

Magnetically Controlled Soft Robotics Utilizing Elastomers and Gels in Actuation: A Review

Hyun-Joong Chung,* Andrew M. Parsons, and Lelin Zheng

A magnetic field has unique advantages in controlling soft robotics inside of an enclosed space, such as surgical catheters or untethered drug-delivering robots operating in the human body. Soft actuators, made of elastomers and gels functionalized with magnetically active materials, are natural choices to drive magnetically controlled motions of soft robots. Recent innovations in soft material technologies, including 3D printing, origami/kirigami, tough hydrogels, mechanical metamaterials, and liquid metal-injected elastomers, offer technological foundations to develop soft actuators and robots with significantly enhanced performance. Herein, an overview of magnetic soft actuators and robots from a materials engineer's perspective is provided. First, the historical background and recent trends of magnetic soft actuators are discussed. Second, the motions of tethered or untethered magnetic soft robotics are classified into aquatic swimmers, terrestrial locomotors, and grippers. Herein, preprogrammed motion under patterned magnetic stimuli is achieved by controlled magnetization of elastomeric materials containing hard magnetic particles. Finally, the applications of magnetically controlled soft robotics in surgical and therapeutic medical devices are discussed.

project an object (chameleons' tongue shooting toward a prey) by replacing the rigid structures with stretchable yet mechanically tough elastomers and gels that can change their shape and dimensions.^[3] As such, unprecedented unique applications, including humanoid corobots for elderly care, wearable joint supports and soft exoskeletons for human motion assistance, surgical devices such as endoscopes and catheters, prosthetic body parts, and artificial organs, have been suggested.^[5,6] The capabilities of soft robotics expand with enhanced "smart functionalities" of elastomers and gels. Smart functionalities include, but are not limited to, controllable modulations of dimension, shape, and stiffness that enable actuation of soft robotics with respect to nonpneumatic or nonhydraulic external stimuli, which may be of electric, magnetic, chemical, thermal, or photonic origin.^[5–8]

The motions of soft robotics are driven by soft actuators. Traditional actuators, that

most of the conventional robots employ, are gearmotor driven with precision linkages regulated by proportional-integral-derivative (PID) controllers. Soft actuators, on the other hand, drive the motion of robotics by regulating the dimension and the stiffness of a soft material, which is exactly how an animal's muscles function. Despite their inherent incapability of precise and linear control of motions, it is well received that soft actuators have advantages in being compact, light weight, highly energy efficient, and mechanically compliant.^[9–14] Considering the proposed applications of soft robotics in confined and uncontrolled environments, soft actuators seem to be well suited to drive soft robotics. Soft actuators are typically classified by the materials that dictate their motion. For example, gels or elastomers without intrinsic functionality (i.e., responsiveness to non-mechanical stimuli) may contain compressed air or fluids to allow pneumatic or hydraulic actuation, respectively. Phase transformation of the polymeric matrix or metallic constituent has been exploited in shape-memory polymer (SMP) or shape-memory alloy (SMA) actuators, respectively. Electronic and ionic conduction, sometimes with associated chemical reactions, have been the principles of actuation for dielectric elastomer actuators (DEAs) and ionic electroactive polymers (IEAPs). The former utilizes the dielectric property and the deformability of elastomers and gels, whereas the latter also involves ionic conductivity within soft materials. Magnetic properties of the compositing materials have allowed remotely controlled actuation of

1. Introduction

Soft robotics, originally developed as a biomimetic design concept to enable adaptive interactions with uncontrolled environments, is now considered to be the core technology to blur the boundary between humans, robots, and the distribution of physical labor.^[1–3] Currently, conventional robots made of skeletal structures with actuators at the joints struggle to perform simple tasks such as grabbing fruits or eggs because they require complicated feedback algorithms to manipulate the rigid structures.^[4] On the other hand, soft robotics are capable of easier mechanisms to grip an object (octopi's tentacles or starfishes' wrapping arms), walk through a tortuous narrow path (caterpillars' locomotion), and

Prof. H.-J. Chung, A. M. Parsons, L. Zheng
 Department of Chemical and Materials Engineering
 University of Alberta
 Edmonton, Alberta T6G 1H9, Canada
 E-mail: chung3@ualberta.ca

 The ORCID identification number(s) for the author(s) of this article can be found under <https://doi.org/10.1002/aisy.202000186>.

© 2020 The Authors. Published by Wiley-VCH GmbH. This is an open access article under the terms of the Creative Commons Attribution License, which permits use, distribution and reproduction in any medium, provided the original work is properly cited.

DOI: [10.1002/aisy.202000186](https://doi.org/10.1002/aisy.202000186)

magnetorheological (MR) fluids and elastomers. Such actuations can be activated by stimuli from various origins, including an electric field, magnetic field, pressure differential, heat, light, and chemical reactions. A review by Sitti and coworkers summarized a succinct table of soft materials and their demonstrated actuation stimuli.^[12] Each material has merits and demerits. The merits and demerits must be quantitatively compared in terms of technological needs contingent with the specific application. A great example to showcase the development of mid-sized (from millimeter to a meter) soft robotics is soft gripper technology. A review by Shea and coworkers summarized the performances of grippers based on each material in terms of the mass and size of the gripper and object, response time, and power consumption.^[15]

Utilizing magnetic force has some unique merits over other stimuli mechanisms. As Sitti and coworkers pointed out in their earlier reviews,^[12,16] magnetic fields can penetrate through a wide range of materials, their spatial gradients can be freely generated over the space, and decoupling from other stimuli is straightforward. In addition, a material's response to the change of magnetic field is relatively fast compared with other modes of actuation. As a result, magnetic actuation has successfully demonstrated swimmers, crawling devices, and micropumps working in enclosed spaces, which are especially suited to enable target drug delivery, microsurgery, microfluidics, and assembly within the body.^[12]

Magnetically responsive soft materials, which regulate their dimensions and/or stiffness with respect to the magnetic field, are the gist of magnetically driven soft actuators. These materials must be mechanically robust with substantial toughness and tear resistance, whereas having a low elastic modulus is essential. Elastomers are polymeric materials with soft, deformable, and resilient mechanical properties. Gels are swollen polymers that can incorporate solutions with various additives.^[8,17] To enable magnetism to steer the translational and rotational motion of the soft actuator, the most popular approach is to blend micro- or nanosized magnetic materials into the elastomers and gels.^[18] The control of motion can become more deterministic by distributing local magnetic susceptibility within the elastomeric composites.^[19] A few novel processing methods, such as continuously distributed magnetization^[20] and programmed magnetic axis alignment,^[21] have been developed to create sections with arbitrary magnetic axis to facilitate the control of motion. Recently emerging fabrication strategies of 3D printing, origami, and kirigami are instrumental in applying these concepts in developing new materials for soft actuators.^[8,22]

In this Review, we intend to summarize the vibrant field of magnetically responsive soft actuators and magnetically controlled soft robotics. In Section 2, a broad overview on the historical background and the recent trends of magnetic soft actuators is provided from materials researchers' perspective. Especially, emerging technologies of 3D printing and origami/kirigami are reviewed and their impact on the recent studies of magnetic soft actuators and robotics is discussed. In Section 3, soft robotics whose tethered or untethered motions are driven by magnetic fields are classified into aquatic swimmers, terrestrial locomotors, and grippers. In the section, we emphasize on the importance of achieving complex programmed motions by the controlled magnetization of elastomeric materials containing

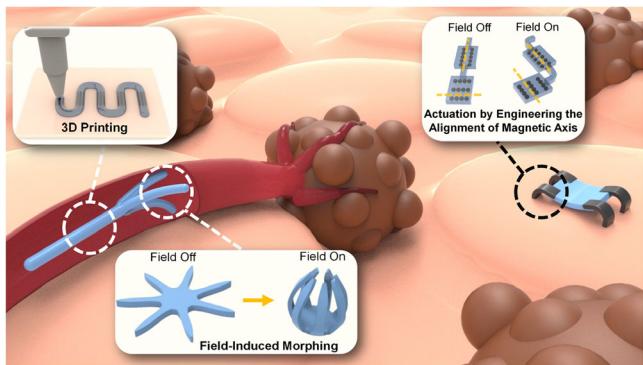


Figure 1. Schematic illustration of magnetically steered swimming and terrestrial robots approaching tumors. Inset at the top left shows a schematic of 3D printing, which is one of the most significant innovations in material fabrication. Inset at the top right shows that the patterned leg motion of a terrestrial crawler can be directed by aligning the magnetic axis within the actuating leg. Inset at the bottom center shows an illustration of a gripper controlled by field-induced morphing, which is a representative example of practical applications of magnetically controlled soft actuators. These three featured innovations, along with the depicted biomedical application, represent the key messages of the Review Article.

hard magnet particles. In Section 4, the current and potential applications of the magnetically controlled soft robotics in surgical and therapeutic medical devices are discussed. Finally, Figure 1 shows a visual summarization of the key features that are covered in this Review Article.

2. Materials: Classification and Fabrication

Magnetically responsive soft actuators require their constituting materials to have high magnetic susceptibility whereas they regulate their dimensions and/or stiffness with respect to the magnetic field. There have been studies to incorporate intrinsic magnetic susceptibility within the polymeric macromolecules, but their synthesis is complicated whereas their magnetic properties are not sufficient for actuation.^[23] Instead, most studies disperse magnetic particles, which are typically inorganic and rigid, into soft and deformable matrix materials.^[24,25] In this approach, controlling the dispersion and distribution of the magnetic particles is critical. While a uniform dispersion of the particles, i.e., preventing random agglomeration, ensures maximal utilization of the magnetic field, controlling the distribution of the particles creates a discrete magnetization profile so that only specific parts of the actuators respond to an applied field to enable motions in a regulated manner.

In this section, constituting materials in the magnetically responsive soft actuators, namely elastomer and gel matrices and magnetic particles, are briefly reviewed and then strategies to ensure the dispersion of the particles are discussed. Then, recently emerging material fabrication strategies to control the distribution of the particles including 3D printing, origami, and kirigami are reviewed. A long-studied class of smart materials that stiffen under an applied magnetic field, MR elastomers and gels, is then reviewed. Also, when an inductive coil is made of flexible conducting materials such as liquid metal, magnetic

actuators can be made without magnetic materials, which is showcased in the following section. Finally, magnetic soft actuators that harness heat-induced material deformation of SMPs or gels are reviewed.

2.1. Matrix Materials: Elastomers and Gels

Elastomers, a portmanteau of elastic polymer, are lightly cross-linked networks of polymer chains above their glass transition temperature so that they deform under external stress and then return to their original shape when the external stress disappears. The term is often used interchangeably with rubbers, whereas the latter generally refers specifically to vulcanized organic polymer networks, such as cis-1,4-polyisoprene (i.e., natural rubber) or polybutadiene (synthetic rubber commonly used as car tire material).^[26] The International Union of Pure and Applied Chemistry (IUPAC) definition of gels is “nonfluid colloidal network or polymer network that is expanded throughout its whole volume by a fluid.”^[27] In other words, gels are crosslinked polymers swollen by solvents and/or low-molecular components, and the degree of swelling can be dependent on various environmental factors that affect osmotic pressure, such as temperature and humidity, as well as the content of ionic and nonionic solutes in a solution-immersed environment. Therefore, “wet” gels have advantages over “dry” elastomers to enable multifunctional stimuli-responsiveness. For soft actuator applications, the gels must possess considerable toughness, tear resistance, and notch insensitivity.^[28] The gels may also exhibit excellent adhesive properties.^[29] There has been extensive research on developing tough (hydro)gels at the interface of polymer chemistry and fracture mechanics.^[30–32] Various matrix materials, such as commercial silicone elastomers (Sylgard®, Ecoflex®, Shin-Etsu®, and Dragon Skin®), natural rubbers, thermoplastic polyurethanes (TPUs), and numerous types of hydrogels and organogels, have been suggested. It is notable that most of commercial silicone elastomer products are indeed composite materials of siloxane-based polymers and silica or clay nanoparticles. Our recent Review provides summaries on the macromolecular origin of the toughness of elastomer and gels, as well as on the key material properties that are important in soft robotics applications.^[8]

Elastomers and gels can become magnetically responsive by attaching/incorporating permanent magnets^[33] or magnetic coils^[34] or by dispersing magnetic particles and/or ferrofluid in the material.^[35] The former method is simpler and resulting magnetism is more straightforward to analyze, but both permanent magnets and magnetic coils are considerably stiffer and more brittle in nature compared with elastomers and gels. Therefore, the latter is more common. As a result, the problem of making a good magnetic elastomer breaks down as follows: a judicious selection of a combination of materials from intrinsic properties, compatibilization techniques between dissimilar materials, and processing methodologies to distribute the magnetic particles and produce a variety of structures and porosities.

2.2. Magnetic (Nano)Particles

The magnetic property comes from the orbital magnetic moment and spin magnetic moment of electrons. Some transition metals,

such as iron (Fe), cobalt (Co), and nickel (Ni), and some lanthanides, such as neodymium (Nd), have a significant magnetic moment from incomplete *d* or *f* orbital states, which render all the atomic magnetic dipole moments to align parallel to each other within a region called a magnetic domain. These metals and their oxides show ferromagnetic or ferrimagnetic behaviors, wherein magnetic ordering can cause macroscopic magnetization that increases the magnetic susceptibility of the material dramatically.^[36]

When the size of a magnetic particle is reduced below a critical size (typically some tens of nanometers), the particle can accommodate only one domain so that it always has a net magnetic moment that exerts a magnetic field to external space. Here, the threshold size is an intrinsic material property related with the size of the Bloch wall, which is the distance between two adjacent magnetic domains to accommodate a 180° contrast in magnetic dipole moments by gradual rotation of magnetic moments of atoms in the wall.^[36] If the particle size becomes too small, however, thermal energy is sufficient to randomize the magnetic dipole in a very short period of time, which is called superparamagnetism.^[37] For magnetite (Fe_3O_4) particles, which is one of the most commonly used ferrimagnetic material, single-domain behavior occurs when the radius of the particle is between 25 and 43 nm.^[37]

A few aspects of material properties are to be considered when choosing magnetic particles. Soft magnetic particles, such as pure iron, carbonyl iron, and Permalloy®($\text{Ni}_{80}\text{Fe}_{20}$), require less energy to align atomic dipoles to magnetize or demagnetize and thus have less residual magnetization in the absence of an external field, which is a critical requirement for MR elastomers.^[38] However, mixing hard magnet particles, such as alnico, cobalt steels, and neodymium alloys, makes the composite behave like permanent magnets. Upon magnetization, magnetic elastomers containing hard magnet particles behave as permanent magnets with a defined magnetic axis. As a result, the motion of such composites can be directed by attractive or repulsive forces against the applied magnetic field.^[39] Zhao and coworkers compiled a comprehensive and simple theoretical foundation to account for magnetically driven dimensional changes and motions of elastomers and gels that have the properties of hard magnets.^[40,41] Quickly after, studies followed to further the application of magneto-mechanical models to implement in finite element frameworks.^[42] Currently, soft robotics that contain magnetic actuators use hard magnet particles more frequently than soft ones. Second, the surface of the magnetic particle has to be engineered to prevent agglomeration. The high surface energy of metallic or ceramic particles causes agglomeration problems in solution, suspension, gel, or ambient environments. Surface treatment with silane coupling agents showed some improvement on the magnetization efficiency of magnetic elastomers.^[43] In magnetic hydrogels, a core-shell approach, specifically covering Fe_3O_4 cores with SiO_2 shells by Stöber's method,^[44,45] was shown to be effective in achieving nearly individual particle-level dispersion when coupled with the negative charging of the particle surface in acidic environments.^[46] Finally, the core-shell structure of neodymium iron boron (NdFeB) covered by silver has merit in obtaining both strong ferromagnetic properties and high electrical conductivity.^[47]

2.3. Strategies to Disperse Magnetic Particles in Elastomers and Gels

Magnetic elastomers and gels are composite materials whose properties are usually maximized when the magnetic particles are uniformly dispersed without agglomeration. In Section 2.2, the core–shell structure was mentioned to be effective in achieving such dispersion. In addition, three fabrication strategies are commonly used: blending, *in situ* precipitation, and grafting onto methods.^[48]

The blending method, also known as physical doping, is the process of mixing magnetic particles, uncrosslinked polymer chains, and crosslinking agents in a single pot often filled with a solvent or a nonpolymerizing silicone oil. During the crosslinking step, the magnetic particles, which do not form chemical bonds with polymeric constituents, are physically trapped in the cross-linked networks of gels or elastomers. The blending method is popular because of its simplicity, but challenges remain in controlling the dispersion and preventing the agglomeration of particles. In swollen gels, bleaching of magnetic particles is also a challenge because of the lack of chemical bonding between the particles and the polymeric network.

The *in situ* precipitation method is applicable to gels but not to elastomers. The method infuses the precursor solution of magnetic metals in a polymeric gel network, followed by triggering a chemical reaction to precipitate magnetic particles in the gel. Compared with the blending method, the magnetic particles prepared by the *in situ* precipitation method show better uniformity in dispersion, and incorporating a large amount particles is more feasible. On the other hand, this method is only suitable for specific hydrogels that can endure alkaline conditions. The necessity of an alkaline solution precludes the method's application in certain biomedical applications such as cell encapsulation. Albertsson and coworkers mixed a hemicellulose oligomer (O-acetyl-galactoglucomannan, degree of polymerization ≈ 40) and a crosslinker (epichlorohydrin) in an aqueous solution of iron chloride salt to make Fe_3O_4 nanoparticles whereas the hydrogel is synthesized from the precursor solution.^[49] The resulting magnetic hydrogel had good magnetic sensitivity with a uniform distribution of nanoparticles in the cellular structure of hydrogel networks although some degree of nanoparticle agglomeration was observed.

Grafting-onto method, in which the magnetic particle serves as a crosslinker by having chemically functional groups on the surface, is also a commonly used method for preparing magnetic elastomers and gels. The covalent bonding between the particles and the polymer chains renders an enhanced structural and chemical stability to the magnetic composite material. Jiang and Liu functionalized magnetic fly ash (the magnetic portion of fly ash was prescreened and collected) and attapulgite (a common form of magnesium aluminium phyllosilicate) with acrylic surface functional groups. Consequently, the authors utilized the nanoparticles as crosslinkers and strengthening agents to synthesize a polyacrylic acid-based hydrogel composite material.^[50] Schmidt and coworkers modified the surface of CoFe_2O_4 magnetic nanoparticles with silicone-based unsaturated methacrylic acid and used it as a crosslinking agent to covalently bond with polyacrylamide to prepare a magnetic hydrogel.^[51] The resulting

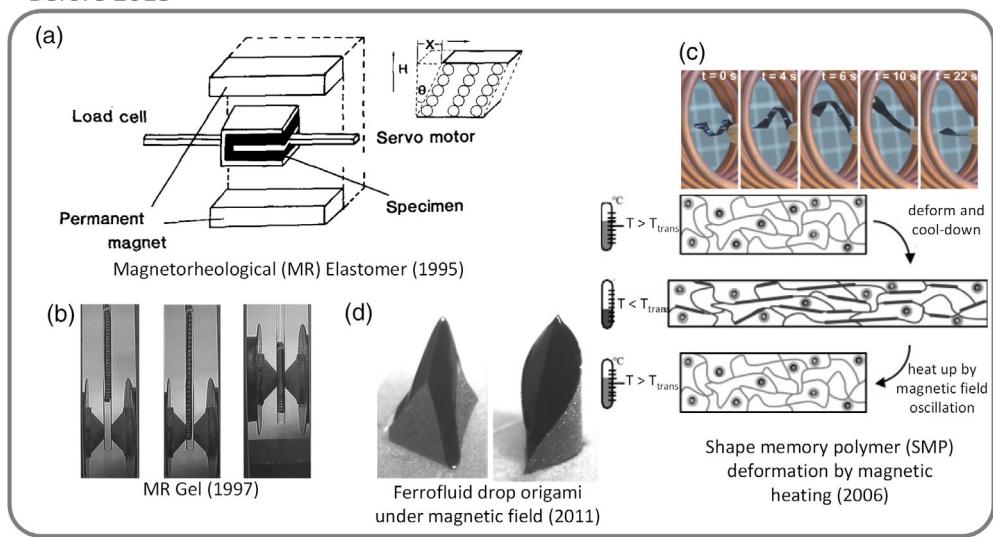
gel was magnetically susceptible. The structural roles of nanoparticles as crosslinking points were convincingly displayed by transmission electron microscopy (TEM) imaging. Evans et al. adsorbed a silicone polymer, poly(dimethylsiloxane-co-aminopropylmethysiloxane), onto the surface of Fe_2O_3 nanoparticles to prevent agglomeration.^[52] Stark and coworkers functionalized the surface of cobalt nanoparticles' 4-vinylbiphenyl group so that magnetic nanoparticles play a role as a crosslinker between 2-hydroxy-ethyl-methacrylate (HEMA) polymer chains during the batch copolymerization process.^[53]

The distribution of magnetic particles can be either isotropic or anisotropic. While isotropic materials are easier to fabricate, the particles can be linearly ordered by applying a magnetic field while curing the elastomer matrix. Such ordering of magnetic particles can introduce directionality in the actuation behavior.^[54] Confining the magnetic particles within structured microsized domains enables both directionality and efficiency in magnetic actuation.^[55] Discretizing magnetic susceptibility by distributing magnetic particles within structured domains allows only specific parts of the soft actuator to respond to an applied field. The emergence of novel fabrication technologies, including 3D printing, origami, and kirigami, facilitated the discretizing strategy; this topic is reviewed in Section 2.5.

2.4. MR Elastomers and Gels

MR elastomers are solid-phase counterparts to MR fluids. MR fluid, a suspension of magnetic particles in viscous liquids, is an established technology since the 1940s and now is ubiquitous in everyday gadgets such as shock absorbers, brakes, clutches, seismic vibration dampers, control valves, and artificial joints.^[56] The physics of MR fluid operation principles has been thoroughly studied.^[57] In short, magnetostatic particle–particle interaction (ordering force) dominates over the thermal motion of particles (randomizing force), resulting in chain-like particle aggregates that align toward the direction of the magnetic field. The formation of chain-like aggregates causes stiffening of the MR fluid with modulus contrast of well over three orders of magnitude with respect to the strength of the applied magnetic field. However, MR fluids have intrinsic limits in practical applications, such as the deposition of iron particles, sealing problems, and environmental contamination.^[58] In the mid-1990s, researchers in Lord Corporation dispersed carbonyl iron particles in a silicone matrix to modulate the viscoelastic properties of elastomer composites by applying an external magnetic field and named the new class of material as MR elastomers.^[55,59] In the same era, researchers in Toyota mixed iron particles with silicone gels to obtain similar results (Figure 2a).^[60] Ford Motors researched carbonyl iron particles mixed in natural rubber as a magnetostrictive material.^[61] Zrinyi et al. swelled crosslinked poly(vinyl alcohol) chains by an aqueous solution containing magnetite particles (Figure 2b).^[62–65] The hydrogel composites were shown to modulate their shape and rigidity with respect to the applied magnetic field and were named ferrogels by the inventors. When the matrix polymer is swollen by large amounts of solvent or low-molecular-weight components, the resulting composite material is classified as MR gels.^[66] However, MR elastomers often contain low-molecular-weight silicone oils to

Before 2015



Breakthroughs in related technologies: 3D printing, origami/kirigami, Tough hydrogels, mechanical metamaterials, and liquid metal injected elastomers

2016 ~ Present

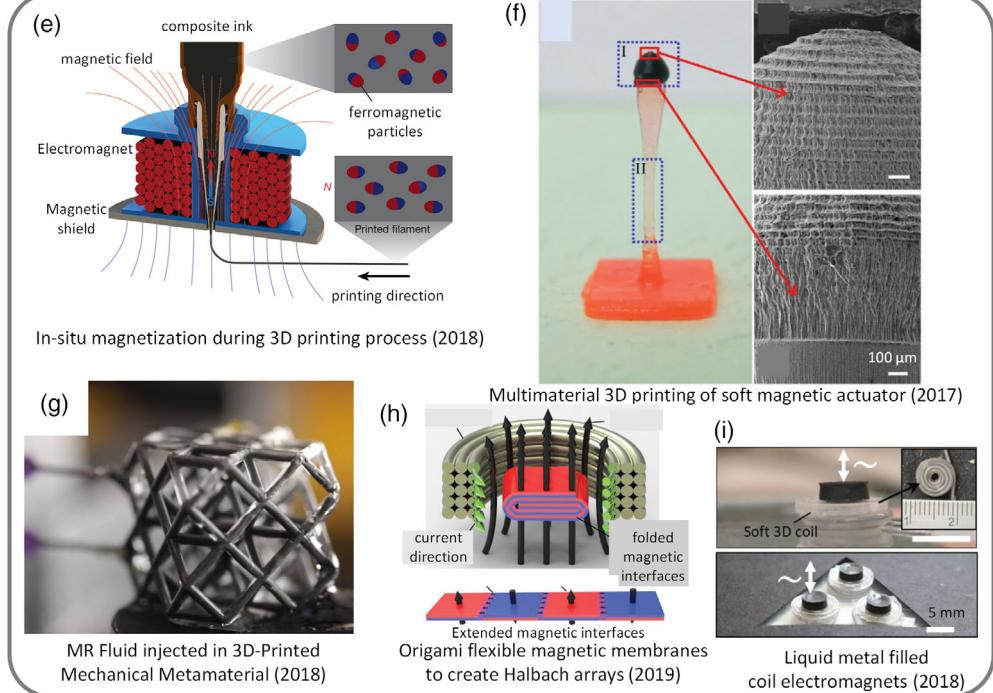


Figure 2. Examples of soft magnetic actuator material development in early (before 2015) and recent years (between 2016 and now). The first examples of magnetically active soft materials are MR. a) elastomer (Reproduced with permission.^[60] Copyright 1995, Wiley) and b) gel (Reproduced with permission.^[62] Copyright 1997, Elsevier), both of which contain micro- or nanosized magnetic particles in a soft material matrix. Magnetically induced heating by a high-frequency oscillating magnetic field has been used to trigger volume-changing phase transformations in c) SMPs (Bottom part: Reproduced with permission.^[105] Copyright 2006, Wiley. Top part: Reproduced with permission.^[106] Copyright 2006 The Author(s). Published by PNAS.) and hydrogels (not shown). d) Early ideas of origami by the aid of ferrofluid drop under a variable-time magnetic field (Reproduced with permission.^[96] Copyright 2011, American Physical Society). Breakthroughs in related technologies in early 2010s motivated various innovative ideas in the field of soft magnetic materials. Most notably, specialized 3D printing techniques, such as e) in situ magnetization (Reproduced with permission.^[98] Copyright 2018, Springer Nature) and f) multimaterial printing (Reproduced with permission.^[88] Copyright 2017, Wiley). g) 3D-printed hollow structures have been filled with MR fluid to enable magnetically induced morphing (Original image provided by the courtesy from the authors of ref. [87]. Copyright 2018, Mancini, Loh, and Spadaccini.) h) The idea of origami has been applied to create membranes with arbitrary Halbach arrays in a simple manner (Reproduced with permission.^[39] Copyright 2019, Wiley). i) Filling liquid metal in an elastomeric matrix has enabled fully flexible coils for electromagnets, and this technology has been utilized to fabricate soft (electro)magnetic actuators without the presence of so-called magnetic materials (Reproduced with permission.^[34] Copyright 2018, Wiley).

increase the plasticity and fluidity of the matrix, as well as dissipate the internal stress concentration.^[58] Considering the definition of elastomers and gels that we reviewed in Section 2.1, these types of elastomers may be classified as gels; the boundary between MR elastomers and MR gels is rather contextual. A Review Article by Ahamed and coworkers provides a self-explanatory table that compares the property differences between different MR materials, including MR fluids, MR elastomers, and MR gels.^[66]

Compared with MR liquids, MR elastomers have a large off-state viscosity (or modulus), which makes the material suitable as a damping isolator for high-end vehicle seat vibration suspension or adaptive base isolation.^[67] The most important feature of MR elastomers and MR gels that is distinctive from MR liquids is, however, its ability to change shapes drastically in addition to viscosity modulation.^[68,69] Thus, MR elastomer research is naturally connected to the development of magnetically controlled soft actuators for soft robotics. As of 2020, MR elastomers and gels are actively researched to enable unprecedented material properties such as lower off-state viscosity,^[70,71] 3D printability,^[72] off-axis anisotropicity for directional actuation,^[73] surface-modified magnetic particles for better dispersion,^[74] novel polymer matrix for lower cytotoxicity,^[75] and fiber reinforcement of MR elastomers to strengthen the matrix and guide the field-induced alignment of magnetic particles.^[76] Recently demonstrated applications that harness the dimensional modulation of MR gels and elastomers include precisely controllable peristaltic pumps,^[77] enhanced contrast for tactile displays,^[78] and grippers for transporting objects.^[76] All these applications are within the category of magnetic soft actuators being used for soft robotics operation.

2.5. 3D Printing, Origami, and Kirigami

3D printing is an additive manufacturing technique that enables the fabrication of magnetic soft actuators with arbitrarily controlled local structure and composition. Due to its viscoelastic nature during and after the printing process, 3D printing of gels and elastomers has unique technological features that are different from the fused deposition modeling (FDM) printing of polymer melts.^[79] In fact, 3D printing technology for gels and elastomers has benefited from 3D bioprinting for cell-laden scaffolds, which can be classified as a subset of direct ink writing (DIW) technology.^[80–82] In addition, conventional 3D printing technologies such as FDM, stereolithography (SLA), and selective laser sintering (SLA) are also used to 3D print gels and elastomers, but material selection is rather restricted.^[83]

3D printing has a few unique advantages in manufacturing soft actuators. Most prominently, controlling the shape and architecture of stimuli-susceptible parts within the actuator allows the elongation, contraction, or twisting deformation of the actuator to be precisely programmed with respect to the applied stimuli; this, indeed, resembles how an animal's muscles deform in a deterministic manner.^[84] Li and coworkers printed 3D patterns of MR fluid encapsulated in a nonmagnetic silicone matrix and studied the effect of the pattern's shape on the efficiency of vibration damping with respect to the magnitude and orientation of the applied magnetic field.^[85,86] Loh, Spadaccini, and coworkers injected MR fluid into the network of 3D-printed

hollow struts (Figure 2g).^[87] The 3D-printed architecture, termed "field-responsive mechanical metamaterial" by the authors, exhibited dynamically controlled movement within a second of response time with respect to the applied magnetic field. Yu, Wang, and coworkers harnessed multmaterial printability of advanced 3D printing technology to produce arbitrarily shaped magnetic actuators with multiple domains that contain different Fe₃O₄ nanoparticle loadings to enable programmed motions.^[88] Pané and coworkers printed and bonded domains filled with soft and hard magnetic particles to find an optimal balance between the reversibility of soft magnetic domains and the rapid actuation ability of hard magnetic domains.^[89] Nuzzo and coworkers harnessed multmaterial printability to its full potential to fabricate magnetically steered jellyfish- and starfish-like aquatic soft robots made of ionic hydrogels.^[90] The structures were printed in planar shapes, and subsequent hydration with preprogrammed amount of salt content resulted in fully 3D structures with locally varying volumes in a deterministic manner; this spatiotemporally controlled fabrication method is an epitome of 4D printing.

Folding and bistability are the two important concepts to enable fast-transforming actuation that is currently used in mimicking the flapping motion of insect wings^[91,92] or snap-buckling-induced strong grabbing of Venus flytraps^[93] and in enabling rapidly deployable structures such as vascular stents, solar panels for spacecraft, or military tents.^[94,95] In a nutshell, bistability is to have two different local equilibrium morphological configurations, whereas the two configurations are switchable with respect to external stimuli. The morphological configurations are often defined by careful cutting of relief patterns (kirigami) and programmed folding of functional patterns (origami), typically from an originally 2D object. Often used in conjunction with 3D printing technologies, origami and kirigami are used to design mechanical metamaterials, which exhibit complex mechanical behaviors including folding, instability, and bistability by the interplay of periodically arranged networks of structured constituent materials; our Review Article covers this topic in depth.^[8] Magnetic soft actuators are adapting the concept of folding, instability, and bistability to enable rapid shape transformation with high output power (Figure 2c).^[33,96,97] Zhao and coworkers fabricated various magnetically deployable 3D structures by controlling the local orientation of ferromagnetic domains by a novel 3D printing method.^[98] In this work, the authors defined the direction of magnetic domains by aligning the polarity of the ferromagnetic NdFeB particles in a strong magnetic field at the moment of 3D printing, immediately followed by curing the silicone matrix to fix the magnetic domains (Figure 2e). Zhao and coworkers introduced a combination of mechanisms, connected by symmetric and asymmetric joints, to enable directed motion when their metamaterial structure undergoes a folding–bending actuation cycle driven by periodically switching the magnetic field from downward to upward.^[99] Here, the word "mechanism" refers to the collections of rigid elements connected with flexible hinges, and an "asymmetric joint" means a joint which folds easily to one side while not folding to the other side, just like our own elbows or knees. Huang and coworkers utilized the origami technique to make various magnetic domain patterns on thin (130–500 μm) polydimethylsiloxane (PDMS) membranes containing Nd₂Fe₁₄B microparticles.^[39] The patterns, called Halbach

arrays, result in enhanced magnetism to be exerted toward the desired orientation at a controllable strength, and thus crawling, flapping, and other complex motions were realized (Figure 2h).

2.6. Magnetic Actuation without Magnetic Materials

Electricity and magnetism are the two resulting phenomena of electromagnetism. Colloquially speaking, the term “magnetic material” refers to matter that is prone to polarize strongly in the presence of external magnetic fields (ferromagnetic or ferrimagnetic materials); often, the scope of the word narrows down to materials that exert a magnetic field toward outer space even in the absence of an external magnetic field (hard magnetic material). However, flowing electric current itself also results in exerting a magnetic field externally, which is how electromagnets work. Therefore, magnetic soft actuators can actually be made without magnetic materials by incorporating induction coils.

A technological challenge, however, is that induction coils require electrical resistivity to be extremely low whereas severe deformation is accompanied with actuation. Liquid metal injected in an elastomer matrix provides a smart solution.^[100] Visell and coworkers made a long and hollow filament of Ecoflex 0030 silicone filled with a eutectic alloy of gallium and indium (EGaIn) and then rolled the electrically conductive wire into a coil form to fabricate a soft gripper (Figure 2i).^[34] Ha and coworkers, Liu and coworkers, and Kaltenbrunner and coworkers independently fabricated a planar film of encapsulated liquid metal coils to demonstrate a loudspeaker,^[101] gripper,^[102] and flipper,^[103] respectively.

2.7. Magnetically Induced Nonmagnetic Actuation

SMPs are a class of programmable shape-morphing materials that can be deformed arbitrarily and then recover their original shape upon external stimuli. The driving force for shape recovery is typically an entropic spring, the tendency of polymers to retrieve random chain conformation to maximize entropy.^[104] As mentioned in Section 1, many soft actuators operate by the morphing of SMPs. Although SMPs have a rather modest energy density for morphing and a slow morphing rate (from a few seconds to minutes), SMPs have strong merits of processability and morphological diversity. When elastomers or gels have SMP properties, vibration damping and large strain durability also become additional merits.^[13]

The shape recovery behavior can be triggered either by the removal of temporary bonding that holds a nonequilibrium chain conformation of polymers (click chemistry) or by enabling the mobility of polymer chains to retrieve their maximum entropy conformation (reverse glass transition). While thermal stimuli are used most often to trigger both mechanisms, inductive heating of embedded magnetic particles under an alternating magnetic field has been used as a stimulus to trigger shape recovery of nonelastomeric SMPs (Figure 2d).^[105–107] In addition, Leidlein and coworkers found that the temperature and magnetic field required for the shape recovery of magnetic particles containing SMPs are dependent on the strength of the applied magnetic field during the event of shape deformation and temporary shape

fixing.^[108,109] Tracy and coworkers studied the interplay between photothermal heating and magnetic deformation.^[110] Zhao and coworkers mixed both soft (Fe_3O_4) and hard (NdFeB) magnetic particles in an SMP matrix, where the former is responsible for rapid heating under an alternating magnetic field and the latter for programmed directional motion of the soft actuator.^[111] While all aforementioned SMPs are not elastomers or gels, Ghosh and Cai mixed Fe_3O_4 nanoparticles in a thermally morphing hydrogel of poly(*N*-isopropylacrylamide) (PNIPAM) and triggered the composite's shape morphing by applying an alternating magnetic field.^[112] A recent work by Wang and coworkers applied origami and kirigami concepts to the same material system to enable sophisticated shape morphing.^[113]

3. Directing Movements by Applying Magnetic Fields

Intuitively speaking, it is obvious that a remotely applied magnetic field can direct the location and movement of a soft robot containing magnetically susceptible materials. One may imagine that pulling the center of mass of the robot by finely controlled magnetic field gradients may be sufficient for navigating untethered small-sized robots submerged in a fluid. Although the direct field gradient method is still used in some applications, it has been theoretically and experimentally shown that magnetically driven helical propulsion or twisting/flapping motions are far more efficient.^[114] In addition, terrestrial and gripping motions require moving mechanisms that are certainly more complicated than simply being pulled by magnetic forces. Therefore, directing movements of soft robotics by applying magnetic fields requires strategically arrayed soft actuators to choreograph coordinated movement. **Table 1** summarizes various designs for steering motions of the robots.

The first requirement for such coordinated movement is the system that generates environmental magnetic fields exerted on soft robotics. The most frequently used setup is the triaxis-nested Helmholtz coil system.^[115] The procedure that involves an electromagnet system to guide medical devices is called “magnetic stereotaxis.”^[116] In clinical setups, magnetic resonance imaging (MRI) systems can be used to manipulate soft robotics.^[117]

The second component for coordinated movement is actual soft robots and distributed soft actuators therein. In this section, we group the magnetically controlled motion of soft robotics into three subsection categories: swimming locomotion, terrestrial locomotion, and gripping motion. Swimming locomotion intends to propel the soft robot by promoting the flow of the surrounding fluid by choreographed movements of tails, flappers, or fins. Thus, controlling the fluid mechanics of the surrounding media is the purpose of the movements of the soft actuators. Terrestrial locomotion, on the other hand, intends to leverage the friction force between the robot and the supporting surface to cause directed movement. Most of the studies use a biomimetic approach to mimic moving legs or slithering bodies utilizing soft actuators. Finally, gripping is the most fundamental motion to manipulate an object; thus, it deserves a dedicated review.

Table 1. A summary of reported studies on magnetically steered untethered swimmers and terrestrial robots.

Class	Subclass	Matrix/magnetic material	Magnetization method	Fabrication method	Magnetic controller	Magnetic stimuli pattern	Ref.
Swimmer motions							
Helical propulsion	Nonmagnetic helix (Rigid)		Magnetic head ^{a)} /Rigid tail ^{a)}	n/a	n/a	Not specified ^{a)}	[122] (2019)
	Nonmagnetic helix (Soft)		Head: NdFeB Tail: Nonmagnetic PDMS ^{a)} or fabric ^{a)} or PET	n/a	n/a	Three-axis Helmholtz coils	[124] (2019)
Magnetic helix (Rigid)			PEGDA-PTA copolymer/Fe ₃ O ₄ (nanoparticle ^{a)})	Uniform magnetization	Two-photon polymerization	Commercial eight-coil controller	[125] (2019)
Elastic tail propulsion	Nonmagnetic tail		Head: Neodymium magnet ^{a)} Tail: PET (100 µm thick)	n/a	n/a	Single-axis Helmholtz coil	[126] (2006)
			Head: NdFeB Tail: Polyimide film (125 µm thick)	n/a	n/a	Single coil	[127] (2008)
Magnetic tail			PDMS ^{a)} /Fe ₂ O ₃ (nanoparticles ^{a)}	Uniform magnetization	Mold casting	Three electromagnetic coils	[128] (2009)
			Ecoflex 00-50/NdFeB (10–50 µm)	Continuously distributed magnetization	Mold casting	Three-axis Helmholtz coils	[20] (2014)
			Polypyrrole (ppy)/nickel (1.75 µm-long rod)	n/a	Linking between ppy and nickel with an elastic hinge	Two electromagnetic coil pairs	[130] (2015)
			Ecoflex 00-50/NdPFeB (nanoparticle ^{a)})	Continuously distributed magnetization	Mold casting	Single-axis Helmholtz coils	[129] (2018)
Frog or jellyfish			Sylgard 184/NdFeB(nanoparticle ^{a)})	Programmed magnetization axis alignment	Mold casting	Single-axis Helmholtz coils	[99] (2019)
			Aginate hydrogel/Fe ₃ O ₄ (nanoparticle ^{a)})	Soft magnetic particle in the composite with a local gradient in particle content	3D printing (gel extrusion)	3 Tesla MRI instrument	[90] (2019)
			Ecoflex 00-10/NdFeB (\approx 5 µm)	Continuously distributed magnetization	Mold casting	Single-axis Helmholtz coils	[131] (2019)
Other types	Direct magnetic steering (no locomotion)		PEGDA/Fe ₃ O ₄ (< 100 nm)	n/a	Photo polymerization	Three-axis Helmholtz coils + two-axis Maxwell coils	[135] (2016)
			PDMS ^{a)} /NdFeB (\approx 5 µm)	Uniform magnetization	Mold casting	Three N38 permanent magnets	[134] (2020)
	Janus microdimer		Silica/Ni coating on one side (100 nm thick)	n/a	Vacuum deposition of nickel on a hemisphere of silica beads	Three-axis Helmholtz coils	[136] (2019)
	Self-assembling microcubes		SU-8/Co coating on one side (100 nm thick)	n/a	Vacuum deposition of cobalt on one side of SU-8 cube	Single-axis Helmholtz coils	[137] (2020)

Table 1. Continued.

Class	Subclass	Matrix/magnetic material	Magnetization method	Fabrication method	Magnetic controller	Magnetic stimuli pattern	Ref.
Terrestrial motions							
No legs	Earthworm	PNIPAm/Fe ₃ O ₄ (15–30 nm)	n/a	Thermal polymerization	High-frequency generator with a water cooling system	High-frequency oscillating magnetic field (heating)	[141] (2017)
Inchworm		PEGDA/Fe ₃ O ₄ @SiO ₂ (~120 nm)	Programmed magnetization axis alignment	Photo polymerization	Not specified ^{a)}	Oscillating magnetic field	[21] (2011)
Rolling and jumping	Ecoflex 00-10/NdFeB (~5 μm)	Continuously distributed magnetization	Mold casting	Three-axis Hemholtz coils	Three-axis Hemholtz coils	Rotating and oscillating magnetic field	[144] (2018)
Two-legged crawler	PNIPAm/NdFeB (microparticle ^{a)})	Uniaxial magnetization	Mold casting	Three-axis Hemholtz coils	Three-axis Hemholtz coils	Rotating and oscillating magnetic field	[145] (2020)
Four-legged crawler	Ecoflex 00-20/Fe3O4 (~5 μm, ≈50 nm, ≈25 nm)	Uniaxial magnetization	Spin coating (~150 μm-thick film)	5-DOF control; one Maxwell pair + one Hemholtz pair + four rectangular coils	5-DOF control; one Maxwell pair + one Hemholtz pair + four rectangular coils	Alternating magnetic field	[147] (2020)
Turtles	Ecoflex 00-10/FePrB (~5 μm)	Continuously distributed magnetization	Mold casting	An array of six electromagnetic coils	An array of six electromagnetic coils	Rotating magnetic field	[146] (2019)
Quadrupeds							
Many-legged crawler	Centipedes	UV curable elastomer ^{a)} /NdFeB (~5 μm)	Programmed magnetization axis alignment	3D Printing (DLP based)	Three-axis Hemholtz coils	Rotating and oscillating magnetic field	[143] (2019)
Multilegged		Sylgard 184/Iron (6–10 μm)	Uniaxial magnetization	Magnetic field- and particle-assisted assembly	Permanent magnet controlled by 3-DOF manipulator	Patterned motion of the permanent magnet controller	[148] (2018)

^{a)}Exact details unknown—followed the nomenclature of the original article.

3.1. Magnetic Swimmers

A magnetic field can be controlled within the volume limited by the size of the nestled Helmholtz coils, and the field penetrates deeply into most media, including optically nontransparent materials. Sitti and coworkers pointed out that these features render the magnetic field the most suitable method to direct movements of small-sized (less than a few centimeters) robots working in enclosed spaces.^[16,118] The human body falls well into this category, wherein many of the accessible cavities and bodily vessels are filled with fluid. Therefore, magnetic swimmers are indeed the key components to enable targeted drug delivery and minimally invasive surgery (MIS), to name a few, whereas they are compatible with many other medical imaging systems such as ultrasound, MRI, and X-ray.^[16]

In their seminal Review Article, Nelson and coworkers categorized the locomotion of magnetic swimmers into two categories: helical propulsion and elastic tail propulsion.^[114] Helical propulsion is powered by a rotary actuator that has screw-shaped tail(s) typically made of rigid materials with either hard or soft magnetic properties^[114] (**Figure 3a**). It should be noted that most helical swimmers are microrobots made of rigid materials and are not classified as soft robots; nevertheless, the technology and its application are closely relevant to soft robotics and thus are included in the current section. This mode of locomotion is inspired by bacterial flagella. To produce propelling torque, a rotating magnetic field must be applied. In one example, $\approx 20\text{ }\mu\text{m}$ -long helical swimmers were fabricated out of a photore sist by the direct laser writing method based on the two-photon polymerization method, followed by nickel coating (25 nm) for magnetic susceptibility;^[119] the small rigid swimmer was then installed onto the tail of an immotile sperm to assist fertilization by enabling magnetically driven motion^[120] (**Figure 3a**). A Review Article by Pané and coworkers and studies therein provide an exhaustive review of magnetically propelled helical swimmer works.^[121] More recent works on helical swimmers include a computational fluid dynamics (CFD) model to predict the actual motion of rigid swimmers in cylindrical channels,^[122] the use of an oscillating magnetic field instead of a rotating one to enable self-propulsion,^[123] a combination of theoretical and experimental studies to account for the dynamic nature of morphology and swimming properties of soft helical tails,^[124] and a degradable microrobot for targeted drug release and hyperthermia therapy.^[125]

Elastic tail propulsion describes the directional motion of soft robotic swimmers whose elastic tail undergoes a traveling-wave motion driven by applying an oscillating or rotating magnetic field. Initially, a swimmer was designed to have a magnetic head whose oscillation drives a passive traveling-wave motion to an elastic tail made of a thin nonmagnetic polymer sheet.^[126,127] Garstecki et al. demonstrated that a tail made of magnetite particles-filled PDMS can drive elastic tail propulsion under a rotating magnetic field.^[128] The concept of distributing magnetic particles in an elastomeric matrix evolved when Sitti and coworkers introduced a continuously distributed magnetization profile in an elastic tail^[20] (**Figure 4a**). In the study, NdFeB nanoparticles were uniformly distributed in a silicone matrix but magnetization was conducted when the elastic tail material was bent

into a circular shape. The idea of continuously distributed magnetic profiles was recently extended by Zhang and Diller, who demonstrated a magnetic sheet swimming on the surface of water.^[129] Pané, Nelson, and coworkers came up with a different approach to distribute the magnetization profile by adding elastic and nonmagnetic hinges between nickel microrods.^[130] The authors found that this increased freedom of the tail was possible by discretizing magnetically susceptible parts in the elastic tail instead of making a continuum. The widespread availability of multimaterial 3D printing and origami/kirigami technologies at the time, indeed, boosted creativity to realize more complex structures with discretized magnetic parts, such as frog-like swimmers by Zhao and coworkers,^[99] starfish-like simmers by Nuzzo and coworkers,^[90] and jellyfish-like swimmers by Sitti and coworkers (**Figure 3c**)^[131] and Huang and coworkers.^[132]

There are a few other recent magnetic swimmer studies that deserve special mention. For example, Yang and coworkers utilized DNA-functionalized magnetic nanoparticles to drive navigational locomotion of a DNA hydrogel-based soft robot.^[133] Feng and coworkers designed a millimeter-scale swimmer in a manta-ray shape to demonstrate a steering precision of $10\text{ }\mu\text{m}$ and a maximum actuation speed of 200 mm s^{-1} .^[134] Park and coworkers used the direct field gradient method to locate the position of a drug-carrying microrobot to a desired location, whereas they utilized environmental pH change as a stimulus to release the drug.^[135] Li and coworkers fabricated $\approx 10\text{ }\mu\text{m}$ -sized Janus microdimer swimmers by coating nickel as a hemispherical dome on a spherical silica particle.^[136] The microdimer swimmers have efficient propulsion behavior under a rotating magnetic field. Finally, Abbott, Velev, and coworkers controlled the self-assembly and folding dynamics of magnetic microbots utilizing the interplay between microcubes and anisotropic media.^[137] In the study, each microcube had a magnetic coating only on one face, whereas the anisotropic media was liquid crystalline fluid whose ordering was influenced by the self-assembly of microcubes and vice versa (i.e., self-assembly is also influenced by the ordering of liquid crystals).

Although it is not untethered, nor truly swimming, it is notable that tethered catheters can “swim” inside a human body because there are many tubular structures such as the blood vessels, gastrointestinal tract, lymphatic system, and urinary tract. Zhao and coworkers developed a catheter-like soft continuum robot whose navigation is precisely steered by magnetic fields, and thus the study deserves a special mention.^[138] In the study, the soft robot has a magnetic core wire made of PDMS or TPU containing nonuniformly dispersed NdFeB nanoparticles, whereas its shell layer is an acrylic-based hydrogel for lubrication against the inner walls of bodily tubes. The soft robot navigated through a tortuous cerebrovascular phantom including multiple aneurysms with remarkable precision. Benchtop demonstrations that show clinical relevance of this study are shown in Section 4.2.

3.2. Magnetic Terrestrial Locomotions

Magnetic terrestrial locomotion refers to the magnetically steered translational movement of soft robots by leveraging the friction force against the surface. Terrestrial and marine creatures

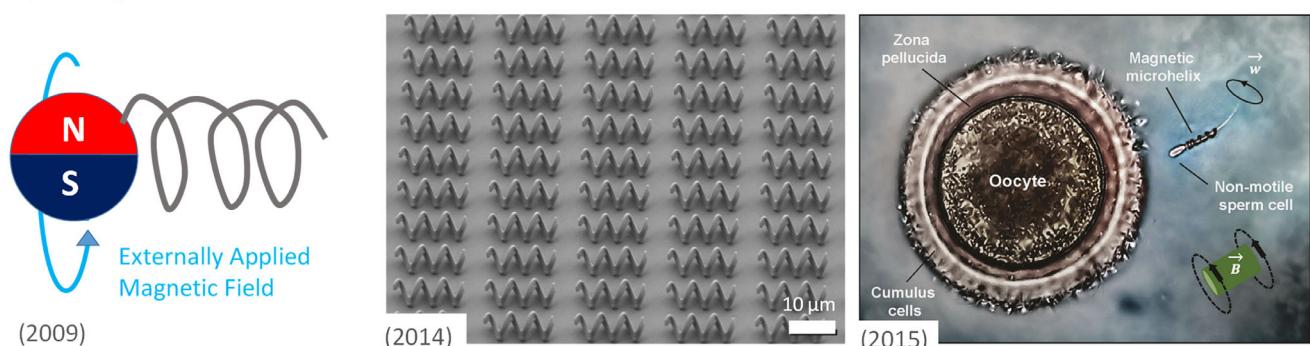
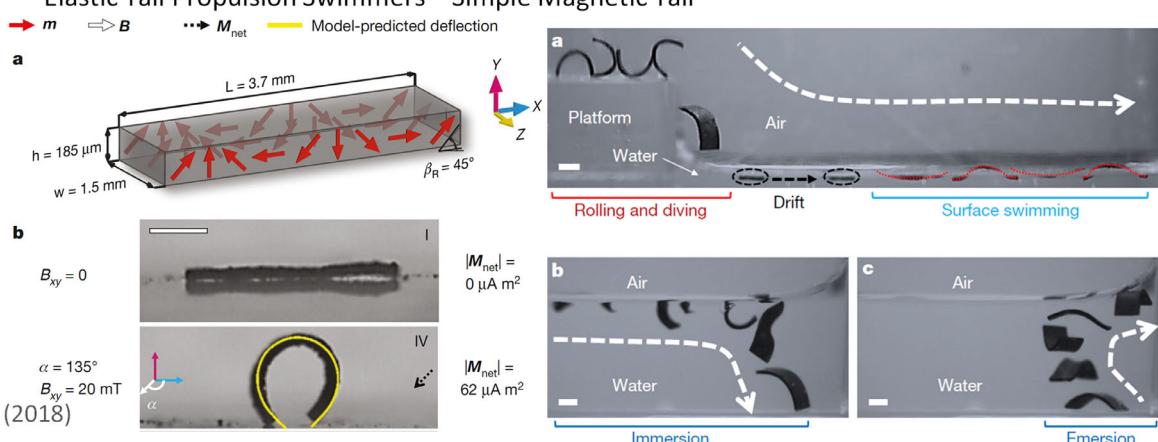
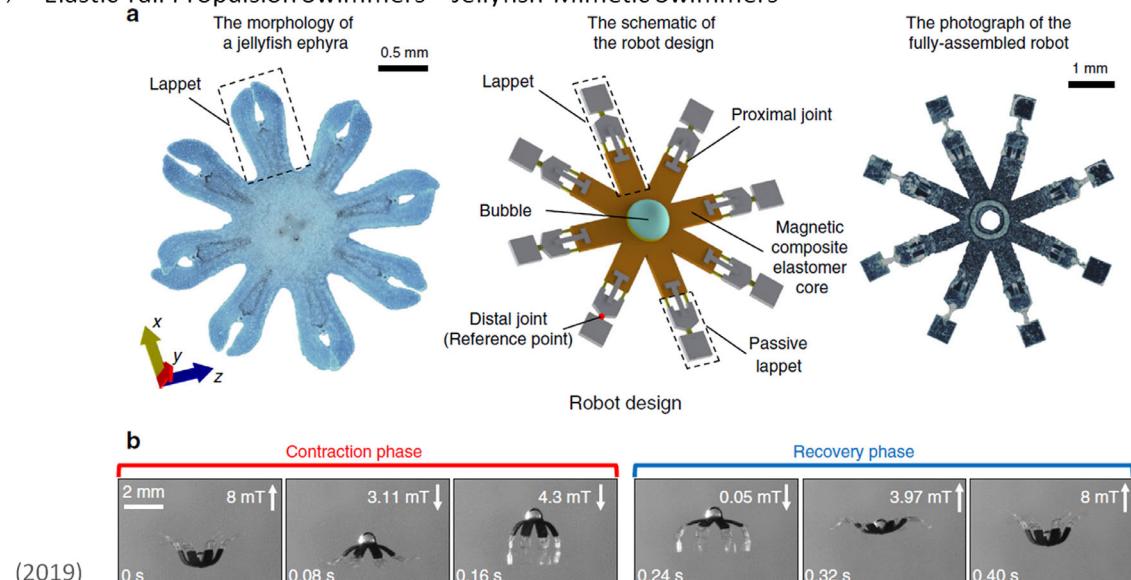
(a) Rigid Tail Helical Swimmers

(b) Elastic Tail Propulsion Swimmers – Simple Magnetic Tail

(c) Elastic Tail Propulsion Swimmers – Jellyfish-Mimetic Swimmers


Figure 3. Three examples of untethered magnetic swimmers: a) Rigid tail helical swimmers. (Left) The basic mechanism of their motion under a rotating magnetic field; an original drawing to explain schematics. Note that the rigid tail helical swimmers are not classified as an example of soft robots. (Middle) Microfabricated helical swimmers (Reproduced with permission.^[119] Copyright 2014, Elsevier). (Right) Application in assisted fertilization (Reproduced with permission.^[120] Copyright 2016, American Chemical Society). b) An example of simple elastic tail swimmers fabricated with a continuously distributed magnetization technique. (Reproduced with permission.^[144] Copyright 2018, Springer Nature). c) An example of a complex elastic tail propulsion swimmer that mimics the swimming of jellyfish (Reproduced with permission.^[131] Reproduced under the terms of the CC-BY 4.0 License. Copyright 2019 The Author(s). Published by Springer Nature.).

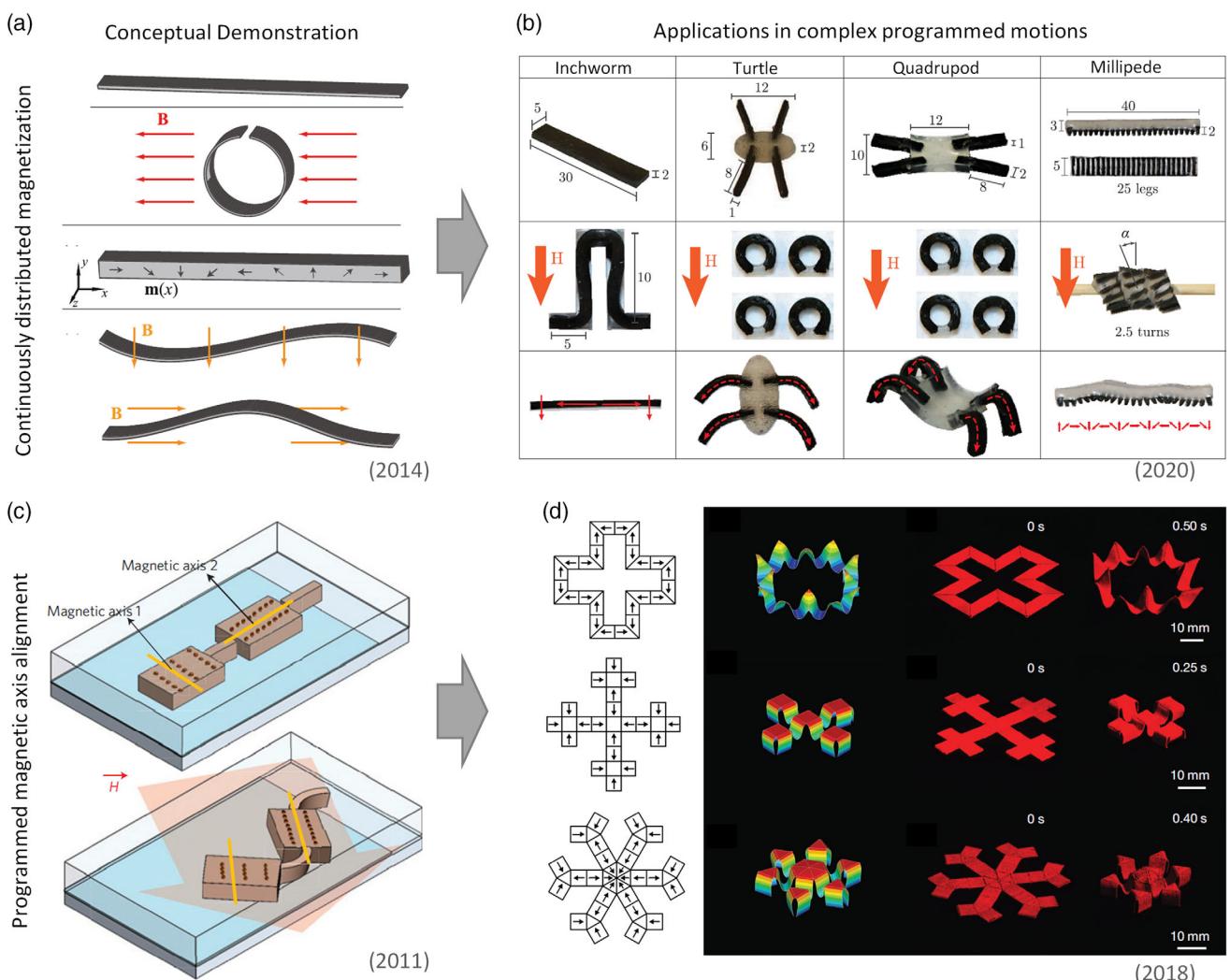


Figure 4. Two key ideas to achieve complex programmed motions by the controlled magnetization of elastomeric materials containing hard magnet particles. Top left and right columns show the first work that demonstrated the concept of a) continuously distributed magnetization (Reproduced with permission.^[20] Copyright 2014, Institute of Physics) and b) its applications on various biomimetic terrestrial locomotions (Reproduced with permission.^[146] Copyright 2019, IEEE; the unit of the scales are millimeter), respectively. Bottom left and right columns show the first work that demonstrated the concept of c) programmed magnetic axis alignment (Reproduced with permission.^[21] Copyright 2011, Springer Nature) and d) its application in programmed folding (i.e., origami) of complex structures (Reproduced with permission.^[98] Copyright 2018, Springer Nature), respectively.

demonstrate a plethora of well-optimized locomotions. These motions are often driven by the movement of protrusive objects such as legs, tentacles, cilia, or bristles, and each animal has its own optimized moving mechanism to adapt to its habitat. Even without involving direct actuation of such protrusions, earthworms vermiculate by peristaltic contractions, snakes slither by undulating their bodies, and inchworms gait by their signature looping motions. In short, terrestrial locomotions are highly diverse in Animalia. Consequently, there are very diverse approaches for soft robots in realizing terrestrial locomotions, and most approaches are based on biomimicry. Indeed, diversity provides a variety of selections to a robot engineer to choose an adequate solution that is specific to the operating environment.

The movement of annelids (including earthworms) involves a peristaltic contraction that regulates local dimensions of their

soft body. In other words, annelids can move forward because they generate a continual bulging wave from head to tail. Mimicking the movement involves a coordinated deformation of elastomeric soft actuators. In fact, such localized and fine-controlled contractions may be straightforward to realize using DEAs^[139] or hydraulic actuators.^[140] Although it is not directly magnetic actuation, Vernerey and coworkers demonstrated an annelid motion by magnetically induced heating of nanoparticles to trigger swelling and deswelling of the hydrogel matrix, whereas the directionality of the soft robot's movement is determined by the array of scale-like features.^[141]

The gait of inchworms has been the prime target of biomimicry in the field of soft robotics.^[142] Kwon and coworkers made a seminal contribution to the field of magnetic soft robots by introducing the idea of programmed magnetic axis alignment.^[21] In

this study, the authors fabricated building blocks of the magnetic hydrogel composite of $\text{Fe}_3\text{O}_4@\text{SiO}_2$ (core@shell) particle-filled poly (ethylene glycol) diacrylate (PEGDA), whereas magnetization during the curing process caused an anisotropic magnetization. Then, the magnetic axes of hydrogel blocks are arranged to have a zig-zag alignment, whereas the connections between the blocks are nonmagnetic and mechanically flexible (Figure 4c). By applying an oscillatory magnetic field to the perpendicular direction against the surface, the soft robot demonstrated inchworm gait. For example, Diller and coworkers conducted a comprehensive work that demonstrates a centipede walker, elastic tail propulsion swimmer, and various types of grippers by advancing the concept of programmed magnetic axis alignment.^[143] In this study, the authors conducted magnetization during the solidification process of digital laser processing (DLP), a type of 3D printing, to create arbitrary alignment of magnetic axes. In fact, the concept of the patterned alignment of the anisotropic blocks is the most frequently used in soft grippers, which is reviewed in Section 3.3.

Sitti and coworkers' concept of a continuously distributed magnetization profile, which was introduced to fabricate an elastic tail swimmer,^[20] was further advanced by the same group to realize millimeter-scale soft robots, demonstrating rolling, walking, crawling, and jumping on a dry surface, as well as swimming in water and on its surface (Figure 3b).^[144] In this study, all these movements were precisely controlled simply using the direction and the strength of the magnetic field. Later, Du et al. added an optical camouflage functionality to a similar magnetic microrobot.^[145]

Crawling requires the periodical motions of legs. Misra and coworkers hypothesized that limb-driven motions are advantageous in traversing uneven terrain and carrying payload without restricting motion and demonstrated mimicry of the biological motion profiles of four different animals: inchworms, turtles, quadrupeds, and millipedes (Figure 4b).^[146] Here, both turtles and quadrupeds have four limbs for movement, but the leg movement of the turtle is entirely in plane with the surface whereas that of the quadruped involves a lifting motion off the surface. The actuating body (inchworm) and limbs (turtle, quadrupeds, and millipedes) were made of silicone rubber containing praseodymium–iron–boron (PrFeB) particles as hard magnets. The locomotion of each soft robot was possible due to the continuously distributed magnetization profile, defined by magnetizing the actuating body and limbs while being arranged to have a specific shape.^[146] Although this study does not provide a direct proof for hypothesis, the study provides variable-time functions to control the magnetic field to drive motions for each soft robot. Each robot's performance parameters such as the displacement rate and the deviation in the lateral direction (y -error) were also defined in two ways: displacement per second and body lengths per actuation cycle. While most studies on crawling involves legs that are actuating, a recent study of Choi, Park, and coworkers demonstrated that magnetically driven crawling is also possible when the main body is anisotropically magnetized whereas the legs are nonmagnetic.^[147]

Finally, the design of soft robotics may be imagined beyond mimicking what exists in Animalia. For example, most vertebrates have four limbs; invertebrates have more feet but with tandem arrangement at the two side ends of the main body. Wang,

Shen, and coworkers developed a millimeter-sized multilegged soft robot, wherein thousands of needle-shaped magnetically susceptible legs protrude all over its thin film-shaped body made of PDMS.^[148] The multilegged robot exhibited superior locomotion speed, strong carrying capacity, and an obstacle-crossing ability on rough terrain that mimics the inner surface of a human's stomach.

3.3. Magnetic Grippers

Gripping—or grasping—is the most fundamental motion that is necessary for animals or robots to manipulate an object around it. A comprehensive Review Article by Shea and coworkers covers virtually all aspects to consider in understanding, designing, and fabricating the soft robotic gripper technology from a materials engineer's perspective.^[15] From a gripping mechanism perspective, the Review categorizes grippers systematically into three classes: grabbing by actuating fingers, immobilizing by modulating the stiffness of an enveloping object, and sticking by controlling the adhesion force. Each gripping mechanism has pros and cons with respect to the shape and texture of the object. Here, an externally applied magnetic field can be used in controlling the actuation of grabbing fingers, the stiffness of enveloping MR elastomers and fluids, and the adhesion force between the gripper and the object. Because Shea and coworkers already reviewed the field of magnetic grippers in depth^[15], this section will briefly overview noteworthy contributions that have been published afterward.

First, multiple-armed (or fingered) magnetic grippers are certainly at the center of attention. As mentioned in Section 2.4, the fabrication of arbitrarily shaped complex objects became unprecedentedly easy due to the recent rise of 3D printing, origami, and kirigami techniques. In addition, the concepts of creating a continuously distributed magnetization profile^[20] and the idea of engineered magnetic anisotropy^[21] triggered a number of innovations in multiple-armed magnetic grippers by programmed folding. Diller and coworkers demonstrated a magnetic gripper with programmed arm motion controlled by an externally applied magnetic field (Figure 5a).^[143,149] Mach and coworkers, on the other hand, controlled the motion of beaks (i.e., not having any joint) and fingers (i.e., two flexible joints separating three magnetic parts) by an electromagnet embedded at the center of the gripper.^[150] In the study, the beaks and fingers were made of MR elastomers that move their center of mass and tune their stiffness by varying the magnetomotive force of the electromagnet. Although it is not strictly magnet-controlled gripping, Gracias and coworkers harnessed magnetically driven heating to trigger dimensional changes of thermally responsive hydrogels (Figure 5b).^[151,152] By a collaboration with Misra and coworkers, the same team demonstrated steering and control of these grippers with the aid of haptic assistance.^[153]

Magnetic fields have also been utilized to enable pick-and-place-type manipulation by controlling the adhesion strength between the gripper's pedal and the object. Gong and coworkers made a cylindrical composite pad which has an MR gel core covered by a PDMS shell.^[154] In this study, dry adhesion is possible due to the intimate contact between the pad and the object. The intimate contact can be maximized because the PDMS surface of

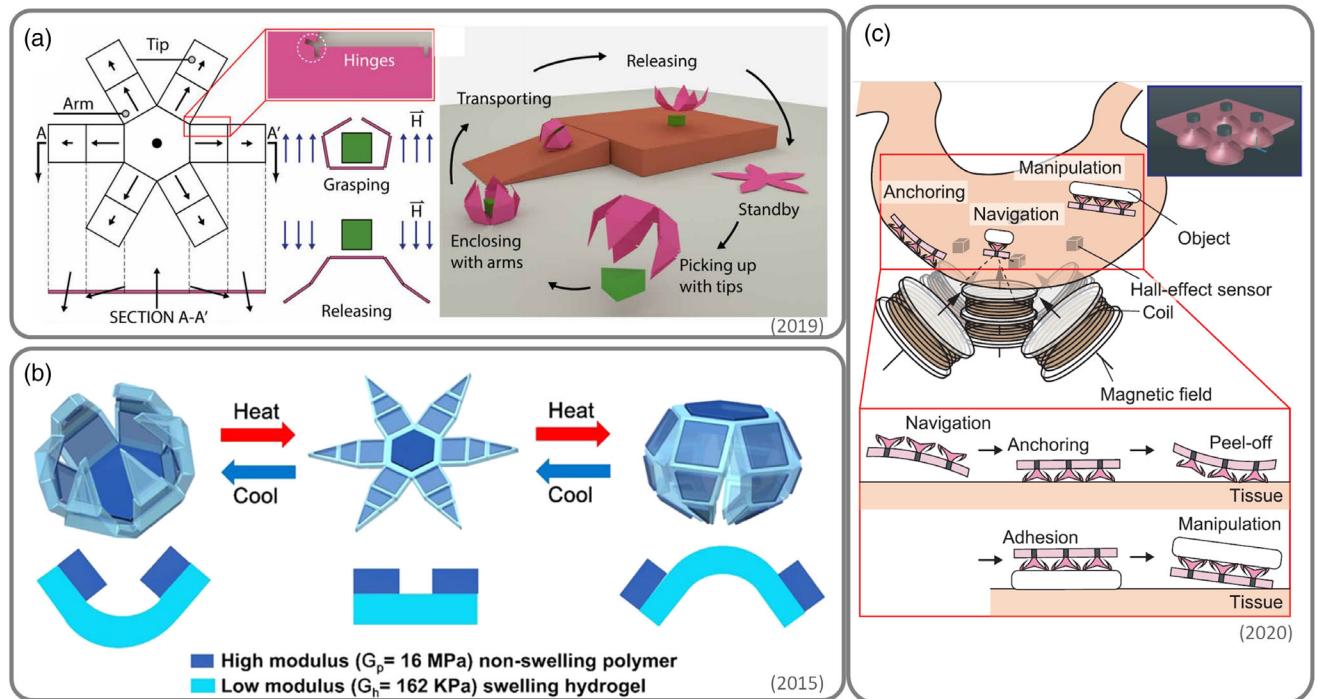


Figure 5. Three examples of recent breakthroughs in magnetically controlled soft gripper technology. a) Gripping and releasing facilitated by the programmed magnetic axis alignment technology (Reproduced with permission.^[143] Copyright 2019, AAAS). b) Folding/unfolding motions of a soft gripper controlled by magnetically induced heating and subsequent cooling of hydrogels (Reproduced with permission.^[151] Copyright 2015, The Author(s). Published by American Chemical Society). c) Adhesion, peel-off, and manipulation of an object controlled by a navigating gripper controlled by magnetically modulated suction (Reproduced with permission.^[156] Copyright 2020, IEEE).

the composite pad is so soft and viscoelastic that it can “flow” into nanoscopic roughness in the object’s surface. When a magnetic field is applied, however, the embedded MR gel core becomes stiff and deformed, and consequently, it disturbs the intimate contact by initiating local detachment (usually termed “crack” in adhesion science). The increased stiffness of the composite pad also prevents viscoelastic adhesion from occurring. As a result, this composite pad with a simple structure may provide a powerful solution for developing pick-and-place and transfer printing machinery, which is an important tool for microelectronics fabrication.^[155] Another interesting idea is from Miyashita and coworkers who attached a neodymium permanent magnet at the center of the nonadhering side of an elastomeric suction cup (Figure 5c).^[156] Using this simple idea, the authors steered the motion of the array of suction cups and then anchored the array to a desired location, all by controlling the magnetic field. The adhesion and detachment of the suction cup were also controlled by the applied magnetic field; thus, manipulation and release of an object was fully possible.

4. Biomedical Device Applications

The biggest merit of using magnetic fields in controlling robots is their ability to penetrate nearly all media. At the same time, one obvious drawback is that massive equipment, such as a triaxis-nested Helmholtz coil system, is required to have full control of the magnetic field over a small enclosed volume of

space. For these reasons, Sitti and Wiersma stated, “magnetic actuation has unique potential for medical applications of microrobots inside nontransparent tissues at high penetration depths, similar to acoustic actuation methods for microrobotics.”^[16] Sia and coworkers pointed out that medical robots require a unique combination of size, function, and material choice and thus put an emphasis on the potential of soft robots made of elastomers and gels.^[157] The ability of magnetic soft robots to navigate in fluid and terrestrial environments with gripping motions, all of which are essential in medical treatments to a targeted location, is summarized in Section 3. In Section 4, the current status of magnetic soft robotics in biomedical device applications is reviewed. Many examples in this section may include robots made of stiffer polymers than elastomers and gels, but we decided to include them to provide an overall perspective of the utilization of magnetic robots in biomedical applications.

4.1. Small-Scale Untethered Medical Robots

Small-scale robots navigating through the human body have been imaginary solutions for medical treatments.^[158] With recent works pushing the boundaries for what is possible on the small scale with micromotors,^[159] biohybrid microscale robots,^[160,161] and medical microrobots,^[118,157,162,163] such imagination is coming closer to reality. Magnetism has been well utilized in these fields to drive robots having the size scales

of milli (between 1 mm and 10 cm) and micro (between 1 μm and 1 mm).^[16] In this Review, we define “small scale” to refer to both sized scales. At this small scale, it is difficult to establish an untethered robot system including power supply, motors, and other components required for transduction. As reviewed in Section 3, controlled and programmed motion is possible simply by managing local magnetic susceptibility of constituting materials. Even when the robot has a battery component embedded within, actuation by an externally controlled field has the advantage of saving power consumption.^[164,165] Therefore, magnetic soft actuators are ideally suited for small-

scale untethered medical robot applications such as targeted drug delivery,^[125,161,166–169] microsurgery,^[170,171] deployable patch applications,^[172] cell delivery/manipulation,^[173] and localized sensing.^[174,175] The five subpanels in Figure 6 showcase representative application examples for each category.

Among the unique functionalities that small-scale untethered medical robots perform, swarming and drilling deserve special mentions. Swarming is the synchronized motion of multiple small robots to flocculate into a specific pattern at a desired location, manipulated by the applied magnetic field.^[176–178] While the terminology is defined to describe the grouping behavior

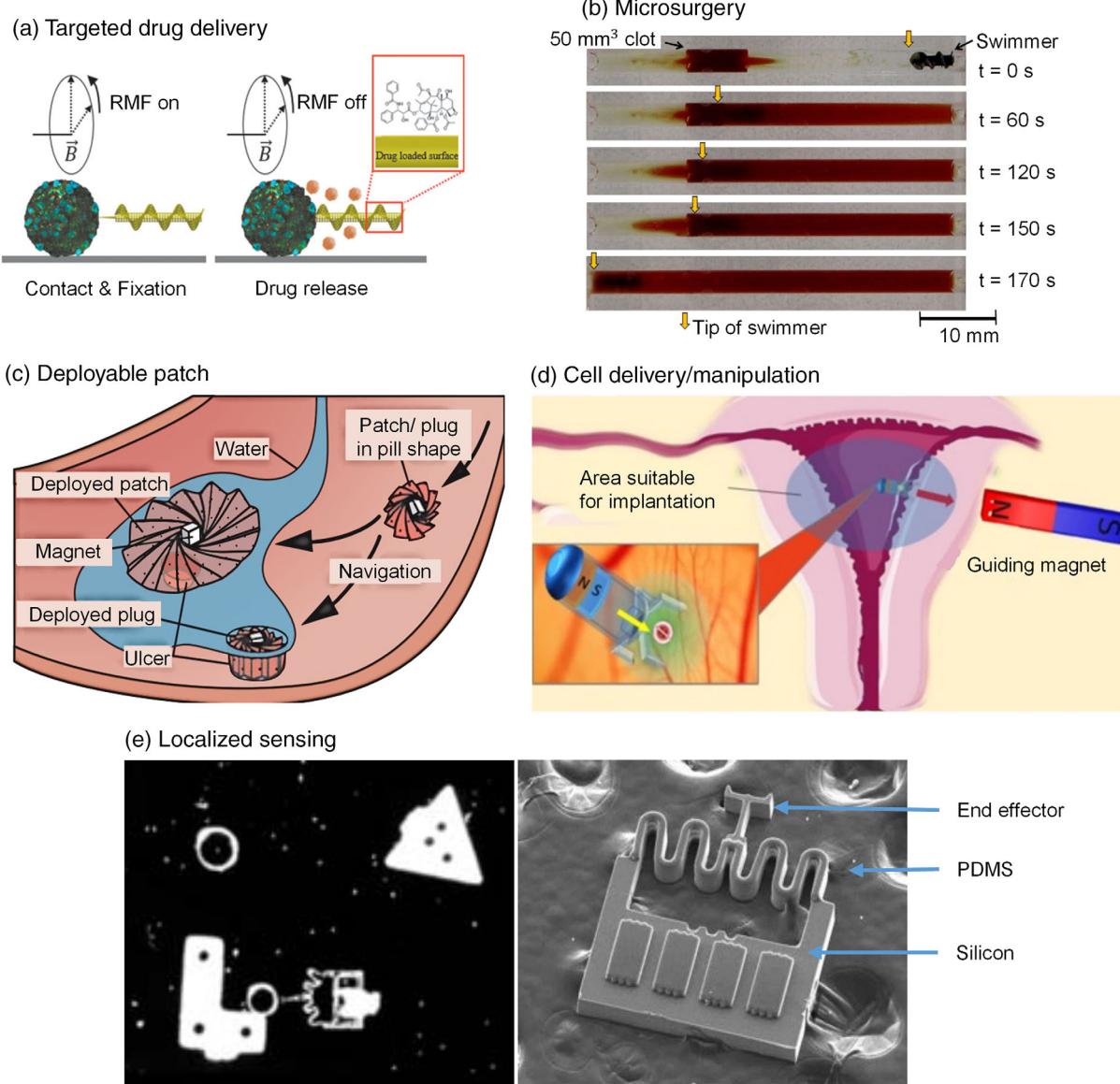


Figure 6. Five categories of applications for magnetically actuated small-scale untethered medical robots and a specific example within each. Note that many of these examples are not classified as soft robotics. a) Needle-type microrobot for targeted drug delivery by affixing to a microtissue (Reproduced with permission.^[167] Copyright 2020, Wiley). b) Drilling millirobot for blood clot removal (Reproduced with permission.^[171] Copyright 2020, IEEE). c) Deployable patch/plug for treatment of ulcers (Reproduced with permission.^[172] Copyright 2018, IEEE). d) Millirobot for transport and implantation of an embryo (Reproduced with permission.^[173] Copyright 2019, IEEE). e) Microforce-sensing microrobot for cell manipulation (Reproduced with permission.^[174] Copyright 2019, IEEE). Refer to Table 2 for more details.

Table 2. A summary of reported studies on magnetically steered small-scale untethered medical robots.

Application class	Application subclass	Tested site	Polymer/magnetic material	Movements/capabilities	Magnetic controller	Magnetic stimuli pattern	Size scale	Ref.
Targeted drug delivery	Drug delivery, hyperthermia therapy	In vitro tests of hyperthermic effect, biocompatibility, and drug release	PEGDA, PETA/Fe ₃ O ₄ nanoparticles	Swimmer- rigid helix/biodegradable	Eight electromagnetic coils in a hemispherical configuration	Rotating magnetic field	Micro	[125] (2019)
Drug delivery, manipulate small objects	In vitro test of movement on rat intestine and drug release	Ultrapure alginate/Au-Ni-Au nanorods	Surface tumbling, rolling in fluid environment/traverse rough tissue, inclines, carry cargo	Coil array ^{a)}	Rotating magnetic field	Micro-milli ^{b)}	[166] (2019)	
Drug delivery	In vitro tests of movement and drug release on cancer cells	Pine pollen/Fe ₃ O ₄ nanoparticles	Surface/swimming: rolling, tumbling, and spinning/ magnetically actuated release	Coil array ^{a)}	Rotating magnetic field	Micro	[161] (2019)	
Biodegradable Microswimmer for Theranostic Cargo Delivery and Release	In vitro tests of biodegradation, drug release, and cell labeling	Gelatin methacryloyl/iron oxide nanoparticles	Swimmer- rigid helix/swells and degrades in response to metalloproteinase-2 enzyme	Six-coil electromagnetic setup	Rotating magnetic field	Micro	[168] (2019)	
Needle-Type Microrobot for Targeted Drug Delivery by Affixing to a Microtissue	In vitro tests of affixing to microtissues, drug release on cancer cells	Photoresist/Ni and TiO ₂ layer deposited	Swimmer- rigid helix/stability due to affixing to microtissue	Electromagnetic setup (Minimag)	Rotating magnetic field	Micro	[167] (2020)	
double-layer drug-loaded microrobot	In vitro tests in acidic versus alkaline environments, drug release, and biocompatibility	[Outer layer] calcium alginate Hydrogel [inner layer] magnetic chitosan microspheres/Fe ₃ O ₄ nanoparticles	Swimmer- rolling/pH sensitive: degrades in alkaline conditions	Coil array ^{a)}	Rotating magnetic field	Micro-milli ^{b)}	[169] (2020)	
Microsurgery	Microsurgery Blood clot removal	Ex vivo blood vessel phantom	SiO ₂ / magnet ^{a)}	Direct magnetic steering ^{a)} / rotation for drilling	Stationary Helmholtz coil along the x-axis and two rotational saddle coils	Rotating magnetic field	Milli	[181] (2015)
Microsurgery	Blood clot removal	Ex vivo blood vessel phantom with simulated soft blood clot	MicroFine-ABS-like polymer/ poly(ethylene-coated NdFeB) magnet	Swimmer- rigid helix/drilling	Electromagnetic setup (Octomag)	Rotating magnetic field	Milli	[170] (2018)
Microsurgery	Blood clot removal	Ex vivo blood vessel phantom with simulated soft blood clot	3D-printed polymer ^{a)} /NdFeB magnet	Swimmer- rigid helix/drilling	Three coil pairs arranged orthogonally	Rotating magnetic field	Milli	[181] (2020)
Deployable patch	Ulcer patch or plug	Ex vivo stomach surface phantom with ulcer to be patched/plugged	Agarose gel/NdFeB magnet	Rolling motion/grow and shrink	Four pairs of coaxial electromagnetic coils	Rotating magnetic field	Milli	[172] (2018)

Table 2. Continued.

Application class	Application subclass	Tested site	Polymer/magnetic material	Movements/capabilities	Magnetic controller	Magnetic stimuli	Size scale	Ref.
Cell delivery/ manipulation	Cell manipulation: Between 50 and 500 μm	In vitro demonstration of cell manipulation	PDMS ^{a)} /NdFeB microparticles	Direct magnetic steering	Three N38 permanent magnets	n/a	Milli	[134] (2020)
	Cell Delivery: transport and implantation of embryo	Ex vivo uterus phantom	[Detachable microrobot] Photocurable resin/NdFeB magnet	Direct magnetic steering/ anchored and aligned by guiding magnet ^{a)}	Guiding magnet ^{a)}	n/a	Milli	[173] (2019)
Cell transfection, gene delivery	In vitro tests of biocompatibility, transfection, and gene delivery	Silica/ FePt L1 ₀ phase	Swimmer- rigid helix/ biocompatible	Three-axis Helmholtz coil	Rotating magnetic field	Micro	[185] (2020)	
Localized sensing	Vision based microforce sensing	In vitro manipulation and microforce sensing of cell analog	Silicon and PDMS other components/Ni body	Swimmer- direct magnetic steering ^{a)}	Four-coil electromagnetic setup	n/a	Micro	[174] (2019)
	Toxin Sensing: Clostridium difficile (C. diff) in human stool	Tests on C. diff of known concentrations and on clinical stool samples	C. lütardum spores as main body/Fe ₃ O ₄ coated on surface	Surface tumbling/C. diff detection by carbon dots	Five-coil electromagnetic setup	Rotating magnetic field	Micro	[175] (2020)

^{a)}Exact details unknown; ^{b)}Very close to 1 mm.

of living creatures such as birds, insects, and fishes, coordinated motions of individual magnetic particles in MR fluids under an applied magnetic field can be described as swarming of individual particles.^[179] In MR fluids, magnetic particles swarm to exhibit string-like patterns under a static strong magnetic field. Likewise, multiple untethered magnetic robots swarm into predictable patterns under magnetic fields. In addition, applying dynamic magnetic fields can result in various swarming patterns of the robots.^[176–178] Considering that immunologic reaction includes white blood cell swarming, understanding the behavior and applying it to medical robots can be instrumental in fighting cancer or in targeted therapies. Drilling is an effective robotic motion that can be used in unclogging a blocked blood vessel or expanding a narrowed blood vessel.^[170,171,180,181] While the current technology of catheter surgery (whose soft robotics examples are introduced in Section 4.2) has a drawback in navigating within twisted and narrowed blood vessels, small untethered magnetic robots can provide a solution to the challenge. Small-scale robots with a helical propulsion mechanism (introduced in Section 3.1) are the most frequently used for magnetically driven drilling motion.^[170,171,180,181] Table 2 shows the key features of each application examples mentioned in Section 4.1.

Whether intended or unintended, untethered small-scale robots may reside in the human body for extended time periods. Consequently, issues in biocompatibility, including issues in tissue and immunologic biocompatibility, must be addressed.^[182] Here, tissue biocompatibility refers to the local nontoxic, non-thrombogenic, noncarcinogenic, and nonmutagenic properties of small robots, and immunologic biocompatibility refers more specifically to the acute inflammatory and chronic immune responses caused by small robots. While long-term biocompatibility issues of some popular magnetic materials exist, such as neodymium alloys,^[183,184] there is increased awareness and consideration among soft robotics researchers about biocompatibility issues.^[157,185]

4.2. Magnetically Controlled Surgical Robots

There has been growing demand for novel surgical procedures that cause less trauma to patients and are safer, faster, and less demanding for doctors to perform. Many abdominal surgeries require a wide opening, which leads to longer recovery times and even to lower survival rates for some procedures. To minimize trauma and avoid damage to peripheral tissues, an endoscope, which has a tiny video camera at the end of a soft and thin tube, has been utilized to perform surgery with minimal opening of the patient's skin. There are three categories of endoscopic surgery techniques: MIS, laparoendoscopic single site (LESS), and natural orifice transluminal endoscopic surgery (NOTES), as shown in Figure 7a.^[186,187] In MIS, multiple incisions are made to allow access by long surgical tools and an endoscope. LESS differs in that there is one larger incision from which the endoscope and other mechanically flexing tools access various internal regions. NOTES accesses the abdomen from a natural orifice in the body and internal incisions where necessary.^[187] Soft robotics research, especially steerable catheters by magnetically controlled movement, is at the forefront of the innovation of surgical tools.^[186–188] These techniques have been

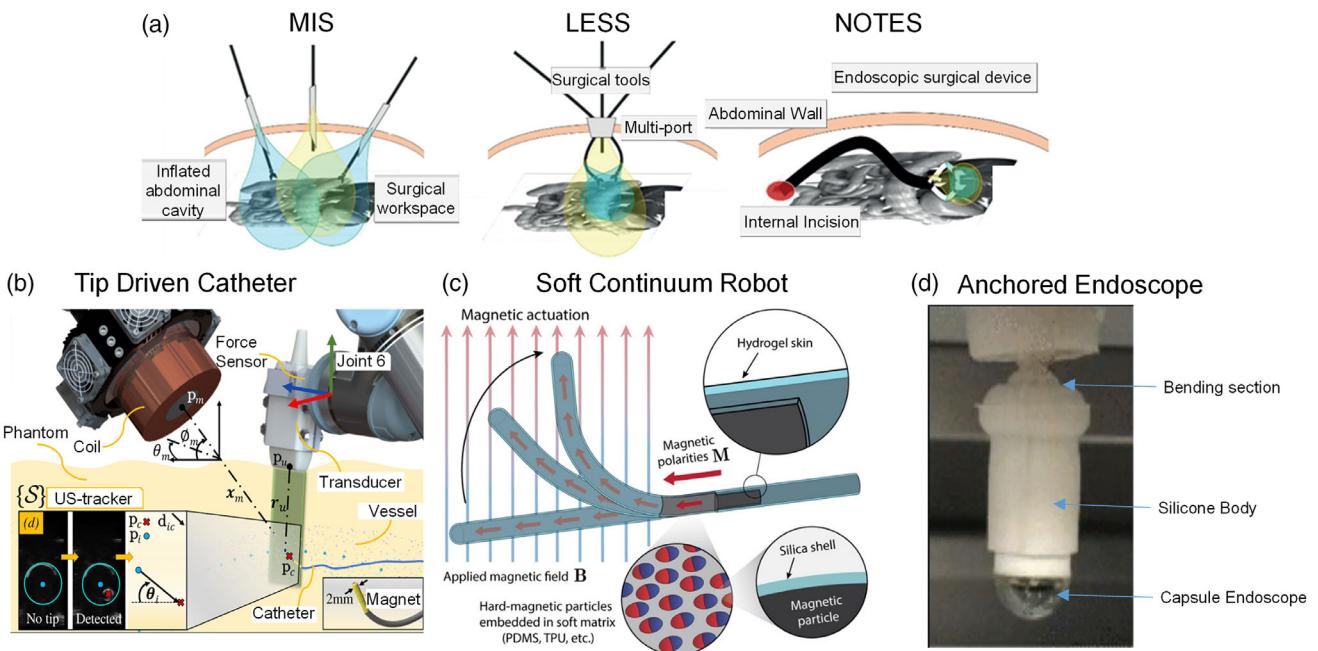


Figure 7. Examples of magnetically controlled soft robotics in endovascular and abdominal surgery. a) Different forms of alternative surgery including MIS, LESS, and NOTES (Reproduced with permission.^[186] Copyright 2016, IEEE). b) Tip-driven endovascular catheter (Reproduced with permission.^[190] Copyright 2020, IEEE). c) Fully deformable continuum manipulator for use as guide wires, etc. (Reproduced with permission.^[138] Copyright 2019, AAAS). d) Magnetically anchored and a guided soft endoscope for abdominal surgery (Reproduced with permission.^[200] Copyright 2017, IEEE).

used in endovascular and abdominal surgeries. Flexible surgical tools needed for these procedures include grafts, tubes, endoscopes, guide wires, and catheters.^[188] Innovating these surgical tools to have finesse, controllability, and smart functionality provides exceptional opportunities for soft actuators with magnetically responsive elastomers and hydrogels; this is what soft materials, more specifically gels and elastomers, can offer in an exclusively unique manner.

Endovascular surgery has challenging requirements particularly for the navigation of catheters and guide wires. Consequently, magnetically steered catheters and guide wires, whose motion of the near-tip region is steered by the applied magnetic field, have been demonstrated in various sizes.^[138,189–193] These catheters and guide wires may have permanent magnets^[190–192] or magnetic microparticles^[138,193] embedded and distributed inside the tube, as shown in Figure 7b,c, respectively. A lubrication layer made of hydrogel can reduce the risk of injury.^[138] As a different application of the magnetically steered catheter technology, an endolaser for treatment of retina conditions is also actively researched.^[194]

Surgical procedures that are assisted by magnetic control is a fairly mature technology. Current clinical examples include rigid retractors, endoscopes, and manipulators.^[195–199] Some of these techniques, such as Levita™ Magnetic Surgical System,^[195] have received the United States Food and Drug Administration (FDA) approval. Recently, concepts from soft robotics are introduced in this important medical field. For example, Li and coworkers developed a novel endoscope that can be anchored in abdominal cavity by magnetic force, whereas its angular motion can be controlled by an externally controlled magnetic

field (Figure 7d).^[200–202] Menciassi and coworkers used ferromagnetic material to anchor a soft retractor, whereas the stiffness of the retractor is controlled pneumatically utilizing the concept of granular jamming.^[203] Generally speaking, the inclusion of magnetically controlled soft robotics in clinical studies of endoscopic or catheter devices is still at its infancy. While many promising new concepts have been suggested from a soft robotics perspective, the motion controllability of these devices can be improved by further developments of the kinematic model and path planning algorithms.^[204] Finally, Table 3 shows the key features of each application examples mentioned in Section 4.2.

5. Conclusion and Outlook

Among all known sources of stimuli that activate the actuation of soft robotics, magnetic fields have an unmatched advantage in their ability to penetrate deeply into various materials. Moreover, magnetically susceptible materials can respond quickly to dynamic changes in the magnetic field. An array of multiple electromagnets or moving permanent magnets can create a spatial gradient within an enclosed volume, whereas the gradient can be controlled precisely. All these factors lead to the conclusion that actuating soft robotic devices by a magnetic field is an exceptionally attractive option for various biomedical applications including MIS and targeted drug delivery. When these medical devices are untethered, controlling the swimming and terrestrial locomotions by an applied magnetic field is the first step for a successful application. For both

Table 3. A summary of reported studies on magnetically controlled surgical robots.

Device	Applications	Target organ (or location)	Tested site	Polymer/magnetic material	Movement type	Magnetic controller	Ref. (year)
Catheter	Magnetically actuated endovascular catheter	Heart, arteries	Ex vivo human abdominal arteries phantom	HDPE catheter/NdFeB magnets	Tip driven	Single electromagnet positioned by a robotic arm	[190] (2020)
	Variable stiffness catheter	Heart chambers, gastrointestinal tract	Demonstration of some movements in free space (no phantom)	Silicone tube Low-melting-point alloy for variable stiffness/ magnet ^{a)}	Tip driven	CardioMag (Aeon Phocus) ^{a)}	[205] (2020)
	Magnetic microcatheter for vitreoretinal targeted drug delivery	Retina	Ex vivo human eye phantom	Long flexible monofilament nylon segment/NdFeB magnet	Tip driven	Eight electromagnetic coils	[191] (2017)
Guide wire	steering sub-millimeter-diameter guide wires	Coronary arteries	Ex vivo human coronary arteries phantom	PDMS/NdFeB magnets	Tip driven	Eight Helmholtz/ Maxwell electromagnetic coils (OctoMag)	[192] (2019)
	fully flexible, shape forming tentacle robot : catheter or guide wire	Vascular networks	Demonstration of some movements in free space (no phantom)	Silicone/NdFeB microparticles in silicone	Fully deformable	Helmholtz coils ^{a)}	[193] (2020)
	Vascular guide wire, laser delivery, Scalable to sub-millimeter	Vascular networks	Ex vivo cerebrovascular phantom network	PDMAA hydrogel skin/ NdFeB microparticles in Silica (PDMS) shell	Fully deformable	Pair of Helmholtz coils: model testing NdFeB magnet: Demonstrations	[138] (2019)
Endolaser	Endolaser for retinal treatments	Retina	Ex vivo human eye phantom	Polyimide tube/NdFeB magnet	Tip driven	Eight Helmholtz/ Maxwell electromagnetic coils (OctoMag)	[194] (2019)
Endoscope	Magnetically anchored and guided soft endoscope for abdominal surgery	Abdominal cavity	Ex vivo experiment using porcine ribs	RTV silicone/NdFeB magnets	Anchored, soft bending	External magnet controlled by robotic arm [R4.2.18]	[200,202] (2017, 2020)
Retractor	Magnetic soft retractor for use in abdominal surgery	Abdominal cavity	Ex vivo surgery on porcine Intestine, liver, and stomach In vivo surgery on a domestic pig	Ecoflex 0030/NdFeB magnets	Anchored, soft bending, variable stiffness	Two NdFeB permanent magnets	[203] (2019)

^{a)}Exact details unknown.

untethered and tethered devices, gripping is the most fundamental and important mode of motion to realize nearly any practical application. At the current stage of magnetically controlled soft robotics development, we consider the use of magnetic elastomers and gels a potential game changer because these magnetic soft actuators can maximize the merits that soft robotics can bring to the field. In addition to their rubber-like mechanical properties, elastomers and gels can be manufactured in nearly any shape. Moreover, controlled magnetization can enable the precisely programmed motion of the magnetic soft actuator by applying a magnetic field with controlled patterns. In this Review, we featured two key ideas to achieve complex programmed motions, namely, continuously distributed magnetization and programmed magnetic axis alignment methods.

During the past decade, the field of soft actuators has seen rapid advancements due to recent innovations in soft material fabrication technologies such as 3D printing, origami/kirigami, tough hydrogels, mechanical metamaterials, and liquid metal-

jected elastomers. The new wave of material technologies has started to stimulate notable innovations in magnetic soft actuator material development. Some of these materials innovations, however, are yet to be incorporated in soft robotics systems. Despite a few intrinsic problems of elastomers and gels that make precisely controlled actuation difficult, there are many unique advantages to justify the use of magnetic soft actuators in soft robotic devices, especially in biomedical applications.^[12,16] Establishing a modeling framework to predict the motion of a magnetic soft actuator under applied magnetic field will be essential for precise control.^[40–42] The successful model can motivate robotics engineers to use magnetic elastomers and gels in their design when precise control is required. All these breakthroughs can also influence a closely related field of soft sensors.^[206–209] In summary, the fields of magnetic soft actuators and soft robotics are rapidly evolving due to recent breakthroughs in materials engineering, and the potentials of these novel materials are just beginning to unfold.

Acknowledgements

This work was supported by the Natural Sciences and Engineering Research Council of Canada (NSERC) by funding through Discovery Grant (RGPIN-2019-04952).

Conflict of Interest

The authors declare no conflict of interest.

Keywords

elastomers, gels, magnetic controls, soft actuators, soft robotics

Received: August 5, 2020

Revised: September 27, 2020

Published online: December 9, 2020

- [1] S. Kim, C. Laschi, B. Trimmer, *Trends Biotechnol.* **2013**, *31*, 287.
- [2] Y. N. Harari, *Homo Deus: A Brief History of Tomorrow*, Penguin Random House, Toronto, ON **2016**.
- [3] G. M. Whitesides, *Angew. Chem., Int. Ed.* **2018**, *57*, 4258.
- [4] A. Billard, D. Kragic, *Science* **2019**, *364*, eaat8414.
- [5] C. Majidi, *Soft Robotics* **2014**, *1*, 5.
- [6] M. Cianchetti, C. Laschi, A. Menciassi, P. Dario, *Nat. Rev. Mater.* **2018**, *3*, 143.
- [7] S. Bauer, S. Bauer-Gogonea, I. Graz, M. Kaltenbrunner, C. Keplinger, R. Schwodiauer, *Adv. Mater.* **2014**, *26*, 149.
- [8] D. Zhammuratova, H.-J. Chung, *ACS Appl. Polym. Mater.* **2020**, *2*, 1073.
- [9] P. Boyraz, G. Runge, A. Raatz, *Actuators* **2018**, *7*, 48.
- [10] Y. Yang, Y. Wu, C. Li, X. Yang, W. Chen, *Adv. Intell. Syst.* **2020**, *2*, 1900077.
- [11] A. Zolfagharian, A. Z. Kouzani, S. Y. Khoo, A. A. A. Mohjadam, I. Gibson, A. Kaynak, *Sens. Actuators, A* **2016**, *250*, 258.
- [12] L. Hines, K. Peterson, G. Z. Lum, M. Sitti, *Adv. Mater.* **2017**, *29*, 1603483.
- [13] J. M. McCracken, B. R. Donovan, T. J. White, *Adv. Mater.* **2020**, *32*, 1906564.
- [14] A. Miriyev, K. Stack, H. Lipson, *Nat. Commun.* **2017**, *8*, 596.
- [15] J. Shintake, V. Cacucciolo, D. Floreano, H. Shea, *Adv. Mater.* **2018**, *30*, 1707035.
- [16] M. Sitti, D. S. Wiersma, *Adv. Mater.* **2020**, *32*, 1906766.
- [17] N. Park, J. Kim, *Adv. Intell. Syst.* **2020**, *2*, 1900135.
- [18] M. Zrinyi, *Smart Polymers and Their Applications* (Eds: M. R. Aguilar, J. San Román), Elsevier, Amsterdam/New York **2014**, pp. 134–165.
- [19] A. Zolfagharian, A. Z. Kouzani, S. Y. Khoo, A. A. A. Moghadam, I. Gibson, A. Kaynak, *Sens. Actuators, A* **2016**, *250*, 258.
- [20] E. Diller, J. Zhuang, G. Z. Lum, M. R. Edwards, M. Sitti, *Appl. Phys. Lett.* **2014**, *104*, 174101.
- [21] J. Kim, S. E. Chung, S.-E. Choi, H. Lee, J. Kim, S. Kwon, *Nat. Mater.* **2011**, *10*, 747.
- [22] L. Xu, T. C. Shyu, N. A. Kotov, *ACS Nano* **2017**, *11*, 7587.
- [23] M. Kamachi, *J. Macromol. Sci. C: Rev.* **2002**, *C42*, 541.
- [24] J. Thevenot, H. Oliveira, O. Sandre, S. Lecommandoux, *Chem. Soc. Rev.* **2013**, *42*, 7099.
- [25] V. Q. Nguyen, A. S. Ahmed, R. V. Ramanujan, *Adv. Mater.* **2012**, *24*, 4041.
- [26] P. C. Painter, M. M. Coleman, *Essentials of Polymer Science and Engineering*, DESTech, Lancaster, PA **2009**.
- [27] S. Slomkowski, J. V. Alemán, R. G. Gilbert, M. Hess, K. Horie, R. G. Jones, P. Kubisa, I. Meisel, W. Mormann, S. Penczek, R. F. T. Stepto, *Pure Appl. Chem.* **2011**, *83*, 2229.
- [28] H. Haider, C. H. Yang, W. J. Zheng, J. H. Yang, M. X. Wang, S. Yang, M. Zrinyi, Y. Osada, Z. Suo, Q. Zhang, J. Zhou, Y. M. Chen, *Soft Matter* **2015**, *11*, 8253.
- [29] X. Hu, G. Nian, X. Liang, L. Wu, T. Yin, H. Lu, S. Qu, W. Yang, *ACS Appl. Mater. Interfaces* **2019**, *11*, 10292.
- [30] H.-J. Chung, H. Charaya, L. Liu, X. Li, *Hybrid Organic-Inorganic Interfaces: Towards Advanced Functional Materials* (Eds: M-H. Delville, A. Taubert, Vol. 2, Wiley, New York/Chichester, UK **2018**, pp. 535–580).
- [31] C. Creton, *Macromolecules* **2017**, *50*, 8297.
- [32] J. P. Gong, *Soft Matter* **2010**, *6*, 2583.
- [33] T. Chen, X. Zhang, X. Yan, B. Zhang, J. Jiang, D. Huang, M. Qi, R. Sun, *Adv. Eng. Mater.* **2019**, *21*, 1801255.
- [34] T. N. Do, H. Phan, T.-Q. Nguyen, Y. Vissel, *Adv. Funct. Mater.* **2018**, *28*, 1800244.
- [35] J. D. Carlson, M. R. Jolly, *Mechatronics* **2000**, *10*, 555.
- [36] S. O. Kasap, *Principles of Electronic Materials and Devices*, 4th ed., McGraw-Hill, New York, NY **2017**.
- [37] S. A. Majetic, T. Wen, O. T. Mefford, *MRS Bull.* **2013**, *38*, 899.
- [38] M. Lokander, B. Stenberg, *Polym. Test.* **2003**, *22*, 245.
- [39] Y. Li, Z. Qi, J. Yang, M. Zhou, X. Zhang, W. Ling, Y. Zhang, Z. Wu, H. Wang, B. Ning, H. Xu, W. Huo, X. Huang, *Adv. Funct. Mater.* **2019**, *29*, 1904977.
- [40] R. Zhao, Y. Kim, S. A. Chester, P. Sharma, X. Zhao, *J. Mech. Phys. Solids* **2019**, *124*, 244.
- [41] L. Wang, Y. Kim, C. F. Guo, X. Zhao, *J. Mech. Phys. Solids* **2020**, *142*, 104045.
- [42] D. Garcia-Gonzalez, *Smart Mater. Struct.* **2019**, *28*, 085020.
- [43] Y. Wang, Y. Hu, L. Chen, X. Gong, W. Jiang, P. Zhang, Z. Chen, *Polym. Test.* **2006**, *25*, 262.
- [44] H. L. Ding, Y. X. Zhang, S. Wang, J. M. Xu, S. C. Xu, G. H. Li, *Chem. Mater.* **2012**, *24*, 4572.
- [45] L. Zheng, T. N. T. Tran, D. Zhammuratova, D. G. Ivey, H.-J. Chung, *ACS Appl. Nano Mater.* **2019**, *2*, 6982.
- [46] D. P. Facchi, A. L. Cazett, E. A. Canesin, V. C. Almeida, E. G. Bonafé, M. J. Kipper, A. F. Martins, *Chem. Eng. J.* **2018**, *337*, 595.
- [47] W. Dai, H. Xu, C. Zhang, Y. Li, H. Pan, H. Wang, G. Wei, X. Huang, *Adv Electron. Mater.* **2019**, *5*, 1900111.
- [48] Y. Li, G. Huang, X. Zhang, B. Li, Y. Chen, T. Lu, T. J. Lu, F. Xu, *Adv. Funct. Mater.* **2013**, *23*, 660.
- [49] W. Zhao, K. Odelius, U. Edlund, C. Zhao, A.-C. Albertsson, *Biomacromolecules* **2015**, *16*, 2522.
- [50] L. Jiang, P. Liu, *ACS Sustainable Chem. Eng.* **2014**, *2*, 1785.
- [51] R. Messing, N. Frickel, L. Belkoura, R. Strey, H. Rahn, S. Odenbach, A. M. Schmidt, *Macromolecules* **2011**, *44*, 2990.
- [52] B. A. Evans, B. L. Fiser, W. J. Prins, D. J. Rapp, A. R. Shields, D. R. Glass, R. Superfine, *J. Magn. Magn. Mater.* **2012**, *324*, 501.
- [53] R. Fuhrer, E. K. Athanassiu, N. A. Luechinger, W. J. Stark, *Small* **2009**, *5*, 383.
- [54] B. G. Bellan, *Int. J. Mod. Phys. B* **2003**, *16*, 2447.
- [55] C. Majidi, R. J. Wood, *Appl. Phys. Lett.* **2010**, *97*, 164104.
- [56] S. B. Choi, Y. M. Han, *Magnetorheological Fluid Technology*, CRC Press, Boca Raton, FL **2013**.
- [57] J. de Vicente, D. J. Klingenberg, R. Hidalgo-Alvarez, *Soft Matter* **2011**, *7*, 3701.
- [58] Y. Li, J. Li, W. Li, H. Du, *Smart Mater. Struct.* **2014**, *23*, 123001.
- [59] M. R. Jolly, J. D. Carlson, B. C. Munoz, T. A. Bullins, *J. Intell. Mater. Syst. Struct.* **1996**, *7*, 613.
- [60] T. Shica, A. Okada, T. Kurauchi, *J. Appl. Polym. Sci.* **1995**, *58*, 787.

- [61] J. M. Ginder, S. M. Clark, W. F. Schlotter, M. E. Nichols, *Int. J. Mod. Phys. B* **2002**, *16*, 2412.
- [62] M. Zrinyi, L. Barsi, A. Buki, *Polym. Gels Networks* **1997**, *5*, 415.
- [63] M. Zrinyi, D. Szabo, H.-G. Kilian, *Polym. Gels Networks* **1998**, *6*, 441.
- [64] D. Szabo, G. Szeghy, M. Zrinyi, *Macromolecules* **1998**, *31*, 6541.
- [65] Z. Varga, G. Filipcsei, M. Zrinyi, *Polymer* **2005**, *46*, 7779.
- [66] R. Ahamed, S.-B. Choi, M. M. Ferdaus, *J. Intel. Mater. Syst. Struct.* **2018**, *29*, 2051.
- [67] S. S. Sun, J. Yang, W. H. Li, H. Du, G. Alici, T. H. Yan, M. Nakano, *Mech. Syst. Signal Process.* **2017**, *83*, 371.
- [68] G. V. Stepanov, D. Y. Borin, Y. L. Raikher, P. V. Melenev, N. S. Perov, *J. Phys.: Condens. Matter* **2008**, *20*, 204121.
- [69] T. Caykara, D. Yoruk, S. Demirci, *J. Appl. Polym. Sci.* **2009**, *112*, 800.
- [70] A. K. Bastola, E. Ang, M. Paudel, L. Li, *Colloids Surf. A* **2019**, *583*, 123975.
- [71] J. Yao, Y. Sun, Y. Wang, Q. Fu, Z. Xiong, Y. Liu, *Compos. Sci. Technol.* **2018**, *162*, 170.
- [72] S. Qi, J. Fu, Y. Xie, Y. Li, R. Gan, M. Yu, *Compos. Sci. Technol.* **2019**, *183*, 107817.
- [73] J. Zhang, H. Pang, Y. Wang, X. Gong, *Compos. Sci. Technol.* **2020**, *191*, 108079.
- [74] Y. Zhou, L. Li, W. Li, S. Wen, L. Jiang, S. Jerrams, J. Ma, S. Chen, *Smart Mater. Struct.* **2020**, *29*, 055005.
- [75] M. Cvek, A. Zahoranova, M. Mrlik, P. Sramkova, A. Minarik, M. Sedlack, *Colloids Surf. B* **2020**, *190*, 110912.
- [76] Q. Shu, L. Ding, X. Gong, T. Hu, S. Xuan, *Smart Mater. Struct.* **2020**, *29*, 025010.
- [77] C. Wu, Q. Zhang, X. Fan, Y. Song, Q. Zheng, *J. Intel. Mater. Syst. Struct.* **2019**, *30*, 1084.
- [78] A. Alkhafaf, A. hoosihiar, J. Dargahi, *Composites, Part B* **2020**, *190*, 107888.
- [79] H. Yuk, X. Zhao, *Adv. Mater.* **2017**, *30*, 1704028.
- [80] R. Levato, T. Jungst, R. G. Scheuring, T. Blunk, J. Groll, J. Malda, *Adv. Mater.* **2020**, *32*, 1906423.
- [81] D. Chirmene, R. Kaunas, A. K. Gahawar, *Adv. Mater.* **2020**, *32*, 1902026.
- [82] L.-Y. Zhou, J. Fu, Y. He, *Adv. Funct. Mater.* **2020**, *30*, 2000187.
- [83] T. J. Wallin, J. Pikul, R. F. Shepherd, *Nat. Rev. Mater.* **2018**, *3*, 84.
- [84] M. Shaffner, J. A. Faber, L. Pianegonda, P. A. Ruhs, F. Coulter, A. R. Studart, *Nat. Commun.* **2018**, *9*, 878.
- [85] A. K. Bastola, M. Paudel, L. Li, *Polymer* **2018**, *149*, 213.
- [86] A. K. Bastola, V. T. Hoang, L. Li, *Mater. Des.* **2017**, *114*, 391.
- [87] J. A. Jackson, M. C. Messner, N. A. Dudukovic, W. L. Smith, L. Bekker, B. Moran, A. M. Golobic, A. J. Pascall, E. B. Duoss, K. J. Loh, C. M. Spadaccini, *Sci. Adv.* **2018**, *4*, eaau6419.
- [88] Z. Ji, C. Yan, B. Yu, X. Wang, F. Zhou, *Adv. Mater. Interfaces* **2017**, *4*, 1700629.
- [89] G. Chatzipiridis, S. Gervasoni, C. Fischer, O. Ergeneman, E. Pellicer, B. J. Nelson, S. Pané, *Adv. Intell. Syst.* **2019**, *1*, 1900069.
- [90] J. M. McCracken, B. M. Rauzan, J. C. E. Kjellman, H. Su, S. A. Rogers, R. G. Nuzzo, *Adv. Funct. Mater.* **2019**, *29*, 1806723.
- [91] K. Saito, S. Nomura, S. Yamamoto, R. Niyyama, Y. Okabe, *Proc. Natl. Acad. Sci.* **2017**, *114*, 5624.
- [92] T. Ozaki, N. Ohta, K. Hamaguchi, *Appl. Sci.* **2020**, *10*, 3771.
- [93] R. Baumgartner, A. Kogler, J. M. Stadlbauer, C. C. Foo, R. Kaltseis, M. Baumgartner, G. Mao, C. Keplinger, S. J. A. Koh, N. Arnold, Z. Suo, M. Kaltenbrunner, S. Bauer, *Adv. Sci.* **2020**, *7*, 1903391.
- [94] Z. Zhao, Y. Wang, H. Jiang, *Proc. Natl. Acad. Sci.* **2018**, *115*, 2032.
- [95] S. Mintchev, J. Shintake, D. Floreano, *Sci. Robot.* **2018**, *3*, eaau0275.
- [96] T. Jamin, C. Py, E. Falcon, *Phys. Rev. Lett.* **2011**, *107*, 204503.
- [97] A. G. Izard, L. Valdevit, *Adv. Eng. Mater.* **2020**, *22*, 1901019.
- [98] Y. Kim, H. Yuk, R. Zhao, S. A. Chester, X. Zhao, *Nature* **2018**, *558*, 274.
- [99] S. Wu, Q. Ze, R. Zhang, N. Hu, Y. Cheng, F. Yang, R. Zhao, *ACS Appl. Mater. Interfaces* **2019**, *11*, 41649.
- [100] M. D. Dickey, *Adv. Mater.* **2017**, *29*, 1606425.
- [101] S. W. Jin, J. Park, S. Y. Hong, H. Park, Y. R. Jeong, J. Park, S.-S. Lee, J. S. Ha, *Sci. Rep.* **2015**, *5*, 11695.
- [102] R. Guo, L. Sheng, H. Y. Gong, J. Liu, *Sci. China Tech. Sci.* **2018**, *61*, 516.
- [103] G. Mao, M. Drack, M. Karami-Mosammam, D. Wirthl, T. Stockinger, R. Schwodauer, M. Kaltenbrunner, *Sci. Adv.* **2020**, *6*, eabc0251.
- [104] Q. Zhao, H. J. Qi, T. Xie, *Prog. Polym. Sci.* **2015**, *49–50*, 79.
- [105] A. M. Schmidt, *Macromol. Rapid Commun.* **2006**, *27*, 1168.
- [106] R. Mohr, K. Kratz, T. Weigel, M. Lucka-Gabor, M. Moneke, A. Lendlein, *Proc. Natl. Acad. Sci.* **2006**, *103*, 3540.
- [107] U. Narendra Kumar, K. Kratz, W. Wagermaier, M. Behl, A. Lendlein, *J. Mater. Chem.* **2010**, *20*, 3404.
- [108] U. Narendra Kumar, K. Kratz, M. Heuchel, M. Behl, A. Lendlein, *Adv. Mater.* **2011**, *23*, 4157.
- [109] M. Y. Razzaq, M. Behl, A. Lendlein, *Adv. Funct. Mater.* **2012**, *22*, 184.
- [110] J. A.-C. Liu, J. H. Gillen, S. R. Mishra, B. A. Evans, J. B. Tracy, *Sci. Adv.* **2019**, *5*, eaaw2897.
- [111] Q. Ze, X. Kuang, S. Wu, J. Wong, S. M. Montgomery, R. Zhang, J. M. Kovitz, F. Yang, H. J. Qi, R. Zhao, *Adv. Mater.* **2019**, *31*, 1906657.
- [112] S. Ghosh, T. Cai, *J. Phys. D: Appl. Phys.* **2010**, *43*, 415504.
- [113] J. Tang, Q. Yin, Y. Qiao, T. Wang, *ACS Appl. Mater. Interfaces* **2019**, *11*, 21194.
- [114] J. J. Abbott, K. E. Peyer, M. C. Lagomarsino, L. Zhang, L. X. Dong, I. K. Kaliakatsos, B. J. Nelson, *Int. J. Robot. Res.* **2009**, *28*, 1434.
- [115] E. Al Khatib, A. Bhattacharjee, P. Razzaghi, L. W. Rogowski, M. J. Kim, Y. Hurmuzlu, *IEEE Robot. Autom. Lett.* **2020**, *5*, 2958.
- [116] D. C. Meeker, E. H. Maslen, R. C. Ritter, F. M. Creighton, *IEEE Trans. Magn.* **1996**, *32*, 320.
- [117] B. J. Nelson, I. K. Kaliakatsos, J. J. Abbott, *Annu. Rev. Biomed. Eng.* **2010**, *12*, 55.
- [118] M. Sitti, H. Ceylan, W. Hu, J. Giltinan, M. Turan, S. Yim, E. Diller, *Proc. IEEE* **2015**, *103*, 205.
- [119] F. Qiu, R. Mhanna, L. Zhang, Y. Ding, S. Fujita, B. J. Nelson, *Sens. Actuators, B* **2014**, *196*, 676.
- [120] M. Medina-Sanchez, L. Schwarz, A. K. Meyer, F. Hebenstreit, O. G. Schmidt, *Nano Lett.* **2016**, *16*, 555.
- [121] X.-Z. Chen, M. Hoop, F. Mushtaq, E. Siringil, C. Hu, B. J. Nelson, S. Pané, *Appl. Mater. Today* **2017**, *9*, 37.
- [122] H. O. Caldag, S. Yesilyurt, *J. Fluid Struct.* **2019**, *90*, 164.
- [123] P. Mandal, G. Patil, H. Kakoty, A. Ghosh, *Acc. Chem. Res.* **2018**, *51*, 2689.
- [124] T. Xu, J. Yu, C.-I. Vong, B. Wang, X. Wu, L. Zhang, *IEEE/ASME Trans. Mech.* **2019**, *24*, 924.
- [125] J. Park, C. Jin, S. Lee, J.-Y. Kim, H. Choi, *Adv. Healthcare Mater.* **2019**, *8*, 1900213.
- [126] S. Sudo, S. Segawa, T. Honda, *J. Intell. Mater. Syst. Struct.* **2006**, *7*, 729.
- [127] S. Guo, Q. Pan, M. B. Khamesee, *Microsyst. Technol.* **2008**, *14*, 307.
- [128] P. Garstecki, P. Tierno, D. B. Weibel, F. Sagues, G. M. Whitesides, *J. Phys.: Condens. Matter* **2009**, *21*, 204110.
- [129] J. Zhang, E. Diller, *Soft Robot.* **2018**, *5*, 761.
- [130] B. Jang, E. Gutman, N. Stucki, B. F. Seitz, P. D. Wendel-Garcia, T. Newton, J. Pokki, O. Ergeneman, S. Pane, Y. Or, B. J. Nelson, *Nano Lett.* **2015**, *15*, 4829.
- [131] Z. Ren, W. Hu, X. Dong, M. Sitti, *Nat. Commun.* **2019**, *10*, 2703.
- [132] M. Su, T. Xu, Z. Lai, C. Huang, J. Liu, X. Wu, *IEEE Robot Autom. Lett.* **2020**, *5*, 806.
- [133] J. Tang, C. Yao, Z. Gu, S. Jung, D. Luo, D. Yang, *Angew. Chem., Int. Ed.* **2020**, *59*, 2490.

- [134] X. Bai, D. Chen, W. Zhang, H. Ossian, Y. Chen, Y. Feng, L. Feng, F. Arai, *Micromachines* **2020**, *11*, 231.
- [135] H. Li, G. Go, S. Y. Ko, J.-O. Park, S. Park, *Smart Mater. Struct.* **2016**, *25*, 027001.
- [136] S. Yu, N. Ma, H. Yu, H. Sun, X. Chang, Z. Wu, J. Deng, S. Zhao, W. Wang, G. Zhang, W. Zhang, Q. Zhao, T. Li, *Nanomaterials* **2019**, *9*, 1672.
- [137] C. W. Shields, Y.-K. Kim, K. Han, A. C. Murphy, A. J. Scott, N. L. Abbott, O. D. Velev, *Adv. Intell. Syst.* **2020**, *2*, 1900114.
- [138] Y. Kim, G. A. Parada, S. Liu, X. Zhao, *Sci. Robot.* **2019**, *4*, eaax7329.
- [139] K. Jung, J. C. Koo, J.-D. Nam, Y. K. Lee, H. R. Choi, *Bioinspir. Biomim.* **2007**, *2*, S42.
- [140] T. Alhart, GE Research to Demonstrate Giant Earthworm-Like Robot for Superfast, Ultra-efficient Tunnel Digging, <https://www.ge.com/research/newsroom/ge-research-demonstrate-giant-earthworm-robot-superfast-ultra-efficient-tunnel-digging> (accessed: July 2020).
- [141] T. Shen, M. G. Font, S. Jung, M. L. Gabriel, M. P. Stoykovich, F. J. Vernerey, *Sci. Rep.* **2017**, *7*, 16178.
- [142] W. Wang, J.-Y. Lee, H. Rodrigue, S.-H. Song, W.-S. Chu, S.-H. Ahn, *Bioinspir. Biomim.* **2014**, *9*, 046006.
- [143] T. Xu, J. Zhang, M. Salehizadeh, O. Onaizah, E. Diller, *Sci. Robot.* **2019**, *4*, eaav4494.
- [144] W. Hu, G. Z. Lum, M. Mastrangeli, M. Sitti, *Nature* **2018**, *554*, 81.
- [145] X. Du, H. Cui, T. Xu, C. Huang, Y. Wang, Q. Zhao, Y. Xu, X. Wu, *Adv. Funct. Mater.* **2020**, *30*, 1909202.
- [146] V. K. Venkiteswaran, L. F. Pena Samaniego, J. Sikorski, S. Misra, *IEEE Robot Autom. Lett.* **2019**, *4*, 1753.
- [147] S. Ijaz, H. Li, M. C. Hoang, C.-S. Kim, D. Bang, E. Choi, J. O. Park, *Sens. Actuators, A* **2020**, *301*, 111707.
- [148] H. Lu, M. Zhang, Y. Yang, Q. Huang, T. Fukuda, Z. Wang, Y. Shen, *Nat. Commun.* **2018**, *9*, 3944.
- [149] J. Zhang, O. Onaizah, K. Middleton, L. You, E. Diller, *IEEE Robot. Autom. Lett.* **2017**, *2*, 840.
- [150] V. Skrivan, O. Sodomka, F. Mach, in *Proc. of 2nd IEEE Int. Conf. on Soft Robotics (RoboSoft)*, IEEE, Seoul, Korea **2019**, pp. 126–130.
- [151] 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. Interfaces* **2015**, *7*, 3398.
- [152] K. Kobayashi, C. Yoon, S. H. Oh, J. V. Pagaduan, D. H. Gracias, *ACS Appl. Mater. Interfaces* **2019**, *11*, 151.
- [153] C. Pacchierotti, F. Ongaro, F. van den Brink, C. Yoon, D. Prattichizzo, D. H. Gracias, S. Misra, *IEEE Trans. Autom. Sci. Eng.* **2018**, *15*, 290.
- [154] H. Pang, L. Pei, J. Xu, S. Cao, Y. Wang, X. Gong, *Compos. Sci. Technol.* **2020**, *192*, 108115.
- [155] S. Y. Yang, A. Carlson, H. Cheng, Q. Yu, N. Ahmed, J. Wu, S. Kim, M. Sitti, P. M. Ferreira, Y. Huang, J. A. Rogers, *Adv. Mater.* **2012**, *24*, 2117.
- [156] H. Iwasaki, F. Lefevre, D. D. Damian, E. Iwase, S. Miyashita, *IEEE Robot. Autom. Lett.* **2020**, *5*, 2015.
- [157] R. D. Field, P. N. Anadakumaran, S. K. Sia, *Appl. Phys. Rev.* **2019**, *6*, 041305.
- [158] M. Sitti, *Mobile Microrobotics*, The MIT Press, Cambridge, MA **2017**.
- [159] M. Fernández-Medina, M. A. Ramos-Docampo, O. Hovorka, V. Salgueirido, B. Städler, *Adv. Funct. Mater.* **2020**, *30*, 1908283.
- [160] M. Sun, W. Chen, X. Fan, C. Tian, L. Sun, H. Xie, *Appl. Mater. Today* **2020**, *20*, 100682.
- [161] M. Sun, X. Fan, X. Meng, J. Song, W. Chen, L. Sun, H. Xie, *Nanoscale* **2019**, *11*, 18382.
- [162] H. Ceylan, J. Giltinan, K. Kozielski, M. Sitti, *Lab Chip* **2017**, *17*, 1705.
- [163] Z. Liu, J. Liu, X. Cui, X. Wang, L. Zhang, P. Tang, *Front. Chem.* **2020**, *8*, 124.
- [164] D. Son, H. Gilbert, M. Sitti, *Soft Robot.* **2020**, *7*, 10.
- [165] M. C. Hoang, V. H. Le, K. T. Nguyen, D. van Nguyen, J. Kim, E. Choi, S. Bang, B. Kang, J.-O. Park, C.-S. Kim, *Micromachines* **2020**, *11*, 98.
- [166] L. O. Mair, S. Chowdhury, G. A. Paredes-Juarez, M. Guix, C. Bi, B. Johnson, B. W. English, S. Jafari, J. Baker-McKee, J. Watson-Daniels, O. Hale, P. Stepanov, D. Sun, Z. Baker, C. Ropp, S. B. Raval, D. R. Arifin, J. W. M. Bulte, I. N. Weinberg, B. A. Evans, D. J. Cappelleri, *Micromachines* **2019**, *10*, 230.
- [167] S. Lee, J.-Y. Kim, J. Kim, A. K. Hoshiar, J. Park, S. Lee, J. Kim, S. Pané, B. J. Nelson, H. Choi, *Adv. Healthc. Mater.* **2020**, *9*, 1901697.
- [168] H. Ceylan, I. C. Yasa, O. Yasa, A. F. Tabak, J. Giltinan, M. Sitti, *ACS Nano* **2019**, *13*, 3353.
- [169] W. Chen, M. Sun, X. Fan, H. Xie, *Appl. Mater. Today* **2020**, *19*, 100583.
- [170] S. Lee, S. Lee, S. Kim, C.-H. Yoon, H.-J. Park, J.-Y. Kim, H. Choi, *Sci. Rep.* **2018**, *8*, 3691.
- [171] J. Leclerc, H. Zhao, D. Bao, A. T. Becker, *IEEE Trans. Robotics* **2020**, *36*, 975.
- [172] A. du Plessis d'Argentre, S. Perry, Y. Iwata, H. Iwasaki, E. Iwase, A. Fabozzo, I. Will, D. Rus, D. D. Damian, S. Miyashita, in *Proc. of 2018 IEEE Int. Conf. on Robotics and Automation*, ICRA, Brisbane, Australia **2018**, pp. 1511–1518.
- [173] S. Koseki, K. Kawamura, F. Inoue, K. Ikuta, M. Ikeuchi, in *Proc. of 20th Int. Conf. on Solid-State Sensors, Actuators and Microsystems and Eurosensors XXXIII, TRANSDUCERS 2019 and EUROSENSORS XXXIII*, art. no. 8808545, IEEE, Berlin, Germany **2019**, pp. 2217–2220.
- [174] W. Jing, S. Chowdhury, M. Guix, J. Wang, Z. An, B. V. Johnson, D. J. Cappelleri, *IEEE Trans. Autom. Sci. Eng.* **2019**, *16*, 518.
- [175] L. Yang, Y. Zhang, Q. Wang, L. Zhang, *IEEE Trans. Biomed. Eng.* **2020**, *67*, 1517.
- [176] H. Xie, M. Sun, X. Fan, Z. Lin, W. Chen, L. Wang, L. Dong, Q. He, *Sci. Robot.* **2019**, *4*, eaav8006.
- [177] X. Dong, M. Sitti, *Int. J. Robot. Res.* **2020**, *39*, 617.
- [178] B. Yigit, Y. Alapan, M. Sitti, *Adv. Sci.* **2019**, *6*, 1801837.
- [179] I. Bahiuddin, S. A. Mazlan, M. I. Shapai, F. Imaduddin, S.-B. Choi, *Appl. Soft Comput. J.* **2019**, *76*, 615.
- [180] S. J. Kim, G. H. Jang, S. M. Jeon, J. K. Nam, *J. Appl. Phys.* **2015**, *117*, 17A703.
- [181] H. Choi, S. Jeong, C. Lee, G. Go, J. Zhen, S. Y. Ko, J.-O. Park, S. Park, *Proc. Inst. Mech. Eng. C* **2015**, *229*, 2443.
- [182] G. Park, H.-J. Chung, K. Kim, S. A. Lim, J. Kim, Y.-S. Kim, Y. Liu, W.-H. Yeo, R.-H. Kim, S. S. Kim, J.-S. Kim, Y. H. Jung, T.-I. Kim, C. Yee, J. A. Rogers, K.-M. Lee, *Adv. Healthcare Mater.* **2014**, *3*, 515.
- [183] K. T. Rim, K. H. Koo, J. S. Park, *Saf. Health Work* **2013**, *4*, 12.
- [184] M. Prasad, M. Manoj-Kumar, S. Gowri-Sankar, N. Chaitanya, G. Vivek-Reddy, N. Venkatesh, *J. Clin. Exp. Dent.* **2016**, *8*, e164.
- [185] V. M. Kadiri, C. Bussi, A. W. Holle, K. Son, H. Kwon, G. Schultz, M. G. Gutierrez, P. Fischer, *Adv. Mater.* **2020**, *32*, 2001114.
- [186] F. Leong, N. Garbin, C. D. Natali, A. Mohammadi, D. Thiruchelvam, D. Oetomo, P. Valdastri, *IEEE Rev. Biomed. Eng.* **2016**, *9*, 66.
- [187] M. Runciman, A. Darzi, G. P. Mylonas, *Soft Robot.* **2019**, *6*, 423.
- [188] C. Heunis, J. Sikorski, S. Misra, *IEEE Robot. Autom. Mag.* **2017**, *25*, 71.
- [189] J. Hwang, J.-Y. Kim, H. Choi, *Intell. Ser. Robot.* **2020**, *13*, 1.
- [190] C. M. Heunis, Y. P. Wotte, J. Sikorski, G. P. Furtado, S. Misra, *IEEE Robot. Autom. Lett.* **2020**, *5*, 704.
- [191] S. L. Charreyron, B. Zeydan, B. J. Nelson, in *Proc. of IEEE Int. Conf. on Robotics and Automation*, IEEE, Marina Bay, Singapore **2017**, pp. 4843–4848.
- [192] S. Jeon, A. K. Hoshiar, K. Kim, S. Lee, E. Kim, S. Lee, J.-Y. Kim, B. J. Nelson, H.-J. Cha, B.-J. Yi, H. Choi, *Soft Robot.* **2019**, *6*, 54.

- [193] P. Lloyd, A. K. Hoshiar, T. Da Veiga, A. Attanasio, N. Marahrens, J. H. Chandler, P. Valdastri, *IEEE Robot. Autom. Lett.* **2020**, 5, 3937.
- [194] S. L. Charreyron, E. Gabbi, Q. Boehler, M. Becker, B. J. Nelson, *IEEE Robot. Autom. Lett.* **2019**, 4, 284.
- [195] I. N. Haskins, A. T. Strong, M. T. Allermang, K. P. Bencsath, J. H. Rodriguez, M. D. Kroh, *Surg. Endosc.* **2018**, 32, 895.
- [196] H. Zhu, Y. Shang, T. Ma, Y. Wang, R. Wu, Y. Lv, D. Dong, *J. Surg. Res.* **2019**, 239, 166.
- [197] M. Brancadoro, S. Tognarelli, G. Ciuti, A. A. Menciassi, *Minim. Invasive Ther. Allied Technol.* **2017**, 26, 7.
- [198] R. L. Steinberg, B. A. Johnson, J. A. Cadeddu, *J. Robot. Surg.* **2019**, 13, 599.
- [199] Y. Shang, H. Guo, D. Zhang, F. Xue, X. Yan, A. Shi, D. Dong, S. Wang, F. Ma, H. Wang, J. Li, X. Liu, R. Luo, R. Wu, Y. Lv, *Surg. Endosc.* **2017**, 31, 274.
- [200] T. Cheng, C. S. H. Ng, P. W. Y. Chiu, Z. Li, in *Proc. of IEEE Int. Conf. on Intelligent Robots and Systems*, IEEE, Taichung, Taiwan **2017**, pp. 2902–2908.
- [201] T. Cheng, W. Li, C. S. H. Ng, P. W. Y. Chiu, Z. Li, *IEEE Robot. Automation Lett.* **2019**, 4, 3098.
- [202] W. Li, T. Cheng, M. Ye, C. S. H. Ng, P. W. Y. Chiu, Z. Li, *IEEE/ASME Trans. Mechatron.* **2020**, 25, 1531.
- [203] A. Cavallo, M. Brancadoro, S. Tognarelli, A. A. Menciassi, *Soft Robot.* **2019**, 6, 161.
- [204] M. W. Gifari, H. Naghibi, S. Stramigioli, M. Abayazid, *Int. J. Med. Robot. Comput. Assisted Surg.* **2019**, 15, e2010.
- [205] C. Chautems, A. Tonazzini, Q. Boehler, S. H. Jeong, D. Floreano, B. J. Nelson, *Adv. Intell. Syst.* **2020**, 2, 1900086.
- [206] C. M. Boutry, L. Beker, Y. Kaizawa, C. Vassos, H. Tran, A. C. Hinckley, R. Pfattner, S. Niu, J. Li, J. Claverie, Z. Wang, J. Chang, P. M. Fox, Z. Bao, *Nat. Biomed. Eng.* **2019**, 3, 47.
- [207] K. Wang, Z. Lou, L. Wang, L. Zhao, S. Zhao, D. Wang, W. Han, K. Jiang, G. Shen, *ACS Nano* **2019**, 13, 9139.
- [208] L. Wang, Z. Lou, K. Wang, S. Zhao, P. Yum, W. Wei, D. Wang, W. Han, K. Jiang, G. Shen, *Research* **2020**, 8716847.
- [209] L. Wang, S. Chen, W. Li, K. Wang, Z. Lou, G. Shen, *Adv. Mater.* **2019**, 31, 1804538.



Hyun-Joong Chung is an associate professor of Chemical and Materials Engineering at the University of Alberta. He received B.S. from KAIST and Ph.D. from the University of Pennsylvania, where he studied the role of jamming nanoparticles in phase-separating polymer blends. After graduation, he worked on oxide semiconductors in Samsung Display, followed by postdoctoral training on stretchable bioelectronics at the University of Illinois at Urbana-Champaign. Currently, he leads an interdisciplinary research program on understanding physicochemical properties of gels and elastomers with or without functional additives and reinforcements, as well as on translating the fundamental understanding to biomedical device applications.



Andrew M. Parsons is an undergraduate student in Materials Engineering at the University of Alberta. In the summer of 2020, he joined Dr. Chung's research group (through an NSERC USRA research grant) because of an interest in polymer science and its biomedical applications. He expects to receive his B.Sc. at the end of winter term 2021.



Lelin Zheng is currently in after-degree program in the Faculty of Education at the University of Alberta. She received her M.Sc. from University of Alberta in 2019, with her thesis focused on colloids and gels containing magnetic nanoparticles. She completed her undergraduate studies in Sun Yat-sen University, China, where she did her capstone project on magnetic nanoparticles.