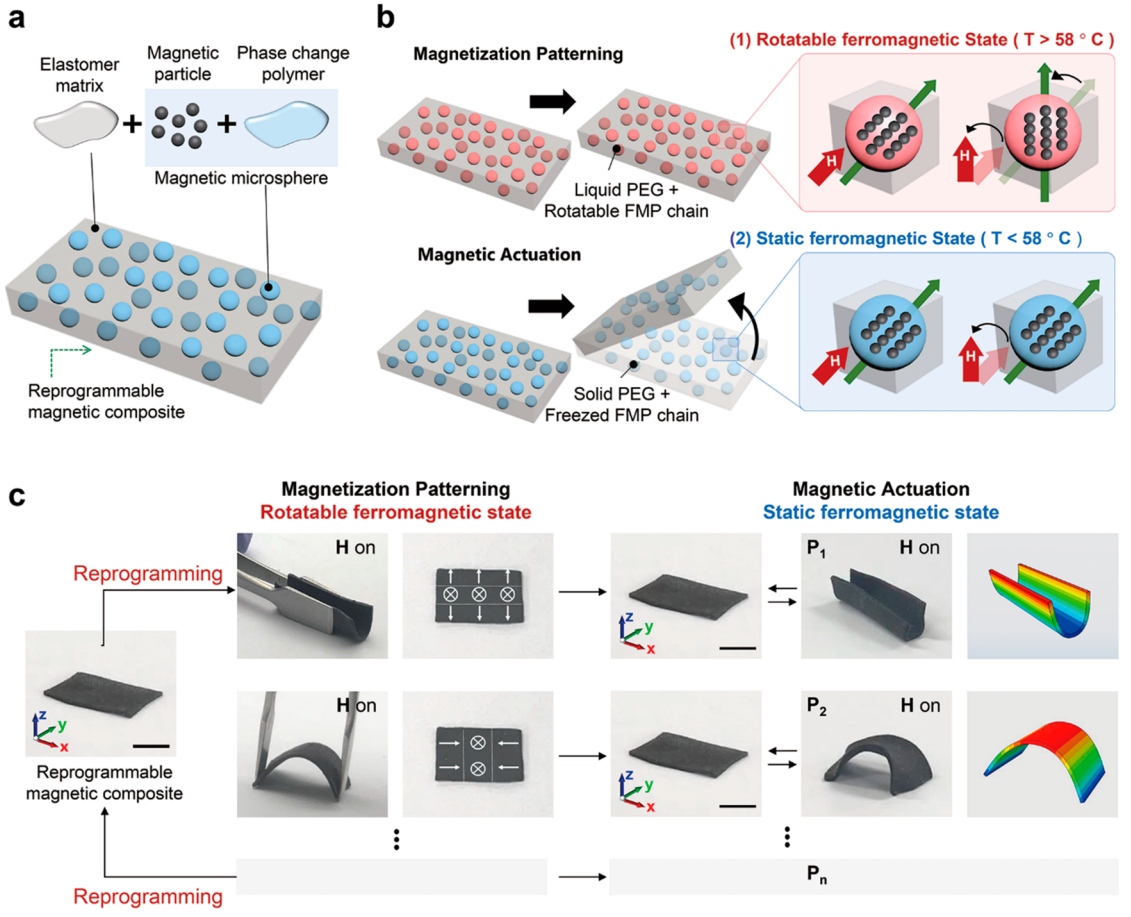

Figure 1. Design and mechanism of a reconfigurable magnetic composite. (a) Composition of a reconfigurable magnetic composite. The proposed magnetic composite has a hierarchical structure comprising magnetic microspheres within an elastomeric matrix, where the microsphere is composed of magnetic microparticles (NdFeB) and encapsulating PEG oligomer. (b) Rotatable and static ferromagnetic states of magnetic microspheres. In the rotatable ferromagnetic state, above the melting temperature of the PEG, the magnetic particle chains can freely rotate. However, in the static ferromagnetic state, below the melting temperature of the PEG, the magnetic particle chains are fixed in the solidified PEG, maintaining the programmed ferromagnetic domain patterns. (c) Reconfiguring process of a reprogrammable magnetic composite. This structure transforms into various shapes under the magnetic field applied perpendicular to the plane of the structure (z-axis direction). 'P' represents 'programmed magnetization profile' and 'n' indicates the number of programming processes of the magnetization profile. All scale bars are 10 mm.

particles, which dictate magnetization in the base material, are hardly changed. On the other hand, complete magnetization change was achieved in ferrofluid through reversible para-to-ferromagnetic transformation by the interfacial jamming of magnetic particles.[25] In this case, magnetization is retained by controlling the physical arrangement of magnetic particles as the magnetic particles freely rotate when the ferrofluid droplet is not jammed. However, these realignments of magnetic particles occur only in the liquid environment, which limits the range of applications. Here, we present a simple composite material to effect the remagnetization by phase transition of polymer encapsulating the ferromagnetic particles without changing intrinsic magnetic properties of embedded magnetic particles or the molecular property of the base material.

Our reprogrammable magnetic composite is composed of spatially separated magnetic microspheres embedded in the elastomeric matrix (Ecoflex 00-30). Each magnetic microsphere consists of magnetizable neodymium−iron−boron (NdFeB) microparticles, with an average size of 5 μm, encapsulated in oligomeric polyethylene glycol (PEG) (Figure 1a). Solid-to-liquid phase transition of this encapsulating oligomer allows us to rearrange the magnetic particles in the microspheres, and the realignments of magnetic particles change magnetization directions in the magnetic microsphere (Figure 1b). Therefore, we can rewrite magnetization patterns in the soft material by heating the structure to induce the phase transition of the microspheres and shaping it into any desired form under external magnetic fields while it cools. In the rotatable ferromagnetic state, the magnetic particles in the microspheres can rotate according to the applied magnetic field direction due to the mobility of the particles in a liquid state of encapsulating polymer. On the other hand, in the static ferromagnetic state, the magnetic composite is actuated when it is exposed to the rotating magnetic field. In this state, magnetic particles cannot be rotated due to the solid state of the encapsulating polymer, and the fixed alignments of the magnetic particle make magnetic torque in response to an external magnetic field.

The magnetic composite was prepared as follows. First, we mixed the magnetic particles with melted PEG oligomers in a 7:3 weight ratio and ground them into diameters between 53 and 150 μm under liquid nitrogen to create the magnetic microspheres. Then, these microspheres were well-blended with silicone elastomer at a 1:1 weight ratio and cured at 100

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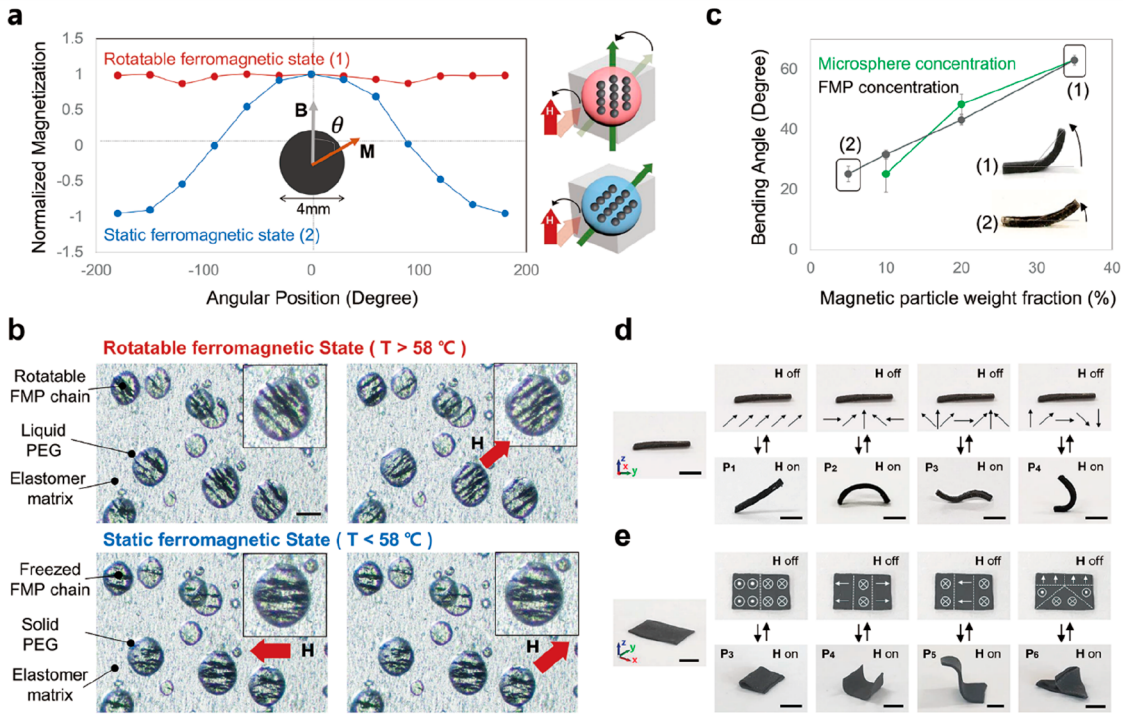

Figure 2. Magnetic characterization, microscopic images, and various shape changes of reconfigurable magnetic composites. (a) Magnetization of magnetic composite for static and rotatable ferromagnetic states. Measurements were conducted at various sample angles with respect to the applied magnetic field. In the static ferromagnetic state (27 °C), residual magnetization changes depending on the angle. However, in the rotatable ferromagnetic state (67 °C), magnetization remains constant regardless of angle. (b) Microscopic images of embedded magnetic particles in rotatable and static ferromagnetic states. Scale bar is 100 μm (c) Reprogrammable transformations for a linear magnetic composite. Actuation capability with respect to NdFeB particle concentration was measured by the magnetic composite bending angle under the external magnetic field. (d) Various transformations for a linear magnetic bar with continuous magnetization pattern under applied magnetic fields. All scale bars are 5 mm. (e) Various transformations for a magnetic membrane with continuous magnetization pattern under applied magnetic fields. All transformation is under the magnetic field of 100 mT applied perpendicular to the plane of the structure. All scale bars are 10 mm.

°C for 10 min. We chose PEG 4000, a commonly employed plasticizer,[26] as the encapsulating material since it has a low melting point of 58 °C and, more importantly, low melt viscosity compared to conventional polymers, giving NdFeB microparticles mobility when melted.[27]

Figure 1c illustrates the process of reconfiguring our reprogrammable magnetic composite by reprogramming its magnetization pattern. The magnetic composite (20 mm × 12 mm × 0.6 mm) which contains 35 wt % magnetic particles transforms into different shapes under the same stimuli, as a demonstration of the ability to reprogram the magnetization of the magnetic composite. The reprogramming process proceeds as follows. First, to program (P_1) the ferromagnetic domain patterns, we heat the composite to 100 °C for 3 min, above the melting temperature of oligomeric-PEG, to accelerate heat transfer and manually morph the composite into the desired shape under a 300 mT magnetic field for 3 min. In the rotatable ferromagnetic state, all magnetic particles align along the applied magnetic field direction, and their realignments are fixed as the sample cools, representing the reprogrammed ferromagnetic domain pattern. In the static ferromagnetic state, the composite transforms its shape into the programmed shape by balancing magnetic torques responding to the 100 mT external magnetic field generated by a permanent magnet and mechanical restoring forces. As the composite transformation occurs by using the permanent magnet, a magnetic field gradient which generates the force that pulls the composite into the place and friction that holds it in place are also considered as important factors to make a final transformation. Thus, they rapidly return to their original shape upon removal of the applied external magnetic field. Next, we reprogram (P_2) the magnetization pattern of this composite by heating it to reach the rotatable ferromagnetic state and manually shaping the structure to rearrange the magnetic particle alignments under the magnetic field. Finally, the magnetic composite transforms into a different shape as reprogrammed by applying an external magnetic field. Their actuation is simulated by finite element analysis under the experimental conditions, including magnetic moment density, mechanical moduli, and applied magnetic field, to anticipate the transformed shapes and actuation sequence according to the designed magnetization pattern. By repeating these steps (P_n), the magnetic composite can be morphed into any desired shape from a single sample, without requiring special equipment or additional fabrication process.

To verify that the magnetic composite magnetization can be reprogrammed in the rotatable ferromagnetic state, we measure the magnetic moment density in samples at various angles relative to the external magnetic field applied by the vibrating sample magnetometer. We then compare the normalized magnetization angular profiles from static and rotatable ferromagnetic states, as shown in Figure 2a. The sample is circular with a 4 mm diameter and 1.3 mm thickness and contains 35 wt % magnetic particles. In the static ferromagnetic state, residual magnetization at 27 °C under magnetic field of 1 mT decreases as the angle changes because

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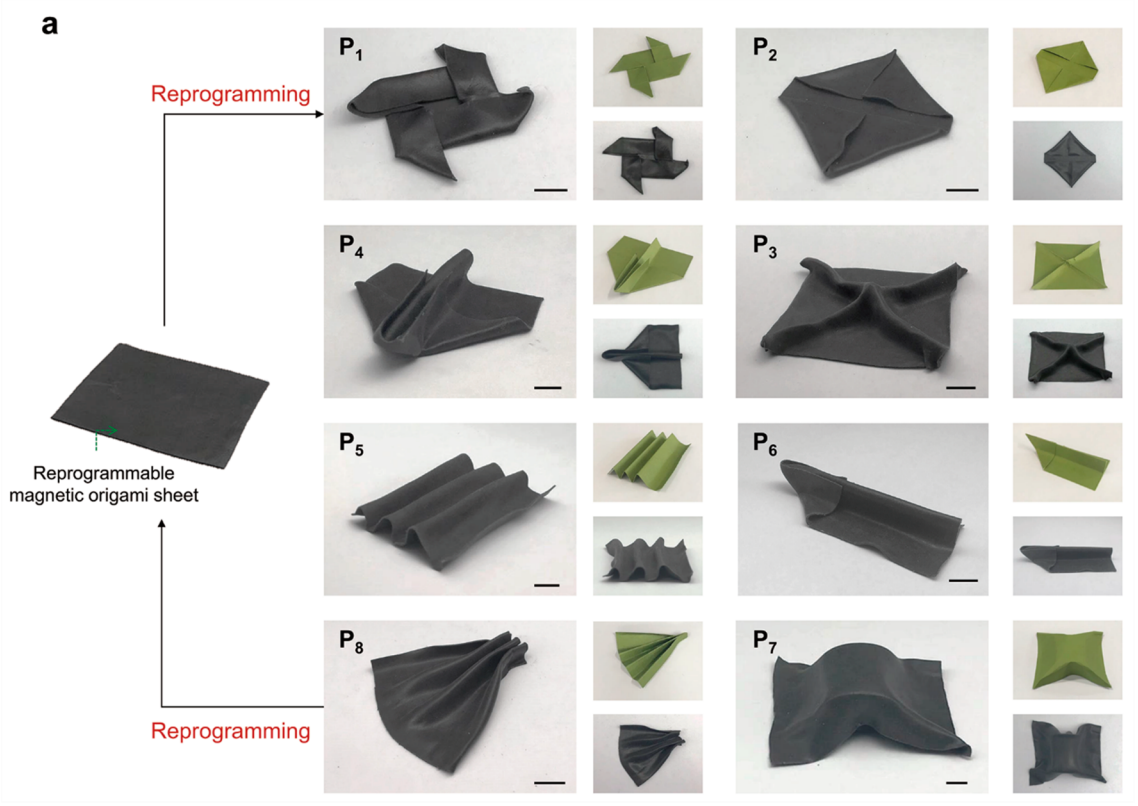

Figure 3. Various transformations of a reconfigurable soft magnetic origami sheet with a continuous magnetization pattern. Photographs and schematics for various transformations of a magnetic origami sheet using a continuous magnetic membrane. The magnetic origami sheet deforms its entire structure into desired shapes under the magnetic field of 100 mT applied perpendicular to the plane of the structure. All scale bars are 10 mm.

the relative directions and positions of all magnetic particles with their intrinsic magnetic moments are fixed. The maximum magnetization value is 14.65 kA/m when the sample magnetic moment direction is parallel to the external magnetic field. However, in the rotatable ferromagnetic state at 67 °C, above the melting temperature of the oligomeric-PEG, the magnetization remains the same at 32 kA/m regardless of the sample angle to the external magnetic field of 300 mT, which is same as the applied magnetic field intensity to reprogram the magnetization pattern in the experiment. Since the magnetic particle chains can freely rotate along the field, it is possible to redirect their magnetic moments to follow the applied magnetic field direction while the PEG oligomer remained in liquid form.

Microscopic images present that the reprogrammabiltiy of our composite arises from the phase transition of the PEG oligomer. As shown in Figure 2b, the magnetic microspheres can be regarded as discrete magnetic domains uniformly dispersed throughout the material, and these ferromagnetic domains can be rearranged at moderate temperatures. Here, the sample contains 5 wt % magnetic particles in total; the elastomeric composite contains 50 wt % magnetic microspheres, and each microsphere includes 10 wt % magnetic particles. When the PEG oligomers encapsulating the magnetic particles transform to low viscosity liquid above their melting temperature (58 °C), the self-assembled magnetic particle chains within the microspheres can freely rotate to align their intrinsic magnetic moments along the applied external magnetic field, which is described as a rotatable ferromagnetic state. On the other hand, when the PEG oligomers become solid below the melting temperature, the realigned magnetic particle chains are physically fixed in the base material, maintaining the programmed pattern of ferromagnetic particles (Movie S1).

Magnetic responsiveness of magnetic composites depends on NdFeB microparticles concentration embedded in the composites. Since individual NdFeB microparticles act as separate and rotatable ferromagnetic domains in the base matrix, the concentration directly impacts the apparent magnetization intensity of the magnetic composite, affecting the magnetic force exerted on the magnetic composite for actuation. Therefore, as shown in Figure 2c, we assess the effect of magnetization intensity on actuation capability by measuring the bending angles of them under the external magnetic field, with respect to NdFeB microparticle concentrations. We control the magnetic microsphere concentration from 14, 29, and 50 wt % while maintaining the magnetic particle concentration at 70 wt % in the microspheres. We also control the magnetic particle concentration in magnetic microspheres from 10, 20, 40, and 70 wt % with fixed magnetic microsphere concentration in the elastomeric matrix at 50 wt %. Thus, the overall magnetic particle concentration in the magnetic composites ranged from 5 to 35 wt %. The bending angle of the magnetic composites increases almost linearly with increasing NdFeB microparticle weight fraction. Furthermore, we repeat the process to program magnetization directions in these samples and measure their bending angles to investigate the functional reliability of this procedure. Their actuation performances are stable over 50 reprogramming trials (Figure S2).

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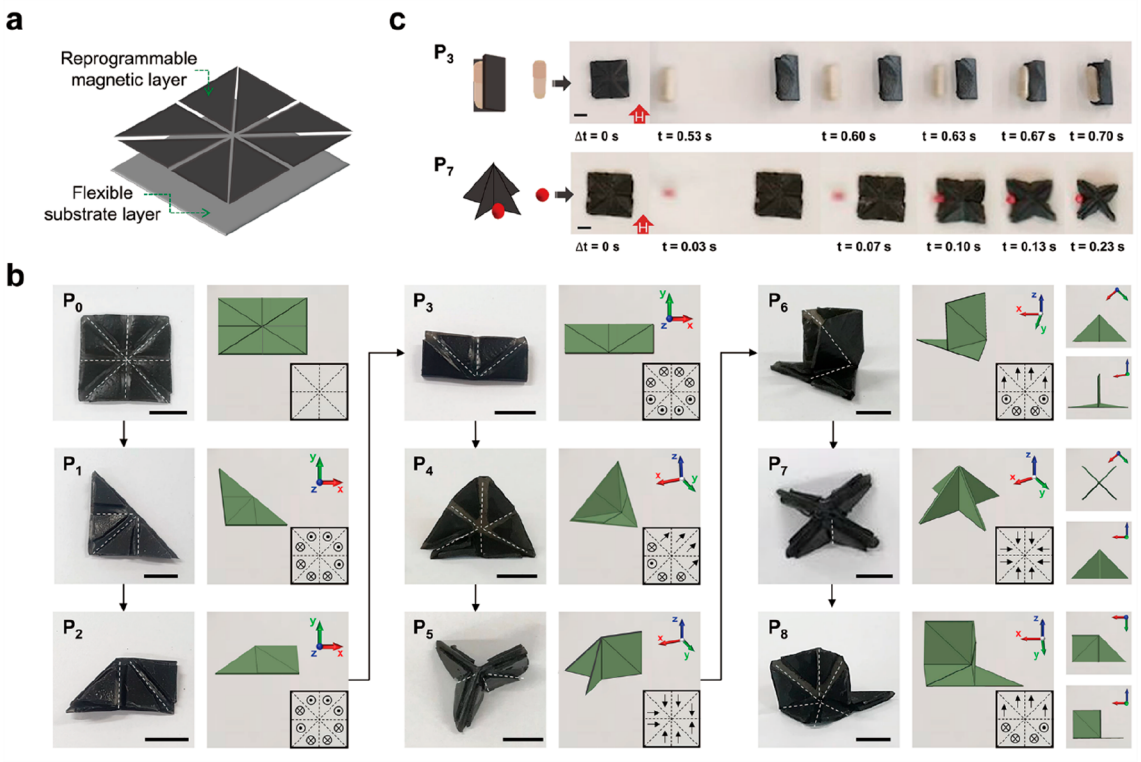

Figure 4. Various transformations a reconfigurable soft magnetic origami sheet with discrete magnetic parts on a substrate. (a) Schematics for various transformations of a magnetic origami sheet with eight distinct actuating parts on a PVC substrate. (b) Photographs and schematics for various transformations of a magnetic origami sheet with eight distinct actuating parts on a PVC substrate. The origami sheet demonstrates two-dimensional planar origami, including triangle, rectangle, and complex three-dimensional structures under the magnetic field of 100 mT applied perpendicular to the plane of the structure. (c) Grasping various fast-moving objects using the magnetic origami sheet. All scale bars are 10 mm.

To demonstrate the capability of writing nonuniform and continuous magnetization profiles in a soft material, we present a linear beam structure (13 mm × 2 mm × 1.2 mm) that transforms into various programmed shapes (Figure 2d). This structure can be programmed and reprogrammed to provide continuous magnetization profiles along its body. The magnetization profile for this composite is designed by heating the structure, morphing the soft body into the desired shape, and subjecting it to a 300 mT magnetic field to rewrite the magnetization profile as it cools to fix the reprogrammed ferromagnetic domain pattern. After cooling, interactions between an external magnetic field of 100 mT applied to the perpendicular to the plane and patterned magnetization produces spatially varying magnetic torques. The composite deforms its entire structure into the programmed shape, such as tilted 'I', 'C', single wavelength, half-wavelength harmonic shape, etc., by balancing magnetic torques and mechanical restoring forces (Movie S2).

Furthermore, the deformation of our magnetic composite can be further extended to a two-dimensional planar structure (20 mm × 12 mm × 0.6 mm) beyond the one-dimensional linear structure. The diverse shape deformations of a continuous magnetic membrane which has a nonuniform and continuous magnetization profile are shown in Figures 1c and 2e and in Movie S3. The magnetization pattern of this magnetic membrane is also reprogrammed simply by morphing the structure as desired under heat and cooling it. Then, it transforms its entire structure into designed shapes under the magnetic field of 100 mT applied perpendicular to the plane of the structure. Origami-inspired design has been noticed in both natural and artificial systems because this design principle provides a novel approach to transform a simple structure into diverse complex structures while being lightweight and compact.[28,29] Thus, we utilize our continuous two-dimensional magnetic composite membrane as a magnetic origami sheet without a predetermined crease structure as shown in Figure 3. This magnetic membrane (64 mm × 54 mm × 0.6 mm) transforms into various shapes, such as a pinwheel, airplane, ruffle, bullet, etc., as the continuous magnetization profiles are reprogrammed. These complex transformations are achieved by simply morphing the membrane during the magnetization reprogramming process. Our material system is suitable to make fully functional and reprogrammable origami, like a real paper origami sheet, because a continuous membrane without any mechanical compartmentalization allows us to provide the universality to magnetic actuators that can realize fast and numerous shape changes and large deflection.

In addition, to utilize our magnetic composite as a functional layer, we develop a magnetic origami sheet with discrete magnetic parts on a flexible substrate layer.

These magnetic origami sheets are fabricated by cutting our magnetic composite into fragments and attaching them to the various flexible substrate layer (Figures S3−S5). We demonstrate the magnetic origami sheet (25 mm × 25 mm × 1.2 mm) with eight isosceles right-angled triangle-shaped magnetic parts on PVC substrate in Figure 4a. As the substrate layer can function as a predetermined mechanical crease which makes the magnetic torque dominant at the interface between each magnetic fragment, this planar two-dimensional structure can be transformed into any three-dimensional shape with high

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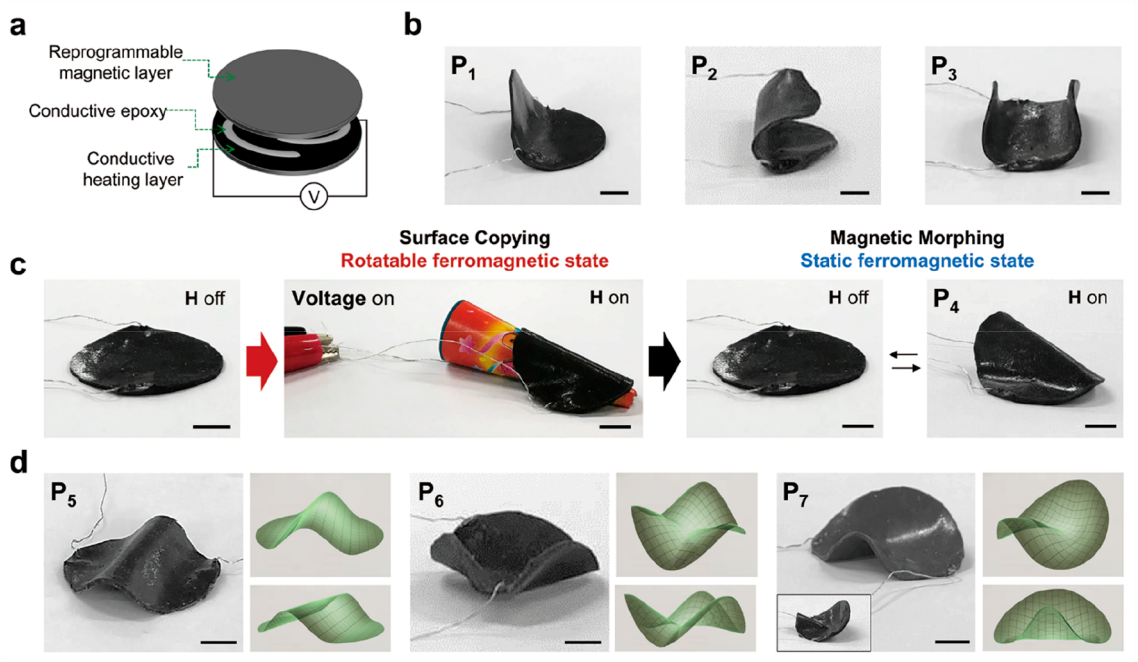

Figure 5. *In situ* reprogramming magnetization profile and various shape transformations with nonuniform curvatures. (a) Schematic of a reprogrammable soft magnetic actuator with reprogrammable magnetic layer and conductive heating layer. (b) Various transformations of a reprogrammable soft magnetic actuator based on *in situ* reprogramming of magnetization profile. (c) Copying the surface curvature of an arbitrary object using the reprogrammable soft magnetic actuator. (d) Photographs and schematics for various reprogrammable morphing with Gaussian curvatures. All transformation is under the magnetic field of 100 mT applied perpendicular to the plane of the structure. All scale bars are 10 mm.

accuracy through the simple magnetization reprogramming and magnetic actuation process. Figure 4b shows eight different transformations, including a triangle, rectangle, trapezium, etc., by repeatedly reprogramming the magnetization profile (Movie S4). Their magnetization profile enables a magnetic field to independently actuate each part of the structure to programmatically determine the final configuration. The schematics of these final configurations are displayed in different views such as 3D, top, side, and front views. Among these, two reprogrammed structures are used to demonstrate the capability of grasping various objects with different shapes (Figure 4c). Taking advantage of the fast response upon external magnetic field, the magnetic origami sheet quickly transformed into the programmed shape within 1 s, providing a space to fit spherical or ellipsoidal fast-moving objects. The result confirmed that our magnetic origami sheet is suitable for fabricating various device components, such as grippers or carriers, and more importantly is able to have its functionality redesigned as desired.

Recently, mimicking complex surface curvatures with soft material actuation has been demonstrated[30,31] as living organisms can realize complex nonuniform curvatures on their surfaces for their development, transformation, camouflage, etc. Thus, we develop a reconfigurable soft magnetic actuator capable of transforming into the shapes with nonuniform curvatures *in situ*. As shown in Figure 5a, the reprogrammable soft magnetic actuator (thickness = 1.0 mm, diameter = 40 mm) is composed of a reprogrammable magnetic layer (thickness = 0.6 mm, diameter = 40 mm) and conductive heating layer (thickness = 0.4 mm, diameter = 40 mm). To reprogram the magnetization profile *in situ*, we apply 30 V to the actuator for 5 min, increasing the temperature of the conductive layer up to 70 °C through Joule heating. The heat is conveyed to the reprogrammable magnetic layer, which changes the magnetic phase of the magnetic composite layer into the rotatable ferromagnetic state. Then, the magnetization pattern is reprogrammed as we morph the sample under the external magnetic field and is memorized as the temperature cools by removing the application of voltage. To show the feasibility of *in situ* reprogramming, we demonstrate several transformations of the magnetic actuator as shown in Figure 5b. This magnetic actuator is able to copy the surface curvature of an arbitrary object or memorize the morphed state as the magnetization profile at the rotatable ferromagnetic state is memorized (Figure 5c). Finally, we also demonstrate several reprogrammable deformations with nonuniform curvatures (Figure 5d). We anticipate that this soft magnetic actuator can be utilized as a surface copying device or *in situ* camouflage skin for soft robotic applications because it can memorize the morphed state and reproduce it when the simple magnetic stimulus is applied.

In conclusion, our material architecture introduces a novel approach to reprogram the magnetization profile in a soft magnetic composite by controlling ferromagnetic particle arrangements using the solid−liquid phase transition of the polymer encapsulating the particles. Using this method, we can freely program heterogeneous magnetization in the elastomeric base material and consequently transform the material into a programmed structure as many times as desired. This enables not only the design of magnetization transformations for magnetic soft materials but also the reprogramming of the actuation of magnetic soft composite materials. Therefore, we expect that this kind of soft magnetic composite with reprogrammable magnetization pattern could be used to make reconfigurable soft material systems for a wide range of applications, including applications in biomedical engineering, flexible electronics, and soft robotics.

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■ ASSOCIATED CONTENT

Supporting Information

The Supporting Information is available free of charge at https://pubs.acs.org/doi/10.1021/acs.nanolett.0c01418.

(Movie S1) Microscopic video of a magnetic composite in its rotatable and static ferromagnetic state (MP4)
(Movie S2) Various transformations of a linear beam structure with a continuous magnetization profile (MP4)
(Movie S3) Various transformations of a magnetic membrane with a continuous magnetization profile (MP4)
(Movie S4) Various transformations of a magnetic origami sheet with eight discrete magnetic parts (MP4)
Materials and methods, Figures S1−S7, and movie descriptions (PDF)

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Notes

The authors declare no competing financial interest.

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