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Soft-Matter Engineering for Soft Robotics

Carmel Majidi

Since its inception, the field of robotics has aimed to create machines that mimic the extraordinary capabilities of the human body. From as early as the 1940s, this has included efforts to engineer actuators and electronics out of elastomers, textiles, and other soft materials in order to mimic the compliance and deformability of natural biological tissue. In the decades since, there is extraordinary progress in the subdomain of soft robotics, with recent efforts focused on novel methods of actuation, sensing, and manufacturing. In this progress report, recent advancements within this field from the perspective of materials and mechanics are highlighted. Wherever possible, efforts in soft robotics are connected to progress in the broader field of soft-matter engineering, which relates to the application of principles and practices in the soft-matter sciences to create machines, electronics, and robotic systems out of fluids, elastomers, gels, and other soft materials. To close, the current challenges and future opportunities within the field of robotics are briefly discussed, with special attention toward the eventual goal of autonomous soft robots that are capable of operating without dependency on external hardware, tethers, or manual intervention.

1. Introduction

The field of robotics as a scientific discipline emerged in the first half of the 20th century as an effort to create fully autonomous machines that are able to independently move and interact with their environment and adapt their physical operation in response to changing conditions. In the decades since, this has led to extraordinary advancements in industrial automation, surgical robots, and autonomous vehicles with machines constructed from hydraulics, motors, and electronics for sensing, computing, and vision. Although engineered with materials that have vastly different properties than biological tissue, these robotic systems are capable of matching, and in many cases exceeding, the performance of natural organisms for prescribed tasks in manufacturing, precision manipulation, and transportation. While there are still opportunities for improved mechanical and computing hardware within these domains, much of the current research in robotics have focused on advanced algorithms for sensing, vision, data fusion, decision making, and data-driven learning. In this respect, existing materials and hardware technologies are good enough and do not represent a crucial bottleneck for further progress within current application domains.

So why the recent interest in soft robotics and the sudden need to create machines and electronics using elastomers, fluids, and other soft matter? After all, rigid materials have a variety of properties that make them well-suited for actuation, sensing, signal processing, and robot housing/packaging. In particular, the stiff plastics, composites, metals, and ceramics used for structural reinforcement, packaging, motors, and printed circuit boards (PCBs) exhibit the following advantages: (i) highly load bearing and can support large mechanical work or large inertial forces; (ii) can maintain fixed mechanical or electrical properties under extreme forces; (iii) enable precision positioning and motion control by rigidly transmitting displacements. So why, then, replace these with materials that have poor load capacity and for which it is difficult to precisely control their motion and geometry?

The reason to engineer robots out of soft matter arises from another fundamental aim within the field of robotics—creating universal and customizable machines that are capable of performing a wide variety of tasks and actively adapting themselves to changing conditions within these tasks. While industrial arms, surgical robots, and autonomous vehicles can perform prescribed tasks with extraordinary precision, speed, or reliability, they are not capable of adapting to other tasks (e.g., from precision manipulation to heavy-duty parts assembly) or operating in fully unstructured environments (e.g., from a flat paved road to a rocky mountainside). In contrast, natural organisms present a proof-by-existence that it is possible to create machines that are not only high performing, but also universal and customizable. Within the field of soft robotics, cephalopods like the octopus are an especially popular example due to its astonishing ability to move in tightly confined spaces, manipulate objects, and camouflage itself through changes in shape, color, and surface texture.[1] While there are many factors that contribute to the rich versatility and multifunctionality of natural organisms, the use of soft materials has a central role in their ability to change their shape or how load is distributed so that they can adapt to new tasks or changing environmental conditions. More broadly, soft robots could help span the gap between the high performance but specialized functionality of conventional machines with the remarkable versatility of biological organisms (**Figure 1**A).

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In this progress report, the author will review some of the recent advancements in soft robotics and describe some of the open challenges that remain. As necessary, the author will relate efforts within this subdomain to the emerging field of soft-matter engineering. Soft-matter engineering represents the practice of creating machines, electronics, and multifunctional structures primarily out of soft materials and fluids using principles and practices in polymer chemistry, condensed soft-matter physics, soft lithography, and soft microfluidics (Figure 1B). In this respect, many of the material technologies used to create the artificial muscle, nervous tissue, and skin used for soft robots fall within the domain of soft-matter engineering since they are largely composed of elastomers, fluids, and gels. In this report, the author will dedicate particular focus on efforts toward autonomous soft robots that do not rely on manual intervention or tethered hardware. Untethered soft robots, unleashed from the constraints of bulky external hardware, represent a necessary step toward the ultimate goal of fully autonomous machines capable of independent mobility and physical interaction with the environment.[4]

Before reviewing progress in untethered soft robots, the author will begin by presenting a working definition of what a robot is (Section 1.1). This will be helpful in evaluating current trends and guiding future efforts within the subdomain of soft robotics. Next, the author will highlight some of the features of natural organisms, in particular soft organisms and biological tissue, that are not present in conventional robotic systems and hardware (Section 1.2). The purpose of this is to motivate the need to revisit the materials and hardware architectures used in robotics. Finally, the author will define what is meant by *soft* and explain how the use of soft materials could enable machines to become more universal and customizable. The remainder of the progress report will explore current practices in soft robotics and relate these to the ultimate goal of untethered, autonomous soft robots that can match the robust mechanical properties, versatility, multifunctionality, and adaptability of natural organisms. This will begin with a discussion of relevant materials (Section 2), including elastomers (2.1), polymer composites (2.2), fluids (2.3), gels (2.4), and liquid metal (LM) embedded elastomer (LMEE) composites (2.5). Next, the author will review various material architectures for artificial muscle actuation (Section 3), with special focus on fluidic actuators (3.1), dielectric elastomers (DE) (3.2), and thermally responsive materials (3.3). This is followed by an overview of current efforts in systems-level soft robotic implementations as well as a brief discussion on remaining challenges and opportunities for future research (Section 4).

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1.1. Robots: A Working Definition

Definitions of the term *robot* vary widely and have been used to describe a broad range of systems, from complex anthropomorphic machines like the Honda Asimo and Boston Dynamics Atlas to air-filled rubber structures that move or grip objects when pneumatically actuated. For the purposes of this progress report, the author will adopt the following definition of a

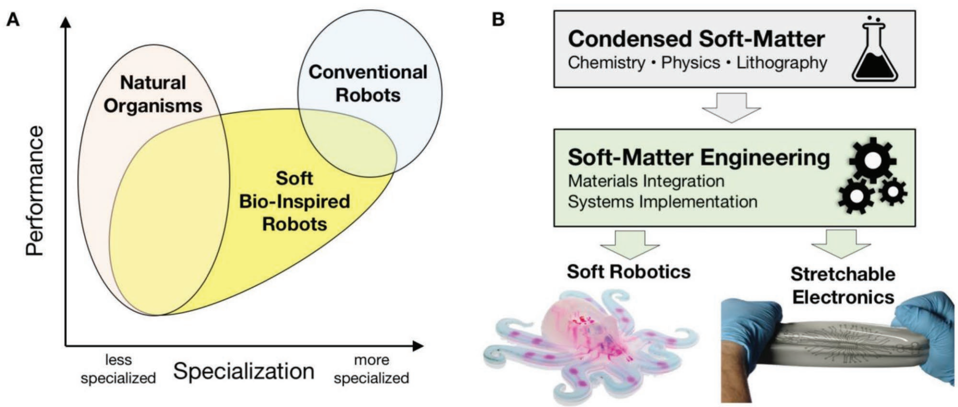

Figure 1. A) Soft bioinspired robots have the potential to bridge the gap between conventional machines and natural organisms in balancing performance with task specialization. B) Soft-matter engineering represents the applications of condensed soft-matter physics, polymer chemistry, and lithography to applications in soft robotics (reproduced with permission.[2] Copyright 2016, Nature Publishing Group) and stretchable electronics (reproduced with permission.[3] Copyright 2018, Springer Nature).

robot adapted from Peter Corke in his book *Robotics, Vision and Control*:[5] a robot is a machine capable of

Sensing—gather information from its environment or about its own physical state;

Planning—use this information to generate decisions on how to change its state or act on its environment;

Action—perform a physical task based on its decision.

While these conditions are necessary, they are not sufficient, for example, many household appliances now have embedded sensors and CPUs that enable the above capabilities, but few would regard these as robots. It should be noted that many would add programmability to the set of conditions. The author has left this out since the notion of machine programmability can be vague. While the concept of digital programmability, that is, coded script with commands or rules uploaded to a microcontroller, is unambiguous, it is less clear how to define programmability in the context of materials selection, mechanical design, and structure. For example, a mechanical system can have prescribed motions, stiffnesses, and resonant frequencies/modes that can be programmed with reconfigurable linkages, cams, or weights. Likewise, a microstructured material can be patterned and actively reconfigured to program certain elastic, tribological, or photonic properties.

Although the definition of what makes a machine a robot can be further restricted, it can also be relaxed. Within the soft robotics community, there are many examples of robots that are not capable of sensing, decision making, or varying their physical motion. Most of these robots use open-loop control and are often limited to a single degree of freedom or single repetitive motion. While it may be reasonable to think of these as robots, there is a danger in broadening the definition too widely to the point that it has not scientific meaning, that is, "if everything is a robot, then nothing is a robot." On the other hand, creating restrictive definitions within a continuously evolving field may create unnecessary, artificial barriers that could cut out meaningful innovation. Therefore, rather than judging whether a machine should or should not be called a robot, it may be more meaningful to ask what scientific lesson is learned from a given implementation and whether it can be used to inform future practices in robotics.

1.2. Why Bioinspired?

Existing robotic systems can be engineered to match or outperform natural organisms for specialized mobility and manipulation tasks. However, there is no single machine that comes close to matching the versatility of animals in their ability to adequately perform a wide range of tasks. This dichotomy has led to the emergence of *bionics* to create humanoid robots, robotic prostheses, artificial muscle, electronic skin, soft multifunctional materials, and other technologies inspired by natural organisms and biological tissue. Such efforts in bioinspired design are based on the recognition that nature has solutions to problems that we are still struggling to solve using existing engineering methods. Indeed, nature has been a rich source of inspiration to create robotic systems or materials with a combination of properties not previously possible in engineered systems. A popular example has been the gecko lizard, which has inspired wall-climbing robots like the Stickybot[6] and the development of dry adhesives that mimic the setal arrays of Anolis lizards in their ability to exhibit shear-controlled adhesion[7] (**Figure 2**A).

Bioinspired engineering provides one potential roadmap, but by no means represents the only approach for creating robotic systems that are universal and customizable. Nonetheless, while alternative design paradigms may eventually emerge, they will likely incorporate many of the same principles observed in natural systems:

(i) Choice of material and material geometry matter—the stiffness and dimensions of a material will determine its load capacity, deformation, and how stress is internally distributed (Figure 2B);
(ii) Integrating materials with different stiffnesses can allow systems to adapt their shape and internal stress distribution to support a wide range of loading conditions and geometric constraints;
(iii) Hierarchical gradations between soft and stiff materials can help with translating the effects of physics at the nanoscale to mechanics and motion at the mesoscale. An example of this is the human musculoskeletal system, which uses a combination of muscle fiber, tendons, and bone to translate chemically driven interactions between actin and myosin filaments into human motor tasks (Figure 2C).

In all these cases, the ability to incorporate soft and elastic materials into the robotic system is crucial. Without these, the system will not be capable of adapting its shape, stiffness, or internal load distribution for accomplishing new tasks or to accommodate changes in environmental conditions.

Of course, it is natural to question why it would be better to have one universal machine capable of all tasks rather than having a collection of specialized machines dedicated to each task. After all, modern households are full of appliances that cover a wide range of tasks and it is not obvious why it would be useful to replace these with a single device. However, the conditions within a household are largely known and static. In contrast, applications of robotics to field exploration, healthcare, and human–machine interaction typically require physical interaction in unknown or changing environments. Moreover, as the demands for robotic automation and machine assistance continue to increase, these systems will be exposed to a wider range of scenarios where it is not always known a priori what the robot is supposed to do or how it is meant to respond. An example could be a healthcare robot that is capable of giving immediate aid to a patient who experienced an unexpected fall during a routine activity. With a collection of specialized machines, the patient might not get the necessary aid because only a certain number of preselected devices might be on hand at any given time.

Another context in which it may make sense to replace a collection of specialized machines with a single bioinspired robot is in resource- or space-limited environments. These include the International Space Station (ISS), sea vessels, aircraft, and remote field operations. In the case of ISS, the need to cut down on collections of specialized tools, equipment, and machines has led to interests in 3D printers and humanoid robots like the Robonaut.[8] Similarly, the military has long expressed interest in programmable matter in which a single volume of material can reconfigure itself into various tools, for example, a clay-like

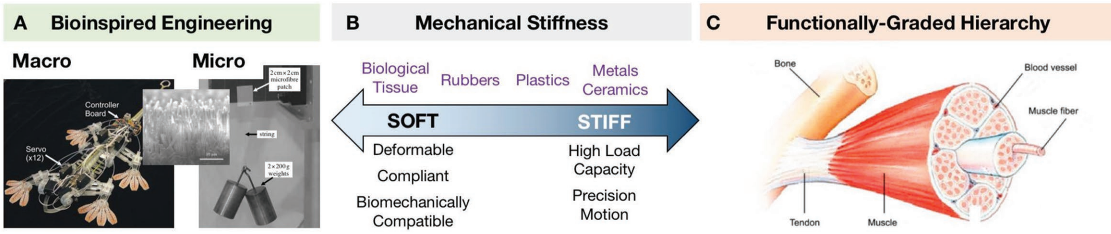

Figure 2. A) Bioinspired engineering has led to advancements at multiple length scales, exemplified by the gecko-inspired Stickybot (reproduced with permission.[6] Copyright 2006, Company of Biologists) and microfibrillar adhesive engineered to mimic the setae in the Anolis lizard. Reproduced with permission.[7] Copyright 2008, The Royal Society Publishing. B) Material stiffness matters in determining how a system deforms and supports load. C) The musculoskeletal system represents a classical example of how robust mechanical functionality can be achieved by combining soft and rigid materials at a variety of length scales (Credit: National Cancer Institute/SEER Training Modules).

material that can reversibly transform itself from a screwdriver to a pair of pliers. Even in the household, there is a proliferation of appliances and Internet of Things (IoT) technologies designed to further improve quality of life through automation. This could eventually result in the need for multitasking humanoids or healthcare robots that would replace these specialized technologies.

1.3. Why Soft?

To date, much of the effort in bioinspired robotics have focused on piecewise-rigid systems composed of motors, PCBs, and semirigid plastics, carbon-fiber composites, or cardboard. These robots are able to walk, crawl, fly, and swim in a manner similar to natural organisms without reliance on soft or highly deformable materials. Given the success of bioinspired robotics with rigid and semirigid materials, why bother to create soft robots?

Despite promising achievements in the field of bioinspired robotics, there are still several critical issues that can only be addressed by using soft materials:

- Mobility in confined spaces—Semirigid robots can have adequate compliance to collapse their shape through folding or bending. However, they are limited in their ability to squeeze and move through tightly confined spaces. In contrast, soft robots can passively deform in order to adapt to the shape of their surroundings.[9,10]
- Impact resistance—When subject to large loads, stiffer materials tend to generate high stress concentrations that can lead to permanent mechanical damage. In contrast, soft materials use their compliance and deformability to distribute loads over a large area, allowing for uniform stress distribution and low peak stress. Likewise, soft materials are better at absorbing dynamical loads and shock.[11,12]
- Design complexity—In order to be mechanically compliant at the device level, piecewise rigid systems require carefully selected geometries. An example is the Softworm peristaltic robot, which achieves compliance using a helical mesh structure for its body.[13] However, these so-called deterministic compliant architectures can introduce significant complexity or limitations to the design. Because soft robots are made of intrinsically compliant materials, such deterministic architectures do not need to be incorporated into their design.

While the last item is not a fundamental technical barrier, it nonetheless represents an important reason to adopt soft materials into robot design. This is because manufacturing represents a significant bottleneck for creating bioinspired robots and so anything that can simplify design will relieve the burden on having to adopt prohibitively complicated fabrication methods.

Another reason to use soft materials is that they are more compatible with the mechanics of the human body. Elastomers and stretchable textiles can be worn on the skin without causing discomfort or interfering with natural motion. When used as an electronic skin for machines used in human–robot interaction, the material can conform to the body and not result in painful stress concentrations or point contacts. Such compatibility is especially important for wearable assistive robots, orthoses, and even implants. An example of the latter is a soft robotic ventricular assist device for help with pumping the heart.[14]

For the purposes of this progress report, *soft* is defined as any fluid, gel, or material with a shear modulus less than 10 MPa. This includes most elastomers along with soft polymer composites that contain a dispersion of nano- or microscale particles. Although not a significant focus of this report, biological materials used in tissue engineering and biohybrid materials also largely fall within the definition here of soft matter.

2. Materials

The soft robotic systems covered in this report are largely composed of soft polymers and elastomers-like silicone rubber. The elastomers are typically used for the limbs or fingers of the robot as well as its housing or carrier medium for embedded electrical circuitry. Soft robots also typically contain fluids for actuation or stretchable circuitry. These fluids can be filled into chambers or incorporated as microfluidic channels or dispersion within an elastomer matrix or gel. In the case of fluidic actuation, the hydrostatic pressure of the fluid also has a central role in the mechanics of the soft. Although relatively small compared to the maximum stresses in the surrounding elastomer, they must nonetheless be included in the potential energy used to determine the mechanical response of the robot. For certain dynamical loading conditions, fluid rheology can also have a role. However, most applications involve low shear rates such that the fluidic shear stresses are negligible compared to the hydrostatic pressure or elastic stresses. Finally, soft robots may contain smart materials like shape-memory alloy (SMA) and liquid crystal elastomers (LCEs), which change shape in response to electrical current or heat. An example of a recent soft robot gripper that contains all three of these material classes—silicone for housing, liquid metal for circuit wiring, and SMA for the actuators—is presented in **Figure 3**.[15] It is significant to recognize here that, as with many robotic implementations, the gripper also contains conventional microelectronics for sensing, signal processing, and radio communication.

For the various materials used in soft robots, it is useful to review the constitutive relationships that relate applied stresses with internal pressure and deformation. In the case of conductive elastomers, it also helps to have a sense of the electromechanical coupling between stretch and electrical resistance. These relationships are then used in Section 3 to better understand the principles of actuation and how material properties and geometric dimensions influence their functionality.

2.1. Elastomers

Elastomers are rubbery polymers that are mechanically compliant and have a high elastic strain limit. Compliance is commonly related to Young's modulus (E), which scales with the tensile stress required to stretch a material by a prescribed amount. In conventional engineering applications, strain is typically small and E can be determined for small deformations in the regime where stress and strain have an approximately linear relationship. However, elastomers and other soft polymers used in soft robotics typically undergo large strains. In this regime, their the stress response is typically nonlinear. More generally, additional elastic coefficients are required to capture the stress–strain behavior of elastomers. Nonetheless, Young's modulus is still a useful measure for comparing the stiffness of materials since the full nonlinear response will converge with the linearized relationship at small strain. In general, elastomers have a modulus between 0.1 and 10 MPa, although soft robots tend to be composed of soft polysiloxanes (silicone) and polyacrylate elastomers, which typically have a modulus between 0.1 and 1 MPa.

A material is elastic if it returns to its original length after it has been stretched under an applied tensile load. Elastomers are considered to be hyperelastic since they exhibit an elastic response over a broad range of strains and have a stress–strain relationship that can be derived from a strain energy density W.[16] Typically, W is expressed in terms of a stretches λ_i where the index $i \in \{1,2,3\}$ corresponds to the orthonormal directions associated with the principle directions of elastic deformation. For a volumetric element with edges oriented along the principle directions, stretch is defined as the final length of the edge (l_i) divided by its initial length (l_0). Likewise, (Cauchy) stress σ_i is defined as the internal pressure (in Pascals) acting along the corresponding principle direction. For a hyperelastic solid that is incompressible, that is, volume remains fixed; $\lambda_1\lambda_2\lambda_3 = 1$, the stress is calculated as $\sigma_i = \lambda_i\{\partial W/\partial\lambda_i\} - p$.[17] The value p is called the hydrostatic pressure and is typically unknown when first deriving the constitutive relationship. It can be determined from the boundary conditions for σ_i or λ_i along with the incompressibility constraint.

The elastic strain energy density W can be derived from the Helmholtz free energy of a solid. For most soft elastomers, it is dominated by entropy of the polymer chains and can be obtained from first principles using statistical mechanics. This includes the commonly used Neo-Hookean constitutive model: $W = C_1\{\lambda_1^2 + \lambda_2^2 + \lambda_3^2 - 3\}$, where the elasticity coefficient $C_1 = E/6$. The form of the strain energy function can also be obtained from experimental measurements. One such phenomenological representation of a hyperelastic solid is the Ogden model.[17]

In soft robotics, there are a wide range of elastomers that have become popular for their compliance, stretchability, and elastic resilience. Polysiloxanes such as poly(di-methylsiloxane) (PDMS) are commonly used for soft microfluidics and robotics due to their low modulus, high strain limit, and relatively low hysteresis between loading and unloading cycles. Pt-cured PDMS elastomers like Ecoflex 30 (Smooth-On) and Sylgard 184 (Dow Corning) are especially popular because they are clear, inert, and mechanically robust. There has also been interest in soft polyurethanes, polyacrylates (e.g., 3M VHB), and block copolymer elastomers like styrene ethylene butylene styrene. When selecting an elastomer, engineers typically focus on properties like strain limit, modulus, and manufacturing processability. However, it is also important to consider factors like creep, stress relaxation, and other forms of inelastic deformation that can lead to hysteresis and energy loss during consecutive loading and unloading. A more complete examination of elastomers for various robotics and stretchable electronics applications have been reported in recent literature.[18–20]

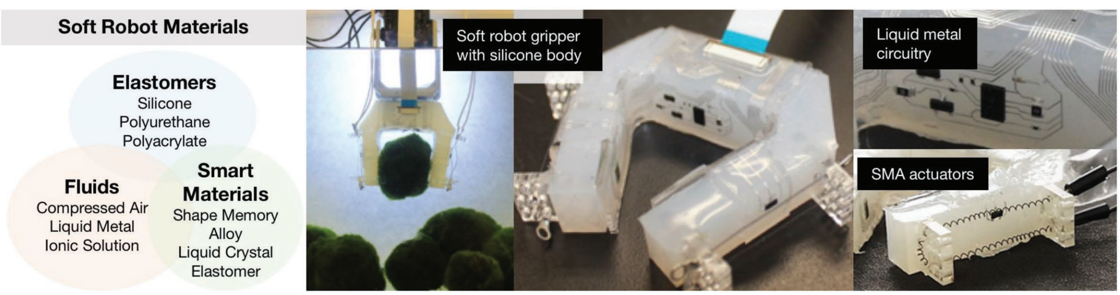

Figure 3. Soft robots are typically composed of elastomers, fluids, and smart materials that respond to electrical or thermal stimulation. An example of this is a soft robot gripper that has a silicone elastomer body, liquid metal circuitry to support embedded microelectronic components, and shape memory alloy for electrical actuation. Reproduced with permission.[15] Copyright 2018, Institute of Electrical and Electronics Engineers.

2.2. Polymer Composites

Elastomers are naturally insulating—both electrically and thermally. In order to introduce electrical and/or thermal conductivity, elastomers are typically filled with nano/microparticles or fluidic inclusions. The dispersion phase is typically conductive, resulting in a substantial enhancement in thermal conductivity and electric permittivity. For high filler concentrations, the dispersion phase forms a percolating network with long-range connectivity. This results in a composite that is electrically conductive. Conductive elastomers typically contain percolating networks of acetylene carbon black, exfoliated graphite, carbon nanotubes, silver nanoflakes, silver nanowires, and other or metal nano/microparticles. An example of a soft sensing skin with traces of carbon-filled conductive elastomer is shown in **Figure** 4A.[21] Electrically conductive elastomers can also be produced by blending elastomers with conductive polymers like polyaniline (PANI) or poly(3,4-ethylenedioxythiophene) polystyrene sulfonate (PEDOT:PSS).[22]

Conductive elastomer composites typically exhibit significant electromechanical coupling and hysteresis, that is, greater electrical resistance during unloading compared to loading. While several theories exist for explaining electromechanical coupling in conductive elastomer composites, one possible explanation is related to electrical tunneling between conductive particles. Rather than forming direct physical contact, nanoparticles in a percolating network are separated from their neighbors by an Å-scale gap, with the contact resistance is controlled by electrical tunneling. As the gap increases during stretch, the interparticle resistance increases dramatically, resulting in a decrease in the effective volumetric conductivity of the composite.[23]

2.3. Fluids

The earliest soft robots used pneumatic artificial muscles (PAMs) for actuation. These actuators are composed of an elastomer shell lined with braided textile that would contract when filled with compressed air. They are called artificial muscle due to the similarities with natural muscle in force output, contraction, stiffness change, and speed.[24] Another early example of soft biocompatible technology was the Whitney strain gauge, which was composed of a rubber tube filled with LM.[25] Strain was determined by measuring the change in the conductivity of the LM channel as the tube was stretched. Because of its compliance and elasticity, the strain gauge could wrap around a human limb and measure muscle contraction or joint motion without interfering with the natural mechanics of the body.

In addition to compressed air and LM, there have been a variety of other fluids commonly used in soft robotics over the subsequent decades. These include carbon grease, water, aqueous electrolytic solutions, and O_2 gas produced through fuel decomposition. The role of fluids in soft actuators is analogous to that in conventional pneumatic and hydraulic machinery. Although performance can be influenced by chemical reactions, rheology, gas dynamics, and other kinetic or rate-dependent properties, this section will focus on static and quasistatic fluid properties.

2.3.1. Compressed Air

The ideal gas law is typically used to model the mechanics of air in PAMs and other soft pneumatic systems. For compressed air with a fixed pressure p, defined relative to atmospheric

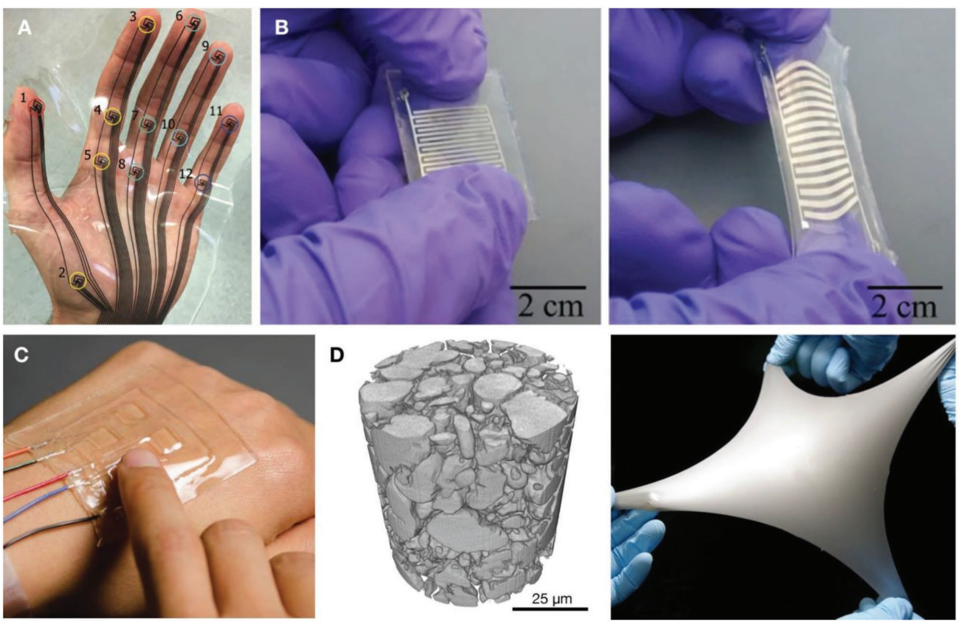

Figure 4. A) Soft sensing skin with traces of carbon-filled conductive elastomer. Reproduced with permission.[21] Copyright 2016, Wiley-VCH. B) Stretchable capacitor with channels of EGaIn liquid metal in a soft silicone film. Reproduced with permission.[20] Copyright 2018, American Chemical Society. C) Tactile skin with ionically conductive gel. Reproduced with permission.[40] Copyright 2014, Wiley-VCH. D) Liquid metal embedded elastomer (LMEE) composite, which exhibits high electrical permittivity and thermal conductivity. Reproduced with permission.[43] Copyright 2016, Wiley-VCH; reproduced with permission.[44] Copyright 2017, National Academy of Sciences.

pressure, the potential energy is $U_p = -pV$, where V is the enclosed volume. To maintain this pressure as the volume changes, air must flow in or out of the enclosure. When the enclosure is sealed, with the air trapped inside, the ratio p/V becomes fixed and the potential energy is computed as $U_p = p_0V_0\ln(V/V_0)$. Here, the subscript 0 denotes the initial value prior to volume change. Gas pressure is typically controlled with electrically powered pumps or valves that are connected to a compressor or pressurized supply.

Recently, there has been interest in using pressurized air created through combustion and fuel decomposition in order to power soft pneumatic actuators. One approach has been to use platinum to catalyze the decomposition of hydrogen peroxide[2,26] or the combustion of butane and oxygen.[11] Such reactions can lead to significant gas pressures (e.g., 50 kPa for H_2O_2 decomposition) that are sufficient for actuator motion. Robots powered with combustion are capable of jumping and require functionally graded materials with gradations in mechanical compliance in order to distribute internal stresses during impact landings.

2.3.2. Water

Soft fluidic actuators are also powered with water instead of air, as water can allow for greater load bearing and a more rapid actuator response.[27] As with the surrounding elastomer, the water is treated as incompressible and so slight displacements to the boundaries of the fluid or surrounding elastomer will typically lead to rapid changes in fluidic pressure and actuator stiffness. There has also been recent interest in underwater soft actuators with hydrogels that allow water to flow out into the surrounding fluidic media.[28] Because the hydrogel has similar density and transparency to seawater, the soft actuators are both optically and acoustically transparent. This could have potential advantages for engineering underwater soft robots that do not interfere with the natural behavior of surrounding marine organisms.

2.3.3. Conductive Fluid

Electrically conductive fluids are commonly used for soft robotic skin and DE technologies. Conductive grease, such as carbon black mixed with silicone oil, has been especially popular for dielectric elastomer actuator (DEAs) and other DE-based technologies.[29] Despite their relatively high electrical resistivity ($\rho \approx 0.1$–$1\ \Omega$ m), these greases have adequate conductivity to function as capacitive electrodes for actuators and capacitive sensors. DE films can also be coated with liquid metals[30] like eutectic gallium indium (EGaIn), which are nontoxic and have much lower resistivity ($\rho \approx 3 \times 10^{-7}\ \Omega$ m).[31] They oxidize in air, forming a nm-thick Ga_2O_3 skin that holds the liquid in place and helps it wet to most polymer surfaces, including soft polyacrylate and silicone.[32]

Ga-based LM alloys are also popular as stretchable circuit wiring and transducers for soft microfluidic electronics. An example of EGaIn microchannels in a soft silicone is presented in Figure 4B.[20] As shown in the figure, the fluidic channels remain intact as the surrounding elastomer is stretched. They can be used to map strain or pressure into changes in electrical resistance. In fact, commercial versions of the Whitney strain gauge are presently made with EGaIn rather than mercury. The resistance (R) of LM channels can be predicted with Ohm's law, that is, $R = \rho L/A$, where L and A are the channel length and cross section, respectively. Soft microfluidic electronics have also been engineered using aqueous electrolytic solutions.[33,34] These ionic conductors are nonetheless useful for sensors or electrodes that provide AC field and are not capable of supporting DC current for digital circuit functionality.

2.4. Gels

Gels have been emerging as a popular material for soft robot actuation and electronics. By definition, gels are binary fluid-solid systems that have colloidal dimensions, that is, nanometer or micrometer-scale distances between the boundaries of the fluid and solid phases.[35] Gels can be thought of as combining the material properties of soft polymers and fluids, with the polymer treated as the continuous dispersion medium and the fluid as the dispersion phase. The polymer typically represents only a small volume fraction of ≈1–10%, with just enough material to give the gel structural integrity, and the gel is usually bicontinuous, with both the solid and fluid phases forming connected networks.

Gels can be infused with a variety of different fluids. These can range from water, as in the case of hydrogels,[36] to ionic solution[37] and gas (i.e., aerogels[38]). Gels with ionic solutions have been used to create optically transparent DEAs[39] and tactile skin, as shown in Figure 4C.[40] The solid phase typically exhibits elastic properties that enable swelling and large mechanical deformation. It can be synthetic, for example, polyethylene oxide, polyvinyl alcohol,[36] or naturally derived using agarose, alginate, or collagen. There has also been recent interest in double network gels that exhibit a remarkable combination of high mechanical compliance (tensile modulus ≈ 0.1–1 MPa) and fracture toughness (≈0.1–10 kJ m^{-2}).[41,42] Gels are an attractive material system for applications in soft robotics due to their potential to combine ionic conductivity with high tear resistance and skin-like elasticity.

2.5. LM-Embedded Elastomers

While gels are predominately composed of fluid, it is also possible to engineer fluid-elastomer composites in which the fluid dispersion makes up less than half of the volume. One approach is to embed elastomers with a suspension of Ga-based LM microdroplets (Figure 4D).[43,44] Compared to conductive polymer composites that have rigid filler, these LMEEs exhibit significantly reduced electromechanical coupling.[3,45] Depending on the concentration and microstructure of the embedded LM droplets, the LMEE composites can be thermally conductive and electrically insulating or electrically conductive. In the case of electrically conductive LMEE, the droplets make direct physical contact and the contact resistance has only limited dependency on stretch. This is due to the ability of the liquid metal to flow between the droplets and preserve the contact resistance of each droplet–droplet interface. Because the total number of contacts does not change during stretch, the absolute electrical resistance remains unchanged.

Soft silicones and urethanes have typically been used as the matrix material for LMEEs, although the LM droplets can be suspended in a much wider range of polymers or deposited as a thin-film coating on an elastomer substrate. To create electrically conductive networks, the droplets can be ruptured through mechanical or laser scribing.[46,47] Interestingly, this same scribing process enables new conductive pathways to form in circuits that have been mechanically damaged. This results in an autonomous self-healing property in which the electrical wiring within a soft robot can form new routes to conduct electricity as the robot is punctured with holes.[3]

LMEEs that have not been scribed remain electrically insulating and exhibit an enhanced electrical permittivity.[43] The increased electric permittivity (ε_r) arises from the polarization of the embedded droplets and can be predicted with effective medium theory (EMT), which provides reasonable agreement with experimental measurements.[48] The field equations for heat transfer have the same differential form as those for electrostatics. Therefore, EMT can also be used to predict the relative enhancement in thermal conductivity (k) of an LMEE composite. The prediction can be further modified to account for stretch in the composite, which leads to elongation of the droplets into needle-like ellipses. Such theoretical predictions have been shown to be in good agreement with experimental measurements of LMEEs stretched to 400% strain.[44] In general, thermally conductive elastomer composites can function as heat dissipating elastomer substrates for heat generating electronics or thermal actuators such as shape-memory alloy.

3. Architectures

Over the decades since the inception of robotics, PAMs and LM-based sensing have been used in humanoids,[49] exoskeletons,[50] industrial automation,[51] and clinical physiology.[25] In recent years, there has been increasing interest in extending pneumatic actuation and LM electronics to create miniaturized systems using emerging methods in soft-matter engineering.[52,53] This includes soft lithography fabrication methods, soft microfluidic architectures, micro/nanocomposite synthesis, and soft polymer engineering. Soft-matter engineering has also been used to create electrostatic actuators, sensors, and energy-harvesting transducers. In the case of DEAs, an insulating elastic film is coated with conductive liquid (e.g., carbon grease or liquid metal) and its deformation is coupled to the electrical capacitance between the liquid electrodes. While the properties of DE transducers have been known since the late 19th century,[29] recent efforts have focused on soft robotic implementations using polyacrylate films and tape, silicone elastomer, conductive grease, and conductive elastomers.[54]

In contrast to conventional robotic hardware, these soft material systems can be incorporated into a host robot or interfaced with the human body without adding bulk, weight, or stiffness. Perhaps the most successful applications of soft material architectures to date are the inflatables used in balloon angioplasty and blood pressure monitoring. While these are not robotic implementations, they do share many of the same design principles that are popular within soft robotics and may represent a starting point for more widespread adoption of soft robotics into commercial or medical products.

As soft materials become increasingly used in robotics and human–machine interaction, there will be continued need for improvements in the performance and functionality of soft matter circuits, sensors, and actuators. This requires progress in not only materials engineering and device-level design, but also advancements in manufacturing for precision materials patterning and robust multimaterial integration. Moreover, as soft robotic technologies continue to mature, there will be an increasing demand for predictive theoretical models that can inform design and operation. As in other areas of robotics and engineering, these models can enable rapid design decisions with reduced dependency on prototype iterations and testing. Moreover, they can furnish design rules and scaling laws that are particularly useful in early stages of design where it is impractical to explore a vast materials space using experimental or computationally intensive finite element techniques. Where possible, the following subsections will apply the constitutive models presented in Section 2 to derive approximate algebraic expressions that relate device performance with material properties, geometry, and external loading conditions. Such models are not only helpful for making early stage design decisions but also can be used to identify the relevant governing physics that can be subsequently incorporated into a more detailed computational simulation.

This section presents an overview of only a few classes of soft robot actuators and is by no means exhaustive. The categories of actuators reviewed here have been selected because of their current popularity in soft robotic implementations. For a more complete discussion of soft actuators used in robotics, the interested reader should refer to recent review papers in the literature.[55,56]

3.1. Fluid-Elastomer Structures

Much of the current work in fluidic soft robots and electronics build on fabrication methods and architectures for soft microfluidics used in biotechnology and medicine.[57–60] These devices are typically produced using soft lithography fabrication techniques to create networks of microfluidic channels embedded in a soft polymer matrix.[61–63] Recent applications include soft microfluidics for organ-on-a-chip devices,[64,65] soft robotics,[66] and resistive strain gauges and pressure sensing with liquid metal.[67] In some cases, the deformation of the elastic medium and embedded microfluidic channels are coupled and this can be exploited to achieve unique functionalities that are not possible with a rigid device. Examples include soft robot actuators that couple fluidic pressure with the contraction, inflation, or bending of an elastic shell; sensors composed of liquid metal or ionic fluid[34] microchannels that change electrical resistance in response to applied pressure or stretch; pressure-controlled microfluidics that filter or capture nanoparticles;[68,69] and microfluidic "Quake" valves that use pressurized control lines to manipulate the flow of fluid within a device.[62]

Theoretical studies have been performed that establish fluid–elastomer coupling for a wide range of soft microfluidic systems. These include studies of PAM actuators that relate internal fluid pressure (p) with various modes of actuator deformation.[70,71] Examples include the pneumatically actuated gripper in **Figure 5**A,[72] 3D-printed flexural actuator in Figure 5B,[73] and contractile vacuum-controlled actuator in Figure 5C.[74] In general, these relationships are established by finding the shape (i.e., contraction length, bending curvature, or twist angle) that minimizes the total Gibbs free energy (Π) of a system. For PAMs and other pressure-controlled mechanical systems, Π is equivalent to the potential energy of a system and accounts for the stored elastic energy of the material ($W \cdot V_0$, where V_0 is the fixed volume of the rubber), mechanical work of the pressurized fluid ($p \cdot V_e$, in the case of air, where p is fixed, V_e is the enclosed air volume), and mechanical work of the applied load ($F \cdot L$, in the case of length change under a fixed force F, where L is the actuator length). In the case of the McKibben PAM actuator, the stored elastic energy has negligible contribution compared to the mechanical work of the pressurized air and applied force. For this special case, the length and diameter (D) of the actuator are coupled by the inextensibility of the braided nylon that make up the actuator's fabric shell: $D = b\{1 - L^2/b^2\}^{1/2}/n\pi$, where b is the fiber length and n is the number of turns.[70] It follows that $\Pi \approx -p\pi D^2 L^2/4 - \mathrm{FL}$. At static equilibrium, the condition that Π is minimized (i.e., $\mathrm{d}\Pi/\mathrm{d}L = 0$) implies

$$L \approx \sqrt{\frac{4n^2\pi F}{3p} + \frac{b^2}{3}} \quad (1)$$

As with most PAMs, another feature of the McKibben actuator is that it increases in stiffness when pressurized. This coupling of contraction and stiffening is similar to that of natural skeletal muscle and is a reason why McKibben actuators are referred to as artificial muscle. A corresponding spring constant η can be defined that relates displacement with applied force, that is, $F = \eta \Delta L$. In the limit as ΔL goes to zero, it follows that $\eta \approx \{b\sqrt{3}/2\pi n^2\}p$. This suggests that stiffness increases linearly with increased internal air pressure.

The theoretical approach presented above for relating internal air pressure and actuator deformation can also be extended to flexural pneumatic actuators that bend rather than contract. Studies have examined such coupling in so-called Suzumori[75] and Pneu-Net actuator,[76] which are composed of soft air chambers that are bonded to flexible but inextensible strain limiting elements that control the direction of actuator bending. Although highly simplified, these analytic approximations provide reliable order-of-magnitude estimate for how bending curvature increases with pressure and can be used to inform how material selection and geometric dimensions can influence flexural response.[77]

3.2. Dielectric Elastomers

While fluidic actuators exhibit contractions and stiffnesses that are in the range of natural muscle, their application in soft robotics is limited by their dependency on bulky pneumatic or hydraulic hardware. A promising alternative is to use DEAs and transducers since these can be powered electrically using a portable power supply. The DE technologies used in soft robotics can be thought of as soft capacitors in which capacitance C is coupled to stretch in the plane (λ_1, λ_2) and thickness (λ_3) of the elastic film. In general, $C = \lambda_1\lambda_2 C_0/\lambda_3$, where C_0 is the natural capacitance of the undeformed film. For the special cases of uniaxial stretch and biaxial stretch, it follows that $C/C_0 = \lambda$ and λ^4, respectively, and for pure compression through the thickness (i.e., $\lambda_3 = \lambda < 1$), $C/C_0 = \lambda^{-2}$. This electromechanical coupling can be used both for capacitive sensing and energy harvesting. For a balloon-like transducer (Figure 5D),[78] energy harvesting is governed by biaxial stretch (i.e., $C/C_0 = \lambda^4$) while for a stacked DE capacitor with alternating layers of insulating and conductive silicone (Figure 5E),[79] the electromechanical coupling is $C/C_0 = \lambda^{-2}$. When a voltage (Φ_0) is supplied to the capacitor, a pressure or force is required to maintain a fixed initial capacitance C_0. In this configuration, the capacitor is loaded with charge $q_0 = C_0\Phi_0$, where C_0 is the capacitance of the undeformed transducer. Next, as the electrodes are momentarily disconnected from the power supply and the pressure or force increases, the voltage will also increase. This is due to the increase in capacitance and the fact that the surface charges must remain fixed (due to being disconnected from the power supply). Subsequently removing charge at this higher voltage allows for net electrostatic energy to be delivered to the power supply.

For DE generators, mechanical deformation is used to manipulate internal electric field and produce electrostatic energy. However, the reverse is also possible—electric field can induce mechanical deformation. This arises from the Maxwell stress that is generated from the electrostatic attraction of the surface charges on the opposing surfaces of the dielectric film. The electromechanical coupling between Maxwell stress and elastic deformation has been exploited for a wide variety of DEA architectures. The simplest of these is the annular diaphragms that were used in the early years of DEA research.[80] Minimizing the potential energy Π of these systems with respect to film thickness yields a highly nonlinear relationship between biaxial stretch and voltage. In addition to being nonlinear, such coupling typically exhibits a limit point instability where the film will undergo sudden wrinkling or creasing when voltage exceeds a critical value.[81,82] At these high voltages, the elastomer may also fail due to dielectric breakdown, which occurs when the applied electric field reaches the electric breakdown strength of the elastomer.[83] Such failures are sometimes difficult to avoid since DEAs only undergo significant deformation at voltages close to these critical values.

The principle of minimum potential energy used to model DEAs and fluidic actuators can also be extended to more complex structures that combine dielectric elastomers with semirigid frames constructed from flexural elements. These so-called dielectric elastomer minimum energy structures (DEMES) are composed of thin dielectric membranes that are stretched and bonded to a flexible (but inextensible) frame.[84,85] An example of a flexural DEMES actuator with liquid metal electrodes is presented in Figure 5F. Applying electric field acts to relieve the residual membrane stresses and allows the frame to return to its natural shape. Another recent development

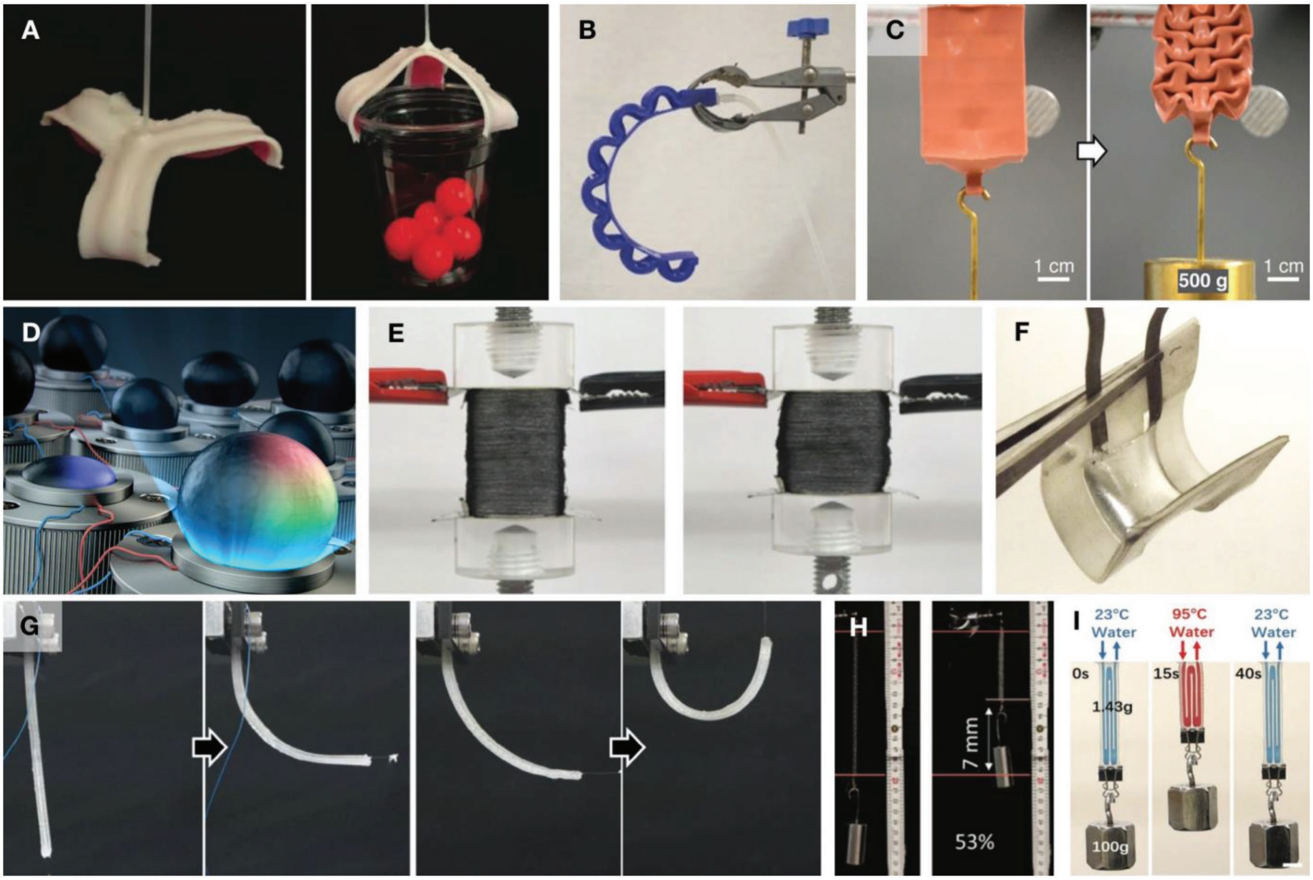

Figure 5. Pneumatic soft robot actuation: A) soft robot gripper produced using direct write printing. Reproduced with permission.[72] Copyright 2018, Wiley-VCH; B) 3D-printed pneumatic flexural actuator. Reproduced with permission.[73] Copyright 2017, Wiley-VCH; C) contractile vacuum-controlled actuator. Reproduced with permission.[74] Copyright 2016, Wiley-VCH. Dielectric elastomer technologies: D) inflation-controlled energy harvesting. Reproduced with permission.[78] Copyright 2012, Royal Society of Chemistry; E) stacked dielectric elastomer actuator (DEA). Reproduced with permission.[79] Copyright 2009, Elsevier; F) flexural dielectric elastomer minimum energy structure (DEMES) with liquid metal electrodes. Thermally responsive actuators: G) bending actuator with shape memory alloy (SMA). Reproduced with permission.[87] Copyright 2017, Elsevier. Reproduced with permission.[108] Copyright 2019, Elsevier Books; H) twisted and coiled polymer muscle composed of a nylon 6 fishing line. Reproduced with permission.[93] Copyright 2018 Wiley-VCH; vascular LCE-based artificial muscle powered by injecting with hot and cold water. Reproduced with permission.[94] Copyright 2018, Wiley-VCH.

has been to combine DEAs with fluidic actuators in order to improve work density and load capacity.[86]

3.3. Thermal Actuators

Although DEAs can operate with mobile electronics, the circuitry need for high-voltage operation can still be bulky and incompatible with miniaturized soft robotic systems. Another alternative for electrically powered actuation is to use thermal actuators like SMAs. Nickel-titanium (nitinol) has been popular for soft actuation because of their high work density and ability to undergo large changes in stiffness and shape when electrically activated. Actuation is controlled by a reversible change from the martensite to austenite crystal phase, which can be induced through resistive (Ohmic) heating of the alloy. Depending on the natural shape of the SMA actuator, this transition can be used to induce bending (Figure 5G),[87] twisting, or contractions, as in the case of an SMA wire that is coiled into the shape of a spring.[88] Since the martensite-austenite phase transition is controlled by temperature, SMA actuation requires careful attention to thermal management. To prevent overheating, researchers have explored techniques ranging from precise voltage control[89] to the use of active cooling[90,91] and integration of thermally conductive soft elastomer[44] for passive heat dissipation.

Other thermal actuators utilize shape-memory polymers that deform in response to applied heat. This includes the twisted and coiled polymer muscle composed of a nylon 6 fishing line originally introduced in ref. [92]. As shown in Figure 5H, the actuator is capable of lifting a 100 g load with a 53% actuation stroke.[93] Another approach to thermal actuation, presented in Figure 5I, involves the integration of microfluidics with LCEs.[94] In this design, the microfluidic channels supply hot or cold water to the LCE in order to rapidly heat it up or cool it down. Compared to external heating and passive cooling, this vascular LCE-based artificial muscle (VLAM) exhibits relatively fast actuation and shape recovery. In addition to shape-controlled actuation, thermally responsive polymer also exhibits reversible stiffness-tuning properties that are also important for artificial muscle applications. Stiffness tuning polymers for robotics has recently been demonstrated in ref. [95] using conductive thermoplastic elastomers and a more general review of stiffness tunable materials for robotics has been presented in ref. [96].

4. Applications and Outlook

The materials and artificial muscle architectures described above have been applied to a wide range of soft robotic systems. Most of these robots are tethered and require external hardware for supplying pressurized air to power pneumatic actuators, high voltage to activate DEAs, or electrical current to heat SMAs. However, progress toward fully autonomous soft robots requires implementations that are untethered and capable of mobility with on-board or embedded hardware. One challenge has been to develop actuators that are powerful enough to carry their own power supply. Some groups have addressed it by significantly scaling up their actuators and creating large soft robots like the 0.65 m long pneumatic quadruped created by Tolley et al.,[97] which is capable of carrying batteries, compressors, valves, and microprocessor. Other attempts at untethered soft robots are engineered to swim or move in an aqueous medium, which reduces load on the actuators through buoyancy and the absence of dry friction. Examples include a fast-moving untethered fish that is actuated using hydrogel-based DEAs that are activated with a built-in high-voltage supply[98] and an autonomous soft robotic fish that swims using hydraulic actuators that are powered with hardware contained within the body.[99,100]

SMA has become popular as an actuator for untethered soft robots due to its low mass, high work density, and ability to be activated using portable electronics for electrical power and control. This has resulted in several recent examples of SMA-powered soft robots that contain an on-board power supply and microcontroller.[101–103] Although promising, the use of SMA for untethered soft robots is limited by the high power consumption and low efficiency of electrical power input to mechanical power output. Moreover, SMA actuators have limited bandwidth due to the time required for the alloy to cool and return to the martensite phase after electrical current is removed. Successful use of SMA in soft robotics therefore requires careful geometric design and choice of surrounding materials in order to ensure adequate thermal management for reliable high-frequency response over extended operating times and lower weight for reduced cost of transport and longer battery life.

Another approach to creating untethered soft robots has been to use combustion. This has led to jumping robots capable of producing large forces.[11,104,105] More recently, controlled combustion has been employed to create an untethered soft octopus robot that does not contain any electronics.[2] Although the robot is not capable of locomotion, it represents a rare example of a fully soft-matter machine that contains absolutely no rigid materials. Other attempts to create fully soft-matter robots typically contain biological tissue, such as the jellyfish-inspired biohybrid robot reported in ref. [106] which can be controlled by applying small electrical charges to the environment. This is accomplished using muscle microtissue that is engineered in a soft elastomer substrate. However, the robots are not autonomous, since the muscle is stimulated using external electrodes or light.

Despite recent successes with engineering untethered soft robots, there is significant room for improvement. In general, creating completely untethered robotic systems is challenging due to issues with materials compatibility and integration. This is because the soft materials used for actuation, sensing, and circuit wiring are not enough for autonomous functionality. These soft material systems also need to interface with rigid microelectronic components for signal processing, communication, and power regulation. Some groups have attempted to create more universal frameworks for creating hybrid material systems that combine soft elastomers and fluids with rigid miniaturized components.[15,107] Improved materials integration can also be accomplished through advancements in functional-graded materials with smooth transitions in material stiffness. This can eliminate the damage-causing stress concentration that often arise at the interface between soft and rigid materials.

Untethered functionality represents just one step toward soft robot autonomy. Referring back to the definition presented in Section 1.1, soft robots must eventually be capable of understanding their physical state and environment or about its own physical state and perform physical task based on decisions that use this information. Soft-matter engineering provides a framework for engineering machines that have the actuators, sensors, and electronics to achieve this autonomy. However, autonomous functionality also depends on advancements in algorithms for sensing, planning, adaptive learning, and control that are specialized to the unique mechanics of soft material systems. Therefore, although many of the near-term efforts in soft robotics should remain focused on materials integration, future efforts must broaden the research to address machine intelligence, planning, and other aspect of robot autonomy.

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Conflict of Interest

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