Facile Manufacturing Route for Magneto-Responsive Soft Actuators

Julia A. Carpenter, Thomas B. Eberle, Simone Schuerle, Ahmad Rafsanjani, and André R. Studart*

Magnetically driven soft actuators are unique because they are fast, remote-controlled, conformal to rigid objects, and safe to interact with humans. Despite these multiple functionalities, a broader utilization of such actuators is hindered by the high cost and equipment-intensive nature of currently available manufacturing processes. Herein, a simple fabrication route for magneto-responsive soft actuators is described using cost-effective and broadly available raw materials and equipment. The method utilizes castable silicone resins that are loaded with magnetic particles and subsequently magnetized under an external magnetic field. The experimental investigation of silicone-based composites prepared with particles of distinct chemistries, sizes, and morphologies enables the identification of the raw materials and magnetization conditions required for the process. This leads to functional soft actuators with programmable magnetic patterns that are capable of performing pick-and-place, lifting, catching, and moving tasks under the remote action of an external magnetic field. By removing manufacturing hurdles associated with costly raw materials and equipment, the proposed approach is expected to facilitate the design, implementation, and exploitation of the unique functionalities of magneto-controlled soft actuators in a wider number of applications.

1. Introduction

Soft actuators have opened compelling new opportunities in the fields of manufacturing, robotics, and medicine.\(^1\) Because of their conformal and mechanically compliant nature, actuators made of soft materials allow for safe interactions of robots with humans and provide an attractive platform to interface flexible electronics with human skin and tissues.\(^2\text{-}^5\) Potential applications that can benefit from these features include robots for manipulation of delicate objects,\(^6\) exoskeletons for motion rehabilitation, medical prostheses,\(^7\) surgical aid devices,\(^8,9\) and intelligent reconfigurable matter in general.\(^10\) In many of these applications, it is desirable to minimize the energy required for actuation, enhance actuation speed, and enable untethered autonomous motion.

Among the several possible types of devices and actuation modes,\(^11\text{-}^20\) magnetically responsive actuators are particularly interesting, because they are fast, contactless, and are driven by magnetic fields that can be safely used in humans.\(^21\) Contactless control allows for the manipulation of untethered devices in confined spaces, which is crucial for minimally invasive medical devices.\(^22\) Making soft objects magneto-responsive is also a promising approach in the ongoing efforts to create devices with complex output motion driven by a simple single input.\(^23\) For example, recent research has shown that the soft objects with tunable local magnetization patterns can undergo complex motions, such as crawling, rolling, and jumping when activated with an external magnetic field. These magneto-responsive soft materials have so far been produced in simple planar geometries using NdFeB microparticles that are pre-magnetized using magnetic fields higher than 1 T.\(^19,24,25\) Although the magnetization of NdFeB particles using these approaches prevents de-magnetization of the actuator, the...
strong magnetic fields required for magnetization increase the costs of the process and may prevent broader exploitation of these materials.

Several types of magnetic particles have been previously used for the fabrication of magneto-responsive soft materials. Superparamagnetic iron oxide nanoparticles (SPIONs) are an attractive option in applications requiring materials that only become magnetized in the presence of an external magnetic field. By aligning such nanoparticles in chains, it is possible to create anisotropic structures with the orientation-dependent magnetic properties needed for soft actuation. In addition to the iron oxide and NdFeB particles explored so far, other magnetic materials may be suitable for the fabrication of magneto-responsive soft actuators but have not yet been considered for this purpose. For example, strontium hexaferrite (SrFe$_{12}$O$_{19}$) could be an interesting alternative to currently employed magnetic particles, because it displays the hard magnet characteristics of NdFeB while also being prone to magnetization using relatively low magnetic fields. Besides chemical composition, the size, morphology, and aspect ratio are also known to affect the magnetic properties of particles and, thereby, the response of particle-laden composites. The availability of magnetic particles of different chemistries, sizes, and morphologies provides room for the design of magneto-responsive soft actuators that fulfill specific application needs. Considering that the cost and access to magnetization equipment are often a limiting factor, the development of simple manufacturing processes that rely on easily accessible tools may broaden the range of applications and potential end users of magnetically driven soft actuators.

In this article, we propose a simple method to fabricate magneto-responsive soft actuators that are fast, scalable, and complex-shaped using readily available manufacturing tools. To this end, silicone-based composites containing magnetic particles of different sizes, morphologies, and chemical compositions are first developed and systematically studied in terms of magnetic and mechanical properties. Particles amenable to magnetization using widely available magnets are then further characterized and used to fabricate soft grippers programmed with distinct magnetization patterns. Using an available model that predicts the magneto-response of soft materials, we correlate the morphing behavior of the soft grippers with the underlying magnetization pattern generated during manufacturing. Finally, our particle-laden silicone resins are used to fabricate a variety of functional soft actuators that can be remotely controlled by an external magnetic field.

2. Results and Discussion

2.1. Manufacturing and Magnetization

Magneto-responsive soft actuators are manufactured by simply mixing magnetic particles with a two-part silicone resin and casting the resulting viscous paste into a mold (Figure 1a). The formation of a percolating network between the particles increases the viscosity of the resin and prevents free-flowing of the resulting paste. Therefore, the casting procedure requires compression of the paste to fill the molds completely and to produce a smooth surface finish.

The magnetization of the soft actuators is carried out during or after curing of the silicone resin using permanent magnets positioned around the mold (Figure 1b). Different magnetization patterns can be generated on the actuator depending on the number and position of magnets used. This procedure is quite efficient and allows for the production of a variety of soft actuators with distinct magnetization patterns.

![Figure 1](image1.png)

Figure 1. Scheme illustrating the fabrication, magnetization, and actuation of silicone parts loaded with magnetic particles. a) Soft objects are fabricated by mixing the magnetic powder with a two-part silicone resin. The resulting viscous paste can be directly cast into molds to create actuators with well-defined geometries. b) Magnetization of the actuators is carried out by placing commercial neodymium magnets at specific positions during or after curing of the silicone resin for 4 h. c) The actuators are activated using an external magnetic field. Motion results from the interplay between the applied magnetic field, the magnetized domains of the actuator, and the gravitational forces.
The markedly larger coercivity of $\text{M}_1$ values and $\text{M}_1$ particles is due to their shape. A magnetic magnetization values obtained with the spherical magnetite upon application of magnetic magnetic dipoles of these particles can be more easily oriented shaped magnetite and the strontium ferrite indicate that the control over the magnetic and mechanical properties of the particle-laden silicone is crucial to describe and predict the response of the soft actuators to remotely applied magnetic stimuli.

2.2. Selection of Magnetic Particles

The versatility of the proposed fabrication route provides a large design space in terms of the chemical composition, size, and morphology of the magnetic particles utilized in the formulation. To explore this design space, we study the magnetic properties, mechanical behavior, and deformation of silicone composites prepared with different types of particles (Figure 2).

Commercially available magnetic particles with three different chemical compositions were selected for the experimental analysis (Figure 2a): magnetite ($\text{Fe}_3\text{O}_4$), strontium ferrite ($\text{SrFe}_2\text{O}_3$), and neodymium iron boron (NdFeB). The magnetite particles have either spherical or rod-shaped morphology with an average particle size below 1 μm. Disk-shaped strontium ferrite particles with a unimodal or bimodal size distribution were compared. The unimodal SrFe$_{19}$O$_{20}$ particles display an average size of 1.0 μm, whereas the bimodal powder consists of a mixture of 3.0 and 0.8 μm particles. Finally, NdFeB particles with an average size of 5 μm were taken as an example of a nonoxide powder with strong magnetic susceptibility. When added to the silicone resin, the spherical magnetite, the strontium ferrite, and the neodymium iron boron particles were found to form a homogenous composite after polymerization of the resin mixture (Figure S1, Supporting Information). In the case of rod-shaped magnetite particles, inhomogeneities were observed within the composite likely due to the formation of larger particle networks in this system.

Magnetization curves were used to evaluate the magnetic response of the particle-laden silicones (Figure 2a). To illustrate the simple fabrication process, we measured the magnetic moment under the external magnetic fields up to 0.3 T, which is the maximum value typically used to obtain readily accessible permanent magnets. The composites were compared in terms of their remanent magnetization $\text{M}_r$, the magnetic moment after the removal of the external field. The experimental results show that silicones containing strontium ferrite and rod-shaped magnetite particles reach remanent magnetizations of $\approx$17–18 and 25 emu g$^{-1}$, respectively. This is significantly higher than the magnetization values obtained with the spherical magnetite and the NdFeB particles, which fall below 7 emu g$^{-1}$.

The strong remanent magnetizations observed for the rod-shaped magnetite and the strontium ferrite indicate that the magnetic dipoles of these particles can be more easily oriented upon application of magnetic fields and maintained after the fields are removed. To permanently orient magnetic dipoles in a specific direction, the imposed external field needs to be comparable to or higher than the coercive magnetic field of the material, $\text{M}_c$. This condition is satisfied by the magnetite and strontium ferrite particles, for which we measured the $\text{M}_c$ values of 0.01 and 0.37 T, respectively (Figure S2, Supporting Information). The low remanent magnetization ($\text{M}_r$) observed for the composites with NdFeB is explained by its hard magnetic character. The magnetic dipoles in this material are very stable under an imposed magnetic field. Indeed, the magnetic dipoles in NdFeB would need magnetic fields higher than the measured coercive field of 0.6 T to become fully oriented and reach the large magnetization theoretically expected for this material (Figure S2, Supporting Information). By contrast, the magnetic dipoles of the spherical magnetite particles were found to be easily oriented within the applied magnetic field, but the orientation cannot be sustained when the field is removed. This observation is in good agreement with the magnetic behavior expected for such spherical submicrometer particles.[31] The markedly larger coercivity of the rod-shaped Fe$_3$O$_4$ particles is due to their shape. A magnetic easy axis emerges along the particle’s long axis, because it provides an optimal configuration of the stray field. This so-called magnetic shape anisotropy stabilizes the magnetic dipoles along the long axis when the external field is switched off.[32]

In addition to the magnetic properties, the mechanical behavior of the particle-laden silicones also affects the magnetically driven motion of the soft actuators. To evaluate the combined effect of magnetic and mechanical behavior on the final actuation, we prepared 1 mm thick composite films with 40 wt% of the distinct particle types. The magnetic properties of the composite films were quantified by measuring the magnetic flux density on the surface of the samples in the out-of-plane direction, whereas the in-plane mechanical compliance of the film was directly obtained from tensile tests. Strong, magnetically driven actuation is expected in films combining high compliance and high magnetic flux density.

Our experiments reveal that the silicone films prepared with both types of strontium ferrite particles outperform the other composites by featuring both high compliance and high magnetic flux density (Figure 2b). Surprisingly, the high remanent magnetization of samples with rod-shaped magnetite particles did not translate into a high magnetic flux density in composite films. This unexpected result is probably caused by the alignment of the rods within the plane of the film due to shear forces applied during molding. As the magnetic dipoles are predominantly oriented along the long axis of the particles, their in-plane alignment is expected to lower the magnetic flux density in the out-of-plane direction. The elongated shape of these particles also reduced the mechanical compliance of the film by a factor of 2 in comparison with the spherical magnetite particles. This low compliance might be related to the fact that the rod-shaped particles form a space-filling network at lower volume fractions compared with spheres. To test this hypothesis, samples with rod-shaped magnetite were also prepared in the presence of a dispersant that prevents the formation of a strong particle network. The compliance of the samples containing dispersant was found to be nearly two times higher than that of the spherical particles, supporting the interpretation above. In line with the remanent magnetization data, films prepared with spherical magnetite and NdFeB particles show negligible magnetic flux density.
The response of the composite films to an external magnetic field reflects the measured magnetic and mechanical properties (Figure 2c). To assess the magneto-response of the films, we measured the bending deflection of free-hanging cantilever composite beams under the presence of an external lateral magnetic field of increasing intensity. By combining high compliance and magnetization, films containing strontium ferrite show strong magneto-responsiveness under small fields of a few millitesla. In contrast, the weak magnetizations of composites with spherical magnetite and NdFeB particles lead to minimal deflection for external magnetic fields up to 20 mT. An intermediate response is observed for films prepared with the rod-shaped magnetite particles, which is consistent with their levels of magnetization and compliance.

2.3. Composites with Bimodal Strontium Ferrite Particles

The possibility to magnetize composites with strontium ferrite particles using relatively low magnetic fields ($H = 0.3$ T) makes...
this an attractive powder for the manufacturing of soft actuators using readily accessible materials and tools. Further experiments were, therefore, carried out to explore the properties and performance of silicone films containing up to 70 wt% of the bimodal strontium ferrite particles (Figure 3). Particles with the bimodal particle size distribution were chosen, because they show minimal interference with the curing of the silicone resin.

Different magnetization approaches were compared in terms of the remanent magnetization achieved (Figure 3a,b). To determine the remanent magnetization \( M_r \), the coercive field \( H_c \), and the associated hysteresis behavior of the composites, we subjected the samples to sequential magnetization cycles with increasing magnitude of the maximum applied field. Samples magnetized during curing of the silicone resin were compared with films cast in the absence of a magnetic field. Cyclic magnetization curves were obtained for an applied magnetic field between \(-1.5 \) and \(1.5 \) T. The obtained results confirm that the composites containing bimodal strontium ferrite behave like hard magnets, which are characterized by a strong hysteresis response and a high coercive field of \( \approx 0.37 \) T (Figure 3a).

To evaluate the properties and performance of films prepared using conventional permanent magnets \((H < 0.3 \) T\), we focused the analysis on samples subjected to an external magnetic field between \(-0.3 \) and \(+0.3 \) T (Figure 3b). The application of a magnetic field during curing was found to increase the remanent magnetization by twofold compared with samples that were magnetized only after curing. This indicates that the disc-shaped strontium ferrite particles orient with their easy long axis parallel to the imposed magnetic field while the resin is still not fully cured. Therefore, it is reasonable to interpret the higher magnetization achieved by samples magnetized during curing as a result of the alignment of the particles with their easy axis along the applied field. Magnetization curves measured along different directions of the composite confirm the anisotropic nature of samples magnetized during curing and support the interpretation above (Figure S3, Supporting Information).

While magnetization is important to achieve a strong magneto-response, a functional actuator also needs to maintain its internal magnetization after exposure to the actuating external fields. Our measurements show that the remanent magnetization introduced in the composite films during manufacturing remains unchanged if the external magnetic field is varied between \(-0.2 \) and \(+0.2 \) T (Figure 3c). This demonstrates that the composite films can be repeatedly actuated without magnetization loss if the driving field is maintained within the range of conventional permanent magnets. Deflection experiments reveal that the actuation of our composite films is already possible using external fields that are one order of magnitude lower than this upper demagnetization limit (Figure 2c).

In addition to magnetization during curing, another strategy to enhance the magnetic response of the composite films is to increase the concentration of magnetic particles incorporated into the silicone resin. However, an increase in particle concentration is also accompanied by a reduction in the compliance of the composite, because a stronger percolating network is formed at high particle volume fractions. These antagonistic effects raise the question whether an increase in particle concentration enhances or lowers the magnetic response of the soft composites. To address this question, we measured the compliance, the magnetic flux density, and the deflection of composite films as a function of the weight fraction of bimodal strontium ferrite particles (Figure 3d). An increase in particle concentration was found to monotonically increase the magnetic flux density and decrease the compliance. Importantly, actuation tests show that the deflection of the composite films under the magnetic fields of 10 and 20 mT increases with the concentration of strontium ferrite particles. These experiments indicate that the increase in

---

**Figure 3.** Magnetic, mechanical, and actuation behavior of silicone-based composites prepared with bimodal strontium ferrite particles. a) Cyclic magnetization curve with H-field increasing stepwise from 0.1 to 1.5 T for composites without prior magnetization (dark orange), magnetized after curing (orange), and magnetized during curing (yellow). b) Closer view of the magnetization curves obtained when the magnetic field was varied between \(-0.3 \) and \(+0.3 \) T for three cycles. c) Remanent magnetization \( M_r \) of the composites as a function of the applied magnetic field. The hollow circles indicate the magnetic remanence of the composite after fabrication and before the cyclic measurements. d) Mechanical compliance, remanent magnetic flux density, and magnetic-driven deflection of composites prepared with varying concentrations of strontium ferrite particles.
magnetization resulting from the higher particle content dominates over the counteracting effect of reducing compliance. We observed that the strongest actuation is achieved with composites with 70 wt% of strontium ferrite particles. Concentrations above this upper limit are hard to incorporate into the silicone and lead to pastes with excessively high viscosity that are difficult to mold.

2.4. Actuation Response of Soft Grippers

Silicone formulations prepared with bimodal strontium ferrite particles were used to manufacture magnetically responsive soft grippers that are untethered and can operate remotely (Figure 4). The grippers consist of 2 mm thick planar films with four orthogonal arms in a cross configuration and large-area hand-like patterns at the ends of the arms. Based on the previous deflection experiments, this simple design should allow for significant geometrical changes in the presence of an external magnetic field. To make the grippers responsive, the soft material was magnetized using either one large single permanent magnet beneath the center of the molds or eight small magnets above and below the “hands” (Figure 1b).

The two magnetization approaches lead to the formation of local magnetic domains that vary in orientation and intensity. We used a Hall-probe scanning system to map out the local flux density ($B$), which changes linearly with the magnetization ($M$) of the soft material through a simple known relation ($B = \mu_0 M$ for $H = 0$). Curing the material on top of a single large magnet was found to create magnetic domains aligned along the same direction within the entire gripper, which we refer to as the mono-oriented actuator (Figure 4a). The “hands” of this gripper show a slightly stronger magnetization than the arms because of the inherent concentration of magnetic flux in the edges of the gripper's structure.

Figure 4. Soft robotic gripper with four-arm architecture and tailored magnetic domain patterns. a,d) Color map indicating the local magnetic flux density on the surface of grippers that were magnetized using either a) one single cylindrical magnet or d) multiple pairs of small magnets at the “hands” of the gripper. Based on the orientation of the magnetic domains obtained, the actuators are referred to as a) mono-oriented and d) dual-oriented grippers. b,e) Graphs indicate the variation of the local magnetic flux density along the length of the gripper arm for the two evaluated patterns. c,f) Snapshots of the experimental and simulated c) mono-oriented and f) dual-oriented grippers when subjected to an external magnetic field of 20 mT oriented in different directions. Applying the external field in the same direction as the magnetization of the “hand” leads to opening of the gripper (left). The grippers close their arms when the external field is applied in the opposite direction (right). The large arrows indicate the direction of the applied $H$-field, and the small arrows indicate the remanent magnetization direction of the grippers. The configuration of the grippers in the rest state (under gravity and with no applied field) is shown in the background in faded colors. The contours shown in the simulated objects represent the maximum principal strain in the soft grippers in the open and closed states.
magnetizing magnet. Magnetization using multiple magnets at the “hands” of the gripper results in a very different magnetic domain pattern (Figure 4d). In this case, the magnetic domains in the “hands” and in the “arms” are oriented in opposite directions, leading to what we call a dual-oriented actuator. Such a pattern arises from the fact that the “arms” are exposed to the closing flux lines connecting the opposite poles of the pairs of magnets placed at the “hands.” The magnetic field along these closing lines is oriented in the opposite direction relative to the field generated within each “hand.”

The magnetically controlled actuation of these two types of grippers is demonstrated by fixing the actuators vertically to allow their arms to hang freely under gravity as the initial configuration. When exposed to an external vertical magnetic field, the arms of the grippers undergo an opening or closing motion depending on the direction of the applied field (Figure 4c,f). The motion of the arms is governed by the interplay between the magnetic domain pattern of the gripper, the applied magnetic field, and the gravitational forces. For the mono-oriented gripper, the presence of magnetic domains aligned in the same direction results in a stretching of the arms in both open and closed conformations. In contrast, the arms of the dual-oriented gripper develop a more curved geometry upon actuation due to the opposite reaction of the local magnetic domains in response to the external field.

To qualitatively describe the motion of the two types of grippers, considering both magnetic and gravitational forces, we performed finite-element (FE) simulations using a continuum-level constitutive model recently developed for soft magnetic actuators.\(^\text{19}\) In this model, the stress that is magnetically induced in the soft material is directly proportional to the mechanical compliance, the applied magnetic field, and the local magnetization. Because magnetization \((M)\) is proportional to the magnetic flux density \((B)\), we normalize the data obtained from the Hall-probe scans and used them to estimate the variation in local magnetization along the arms and hands of the grippers (Figure 4a,b,d,e). Such estimation assumes that the local magnetization in the gripper is at maximum where the composite was in close contact with the permanent magnet \((H = 0.3\) T). Under this assumption, the maximum \((M_s)\) value can be taken from the cyclic magnetization data for the composition used to manufacture the actuators \((M_s = 55.5\ \text{emu}\cdot\text{g}^{-1},\) Figure 3c). Using these estimated values and the measured compliance as input in the FE analysis, we find that the simulated conformations of the two grippers in the closed and open states are qualitatively very similar to the experimentally observed geometries. Indeed, the simulations correctly capture the stretching of the arms in the mono-oriented gripper and the more curved geometry of the arms of the dual-oriented actuator. This computational analysis demonstrates that the FE simulations combined with experimentally measured magnetic and mechanical input data can be used to qualitatively predict the motion of our soft magnetic actuators.

2.5. Untethered Functional Actuators

Magneto-responsive soft materials can be used to create untethered fast robots and actuators for pick-and-place, grasping, and moving tasks, as well as 3D objects with remotely controlled morphing capabilities. To illustrate this potential, several demonstrators were built and evaluated in terms of their magnetically driven actuation behavior (Movies S1–S5, Supporting Information).

We start by exploring the morphing capabilities of the magnetic soft grippers with the four-arm design (Figure 5a–c). Grippers with the mono-oriented magnetic pattern are most suitable for pick-and-place tasks, because the out-stretched arm conformation induced by the external field increases the contact area with the object to be picked and leaves the bottom of the gripper open for gentle sliding of the object during placing. This capability was harnessed to pick, transport, and place a raspberry between two distant sites using a standard neodymium magnet permanently installed above the gripper to keep it closed and a manually controlled magnet underneath the platform to induce the opening motion (Figure 5a). The soft conformable nature and the resulting large contact area of the gripper enable controlled manipulation without damaging the delicate raspberry.

Because of its large contact area with the picked object and the stiffening of the stretched arms in the closed state, the mono-oriented soft gripper can also be used to lift objects that are significantly heavier than the actuator itself (Figure 5b). We quantify this lifting capability by measuring the external magnetic field strength required to hold an object with different weights for a given amount of time. The experiments show that the gripper can lift objects that are 60 times heavier than its own weight \((120\) g) for at least 10 s if an external field of \(80\) mT is applied. As expected, the magnetic field required for lifting increases with the weight of the object.

Interestingly, lifting is also possible using lower external fields as long as the application requires holding for a shorter period of time. For example, the field required to lift a \(115\) g object decreases from \(\approx 80\) to \(30\) mT if the hold time is reduced from 10 to 1 s. Such transient lifting capability is possible due to friction at the contact between the gripper’s arms and the surface of the object. Frictional forces delay the gravity-driven sliding of the object through the stretched arms of the gripper. To qualitatively evaluate the role of friction on the lifting performance of the actuator, we applied talc powder as a lubricating agent on the surface of the gripper’s arms. Experiments with talc-coated actuators confirm that lowering friction at the arms significantly reduces the lifting capacity of the gripper and increases the required applied magnetic field (Figure 5b).

Beyond lifting and pick-and-place functionalities, our soft grippers are also unique in terms of actuation speed despite their relatively large size. High actuation speeds can be exploited for catching flying objects, efficient pumping of fluids, or quick morphing of 3D objects. To demonstrate fast catching, we use the dual-oriented four-arm gripper. This design is suitable for catching, because its closed state allows for the full enclosure of the object. An electromagnet is used in this demonstration to enable quick switching of the external magnetic field. Catching experiments were carried out by propelling a blueberry in the direction of a gripper positioned right above the lever catapult (Figure 5c). Snapshots of the catapulting experiment show that it takes less than \(400\) ms for the gripper to close and catch the flying blueberry upon switching on an external magnetic field of \(20\) mT.
The actuation timescale achieved enables the design and fabrication of soft magnetic membranes for contactless actuation at a pulsating frequency in the order of a few Hertz (Figure 5d). This is achieved by applying a cyclic magnetic field with the help of a rotating permanent magnet next to the membrane. Contactless magnetic actuation is not limited to simple membrane geometries but can be extended to also create 3D objects with remotely controlled morphing functionalities. Magneto-responsive 3D objects can be manufactured by casting the particle-laden silicone resin in complex-shaped molds. As an example, we prepared a bunny-shaped bulk object using a 3D printed polymer mold. Exposure of the untethered soft rabbit to an oscillating magnetic field causes the ears of the bunny to quickly move back and forth at a frequency controlled by the rotating magnet used to generate the external field.

3. Conclusion

Magneto-responsive soft actuators that are conformal, fast, and cheap can be fabricated via simple casting of particle-laden silicone resins followed by controlled magnetization using widely available permanent magnets. The level of magnetization achieved after manufacturing is strongly influenced by the chemical composition, size, and morphology of the magnetic particles incorporated into the silicone resin. Because of their intermediate coercive fields compared with magnetite and NdFeB, SrFe12O19 particles require a lower field for magnetization, while still being stable against demagnetization when exposed to the field ranges typically used for actuation. The complex actuation behavior of exemplary four-armed grippers manufactured with these particles depends on the magnetization pattern imposed during fabrication and can be reasonably predicted using an available FE simulation code. This allows for tuning of the magnetization pattern according to the desired functionality. Functional soft actuators manufactured using the proposed route can lift objects 60 times their own weight, pick-and-place delicate items without damage, oscillate with controlled periodicity, and catch delicate flying objects. Because it relies on a simple procedure and on cheap magnets and raw materials, this fabrication route is expected to facilitate the exploitation of magneto-responsive soft actuators by a broader community, including roboticists, architects, designers, engineers, and the general public.

4. Experimental Section

Magnetic Particles: Five types of magnetic particles are investigated: 1) spherical magnetite, Fe3O4 (Bayoxide E 8712, Lanxess); 2) rod-shaped magnetite, Fe3O4 (Bayoxide E 8840, Lanxess); 3) strontium ferrite, SrFe12O19, with bimodal particle size distribution (UF-S2, DOWA Electronics Materials Co. Ltd.); 4) strontium ferrite, SrFe12O19, with...
unimodal particle size distribution (OP-56, DOWA Electronics Materials Co. Ltd.) and 5 wt% neodymium alloy, NdFeB (Magnequench MGFP-B + (D50 = 5 μm)-10215-089 // #F00459). As-received particles were incorporated into the silicone resin mixture to generate the pastes for composite fabrication. For one experimental series, rod-shaped Fe₃O₄ particles were also surface modified by suspending them in an ethanol solution containing 4 wt% poly(acrylic acid) (sodium salt) and ball-milling the resulting suspension (20 wt%) before drying (12 h, 60 °C).

**Formulation and Shaping of Magneto-Responsive Pastes:** The particles and the platinum-catalyzed silicone resin (Ecobond 00-20, Smooth-On-Inc.) were mixed with a spatula until a smooth, viscous paste was formed. The paste was then shaped by casting into 3D printed molds or pressing between two plates separated by spacers to create 1 mm thick films. Due to the high paste viscosity, the inclusion of air bubbles could not be avoided but they did not noticeably affect the final properties of the composites. After shaping, the composite was cured under the magnetizing fields of commercial neodymium magnets at room temperature for at least 4 h.

**Magnetization of Films:** Fabricated films were magnetized by placing cylindrical neodymium magnets (5 mm thick, 60 mm in diameter, N42, Supermagnet) on both sides of the mold to achieve homogeneous, out-of-plane magnetization.

**Magnetization of Four-Arm Grippers:** Two configurations were implemented to magnetize the soft grippers (Figure 1b): 1) eight square magnets (15 × 15 × 8 mm, N42, Supermagnete) attached to the tip of each arm of the gripper and 2) a single cylindrical magnet (5 mm thick, 60 mm in diameter, N42, Supermagnete) placed on one side. All grippers were made from a composite paste containing 60 wt% bimodal strontium ferrite (SrFe₁₂O₁₉) particles.

**Microstructural Characterization:** Electron microscopy images of magnetic particles were taken with an LEO 1530 instrument (Zeiss GmbH, Germany).

**Magnetic Characterization:** Hysteresis loops and the remanent magnetization of magneto-responsive composites were measured in a Physical Property Measurement System (PPMS; Quantum Design) at 300 K with a rate of 2.5 mT s⁻¹. Samples with and without prior magnetization during curing were characterized. Magnetic flux measurements were performed with a three-axis scanning Hall probe (M3D-2A-Port, Senis) positioned 0.5 mm above the sample surface at a pixel resolution of 0.5 mm. The magnetic flux density of the neodymium magnets was measured with a handheld Hall probe (LakeShore 410, Gaussmeter).

**Mechanical Characterization:** The mechanical compliance of the composites was measured in a universal mechanical testing machine (AGS-X, Shimadzu) equipped with a 100 N load cell. Dogbone samples were punched out of 1 mm thick composite films, resulting in a gauge length of 23 mm and a test diameter of 3 mm. Tests were performed by applying a constant displacement rate of 12 mm min⁻¹. Five samples were measured for each composition.

**Magnetor-Response of Composite Films:** The deflection of films was measured using a microprocessor-controlled electromagnet (MFG-100, Magnebotix) that allowed for precise control over the applied field. Samples were prepared by cutting 1 mm thick composite films containing 30–70 wt% particles into rectangular beams (4 × 20 mm). The sample was suspended vertically and subjected to a magnetic field along the long axis of the film, perpendicular to the magnetization direction. The field was applied in increments from 1 to 20 mT. The motion of the films was recorded using a digital camera, and the displacement was extracted using a MATLAB image processing toolbox.

**Functional Soft Actuators:** In most experiments, the four-arm soft grippers were actuated manually using neodymium magnets. The lifting measurements were performed with grippers suspended below a permanent magnet (25.4 mm thick, 63.5 mm outer diameter, 9.53 mm inner diameter, N50, Supermagnetman), which was shielded by four 1 mm thick steel plates (EN-AW 1.0330, Debrunner Acifer). The applied magnetic field was controlled by adjusting the distance between the gripper and the magnet. The opening and closing of the gripper were achieved by overlapping the magnetic fields with a secondary magnet controlled by hand. The load cell lifted by the gripper consisted of a plastic container filled with various weights attached to a 1 cm diameter wooden ball, which worked as a handle (Figure 5b). Once the actuator was gripping the handle, the substrate was quickly removed. Two scenarios were considered: 1) momentary lifting strength for 1 s, after which the load cell slipped out of the gripper’s grasp and 2) persistent lifting strength for at least 10 s, after which the experiment was terminated. To test the effect of friction, experiments were also performed with grippers that were covered in talc and then cleaned with water and ethanol before testing.

To catch a flying object, the gripper was controlled with an electromagnet setup (MFG-100, Magnebotix) which applied 20 mT to close the gripper. The object, a blueberry, was catapulted into the gripper’s direction, and the magnetic field turned on simultaneously. The whole procedure was monitored with a high-speed camera at 960 frames per second (Sony RX100 VI).

The vibrating membrane (2 mm thick) and the molded bunny were actuated with a permanent magnet rotating at 700 rpm. The magnet generated a field of 30 mT at the chosen distance from the sample.

**FE Simulations:** The simulations were carried out using the commercial FE package ABAQUS 2017. All calculations were performed using an Abaqus/Standard solver. A python script was developed for pre- and post-processing of the models, and a user-material subroutine (UMAT) was used to assign a constitutive law to the structures assuming an ideal hard magnetic soft material. The constitutive law was originally proposed and implemented in UMAT by Zhao and co-workers[19]. This law assumes that the material displays a residual magnetic flux density, and that there is a linear relationship between the induced magnetic flux density and the applied magnetic field over the range of field strengths used for actuation. To account for the spatial distribution of the remanent magnetization within the samples, we normalized the remanent magnetization with locally scanned magnetic flux measurements (Figure 4b,e).

**Supporting Information**

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

**Acknowledgements**

The authors thank the financial support from the Swiss National Science Foundation (SNSF) through the SNSF consolidator (Project number BSCG10_157696) and mobility grant to A.R. (Project number P3P3P2_174326). They are grateful to Ruike Zhao and Xuanhe Zhao (Soft Active Materials Laboratory, MIT) for kindly sharing the user-material subroutine used for the finite-element simulations.

**Conflict of Interest**

The authors declare no conflict of interest.

**Data Availability Statement**

Research data are not shared.

**Keywords**

actuators, magnetic particles, silicone, soft robotics

Received: December 29, 2020
Revised: February 17, 2021
Published online:
[1] N. El-Atab, R. B. Mishra, F. Al-Modaf, L. Johari, A. A. Alsharif, H. Alamoudi, M. Diaz, N. Qaiser, M. M. Hussain, Adv. Intell. Syst. 2020, 2, 2000128.
[2] M. Zarek, M. Layani, I. Cooperstein, E. Sachyani, D. Cohn, S. Magdassi, Adv. Mater. 2016, 28, 4449.
[3] M. D. Bartlett, E. J. Markvicka, C. Majidi, Adv. Funct Mater 2016, 26, 8496.
[4] E. J. I. Barron, R. S. Peterson, N. Lazarus, M. D. Bartlett, ACS Appl. Mater. Interfaces 2020, 12, 50909.
[5] H. Souri, H. Banerjee, A. Jusu, N. Radacsi, A. A. Stokes, I. Park, M. Sitti, M. Amjadi, Adv. Intell. Syst 2020, 2.
[6] G. Chowdhary, M. Gazzola, G. Krishnan, C. Soman, S. Lovell, Sustain. Basel 2019, 11, 6751.
[7] J. J. Huaroto, E. Suarez, H. I. Krebs, P. D. Marasco, E. A. Vela, IEEE Robot. Autom. Lett. 2019, 4, 17.
[8] J.-H. Hsiao, J.-Y. Chang, C.-M. Cheng, Adv. Robot. 2019, 33, 1099.
[9] X. Yang, W. Shang, H. Lu, Y. Liu, Y. Yang, R. Tan, X. Wu, Y. Shen, Sci. Robot. 2020, 5, eabc8191.
[10] L. S. Novelino, Q. J. Ze, S. Wu, G. H. Paulino, R. K. Zhao, P Nat. Acad. Sci. USA 2020, 117, 24096.
[11] M. Schaffner, J. A. Faber, L. Pianegonda, P. A. Ruhs, F. Coulter, A. R. Studart, Nat. Commun. 2018, 9, 878.
[12] S. W. Kwok, S. A. Morin, B. Mosadegh, J. H. So, R. F. Shepherd, R. V. Martinez, B. Smith, F. C. Simeone, A. A. Stokes, G. M. Whitesides, Adv. Funct. Mater. 2014, 24, 2180.
[13] C. Pacchierotti, F. Ongaro, F. van den Brink, C. Yoon, D. Prattichizzo, D. H. Gracias, S. Misra, IEEE T Autom. Sci. Eng. 2018, 15, 290.
[14] J. C. Breger, C. Yoon, R. Xiao, H. R. Kwag, M. O. Wang, J. P. Fisher, T. D. Nguyen, D. H. Gracias, ACS Appl. Mater. Inter. 2015, 7, 3398.
[15] K. Kobayashi, C. Yoon, S. H. Oh, J. V. Pagaduan, D. H. Gracias, ACS Appl. Mater. Inter. 2019, 11, 151.
[16] J. L. Guo, C. Q. Xiang, P. Zanini, J. Rossiter, IEEE Robot. Autom. Lett. 2019, 4, 2364.
[17] D. Martella, S. Nocentini, C. Parmeggiani, D. S. Wiersma, Adv. Mater. Technol. US 2019, 4, 1800571.
[18] M. P. da Cunha, Y. Foelen, R. J. H. van Raak, J. N. Murphy, T. A. P. Engels, M. G. Debije, A. P. H. J. Schenning, Adv. Opt. Mater. 2019, 7, 1801643.
[19] Y. Kim, H. Yuk, R. K. Zhao, S. A. Chester, X. H. Zhao, Nature 2018, 558, 274.
[20] W. Gao, L. L. Wang, X. Z. Wang, H. Z. Liu, ACS Appl. Mater. Inter. 2016, 8, 14182.
[21] B. J. Nelson, I. K. Kaliakatsos, J. J. Abbott, Annu. Rev. Biomed. Eng. 2010, 12, 55.
[22] Y. Kim, G. A. Parada, S. D. Liu, X. H. Zhao, Sci. Robot. 2019, 4, eaax7329.
[23] D. Rus, M. T. Tolley, Nature 2015, 521, 467.
[24] T. Xu, J. Zhang, M. Salehizadeh, O. Oanaizah, E. Diller, Sci. Robot. 2019, 4, eaav4494.
[25] W. Q. Hu, G. Z. Lum, M. Mastrangeli, M. Sitti, Nature 2018, 554, 81.
[26] C. Peters, O. Ergeneman, P. D. W. Garcia, M. Muller, S. Pane, B. J. Nelson, C. Hierold, Adv. Funct. Mater. 2014, 24, 5269.
[27] H.-W. Huang, M. S. Sakar, A. J. Petruska, S. Pané, B. J. Nelson, Nat. Commun. 2016, 7, 12263.
[28] J. Kim, S. E. Chung, S.-E. Choi, H. Lee, J. Kim, S. Kwon, Nat. Mater. 2011, 10, 747.
[29] M. Stingaciù, A. Z. Eikeland, F. H. Gjorup, S. Deledda, M. Christensen, Rsc Adv. 2019, 9, 12968.
[30] A. K. Singh, O. N. Srivastava, K. Singh, Nanoscale Res. Lett. 2017, 12, 298.
[31] Q. Li, C. W. Kartikowati, S. Horie, T. Ogi, T. Iwaki, K. Okuyama, Sci. Rep. UK 2017, 7, 9894.
[32] H. P. Johnson, W. Lowrie, D. V. Kent, Geophys. J. Roy. Astr. S 1975, 41, 1.