Embedded Three-Dimensional Printing of Autonomous and Somatosensitive Soft Robots

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Embedded Three-Dimensional Printing of Autonomous and Somatosensitive Soft Robots

A dissertation presented

by

Ryan Landon Truby

to

The John A. Paulson School of Engineering and Applied Sciences

in partial fulfillment of the requirements

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Embedded Three-Dimensional Printing of Autonomous and Somatosensitive Soft Robots

ABSTRACT

Soft robots possess attributes that are difficult, if not impossible, to achieve with traditional robots. However, soft robots remain dependent on rigid hardware components, inhibiting the full benefits of being soft. New fabrication methods that enable the assembly of soft matter with multifunctional properties are needed to realize soft robots’ full potential. This dissertation focuses on new material and manufacturing strategies for creating soft robots that possess soft analogues of power, control, and sensory components.

Central to this research is a technique called embedded three-dimensional (EMB3D) printing, in which functional inks are extruded through nozzles that translate omnidirectionally within viscoplastic matrix materials. For this work, several functional inks and elastomeric matrices that cure after printing were designed for printing soft robots with pneumatic/fluidic, catalytic, and sensory elements. We first investigate how matrix rheology, various printing parameters, and print path influence printing results. We find that maximizing the Oldroyd number associated with EMB3D printing conditions improves print fidelity.

Next, we use EMB3D printing to assemble the “Octobot,” the first entirely soft, autonomous robot capable of untethered operation. Octobots are controlled by microfluidic logic-based devices that autonomously regulate the catalytic decomposition of an on-board monopropellant fuel supply. Gas generated from fuel decomposition inflates networks of fluidic actuators. The Octobot’s body and microfluidic logic are fabricated using molding and soft lithography, respectively, while all mesofluidic
and catalytic features are EMB3D printed. The fluidic and elastomeric architectures required for Octobot operation are straightforwardly achieved with EMB3D printing.

Lastly, we EMB3D print soft, somatosensitive actuators (SSAs) that are innervated with conductive features that enable bioinspired haptic, proprioceptive, and thermoceptive sensing. Each sensor is printed with our ionogel-based sensor ink and exhibits long-term stability and hysteresis-free conduction. For a simple demonstration, SSAs were assembled into soft robotic grippers, and their somatosensory feedback during handling of spherical objects of varying stiffness, textures, and temperature was characterized.

With EMB3D printing, we have created soft robots with novel attributes. Our manufacturing platform can be adopted to design other soft robots that are entirely soft, require somatosensory feedback for improved control, or cannot be made with other manufacturing methods.
TABLE OF CONTENTS

ABSTRACT .................................................................................................................................................... iii

ACKNOWLEDGEMENTS .............................................................................................................................. vii

CHAPTER 1 - INTRODUCTION .................................................................................................................... 1
  1.1 Dissertation Scope .............................................................................................................................. 2
  1.2 Dissertation Organization .................................................................................................................... 3

CHAPTER 2 - LITERATURE REVIEW .......................................................................................................... 5
  2.1 Motivation and Scope .......................................................................................................................... 5
  2.2 Soft Robotic Systems .......................................................................................................................... 6
    2.2.1 Soft Actuators .............................................................................................................................. 6
    2.2.2 Molding and Digital Fabrication ................................................................................................. 17
    2.2.3 Power and Control for Soft Robotic Systems .............................................................................. 27
    2.2.4 Soft Sensors ............................................................................................................................... 30
    2.2.5 Progress in coupling soft robotic sensors and actuators ............................................................ 40
  2.3 Applications ....................................................................................................................................... 42
  2.4 Conclusions ........................................................................................................................................ 45

CHAPTER 3 - OPTIMIZING MATRIX RHEOLOGY FOR EMBEDDED THREE-DIMENSIONAL PRINTING .......... 46
  3.2 Experimental Methods ....................................................................................................................... 50
    3.2.1 Matrix and Ink Preparation .......................................................................................................... 50
    3.2.2 Rheological Measurements ......................................................................................................... 50
    3.2.3 Embedded 3D Printing ................................................................................................................ 51
    3.2.4 Imaging and Videography ........................................................................................................... 52
  3.3 Results and Discussion ........................................................................................................................ 52
    3.3.1 Matrix Design and Rheology ....................................................................................................... 52
    3.3.2 Embedded 3D Printing ................................................................................................................. 56
    3.3.3 Matrix Material Yielding During Embedded 3D Printing ........................................................... 59
  3.4 Conclusions ........................................................................................................................................ 63

CHAPTER 4 - EMBEDDED THREE-DIMENSIONAL PRINTING OF ENTIRELY SOFT, AUTONOMOUS ROBOTS .... 64
  4.2 Experimental Methods ....................................................................................................................... 65
    4.2.1 Soft Controller Fabrication ......................................................................................................... 65
    4.2.2 Preparation of the Inks, Matrix Materials, and Fugitive Plug Material ..................................... 66
    4.2.3 Rheological Characterization of Inks and Matrix Materials ...................................................... 68
    4.2.4 Octobot Mold Fabrication .......................................................................................................... 69
    4.2.5 Octobot Assembly ....................................................................................................................... 69
    4.2.6 Actuator Characterization ........................................................................................................... 72
    4.2.7 Untethered, Autonomous Soft Robot Operation ......................................................................... 74
    4.2.8 Imaging and Videography ........................................................................................................... 74
  4.3 Results and Discussion ........................................................................................................................ 75
    4.3.1 Octobot Design ............................................................................................................................. 75
    4.3.2 Matrix Materials and Inks ............................................................................................................. 77
    4.3.3 Printing and Auto-Evacuation of the Fugitive and Catalytic Inks .............................................. 79
    4.3.4 Free Displacement and Blocked Force Characterization of the Actuators .................................. 81
    4.3.5 Autonomous, Untethered Octobot Operation ............................................................................. 82
  4.4 Conclusions ........................................................................................................................................ 86
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For my first teacher, my greatest hero, and my best friend –

Thank you for giving me the world, Mama.

I love you.
“Climb the mountain not to plant your flag, but to embrace the challenge, enjoy the air and behold the view. Climb it so you can see the world, not so the world can see you.”

David McCullough, Jr., *You Are Not Special...And Other Encouragements*
CHAPTER 1

INTRODUCTION

Robots comprised of soft materials can exhibit many attributes that are difficult, if not impossible, to realize with robots based on conventional rigid materials or power and control strategies. Owing to their soft, compliant nature and dynamic physicochemical properties, “soft robots” have tremendous potential in applications requiring robots with exceptionally complex motions, deformability, dynamic and biomimetic functionalities, abilities to perform or adapt to undefined, unstructured environments, and resilience in environments ordinarily considered inappropriate for robot deployment. The inherent mechanical similarities between the materials comprising soft robots and our own bodies are opening new opportunities for technology development at the human-robot interface. In short, soft robots are excelling in areas where traditional robots have not, opening new avenues for applications such as automation, manufacturing, medicine, rehabilitation, wearable technologies, soft tactile interfaces, search-and-rescue, and more [1–3].

The field of soft robotics emerged in earnest in the early 2000s [1], decades after seminal works in stimuli-responsive materials [4], artificial muscles [5, 6], and deformable “soft” electronics [7–10]. At present, inflatable fluidic elastomer actuators and electroactive polymer actuators remain a foundation of soft robotics design strategies [2, 11–13] and have enabled soft robotics to establish itself as an eminent field of research at the interfaces of materials engineering with other scientific and engineering disciplines. However, despite tremendous progress in soft robotics over the last decade, soft robots still rely on conventional robotic hardware for power and control [2, 3, 11, 14–17]. These dependencies fundamentally prohibit soft robots from being fully soft.

The ability to programmably assemble soft matter with disparate mechanical and material properties into soft robotic architectures is central to realizing the full potential of this new class of robots [2]. My Ph.D. thesis focuses on new material and manufacturing strategies for creating soft robots with
soft analogs of crucial power, control, and sensory components. Central to my research is the development of a free-form, multimaterial assembly technique called embedded three-dimensional (EMB3D) printing. In this additive manufacturing approach, functional inks are extruded through a nozzle that is translated omnidirectionally within soft, viscoplastic matrix materials [18–20] that surround and support the printed features. By using specifically tailored functional inks, we can pattern pneumatic, catalytic, and sensory elements that enable autonomous and somatosensory soft robots, which operate without auxiliary hardware and are innervated with multiple soft sensors, respectively.

1.1 Dissertation Scope

My dissertation first aims to develop and characterize a model matrix material for EMB3D printing. Next, I use EMB3D printing as part of an integrated design and fabrication approach for creating the Octobot, the first entirely soft robot that operates autonomously. The Octobot takes the form of an octopus because this soft-bodied organism has long been a source of inspiration for the field of soft robotics. Finally, I use EMB3D printing to create soft somatosensitive actuators (SSAs), i.e., fluidic elastomer actuators that are innervated with multiple ionically conductive features, that provide several modes of somatosensory feedback, including proprioceptive, tactile, and thermoceptive sensing.

The principal accomplishments of my Ph.D. dissertation are the design and fabrication of autonomous, entirely soft robots and soft robotic actuators with multiple somatosensory feedback motifs. Central to these outcomes was the development and characterization of several functional inks and elastomeric matrix materials for EMB3D printing. For our entirely soft Octobots, we designed a microfluidic logic [21] device – a “soft controller” – to autonomously regulate the selective decomposition of an on-board, monopropellant fuel supply [22] that powers alternating actuation cycles of printed fluidic elastomer actuators. The body and microfluidic logic of the robot are fabricated using molding and soft lithography, respectively, and the pneumatic actuator networks, on-board fuel reservoirs, and
catalytic reaction chambers needed for movement are patterned within the body via EMB3D printing. The Octobots represent the first machine controlled via microfluidics. The fluidic and elastomeric architectures required for Octobot function span several orders of magnitude from the microscale to the macroscale. With our straightforward approach, all features required for Octobot operation are seamlessly and rapidly achieved. Finally, the SSAs’ possess new types of somatosensory feedback, including thermoceptive and fine-and-deep touch sensing, which are enabled by a new sensor ink based on an organic ionogel. The ionogel-based sensors exhibit superior long-term stability and hysteresis-free performance, unlike other soft sensor materials. Seamlessly innervating soft robotic actuators with complex sensing networks via EMB3D printing represents a foundational advance with potential applications in soft robotic, wearable, and haptic devices.

Both of these novel soft robotic systems possess new capabilities not previously demonstrated by other researchers in the soft robotics field. Our integrated design, materials, and 3D printing approach can be readily adopted to create other soft robotic systems that are entirely soft, require somatosensory feedback for improved control, or cannot be made by traditional manufacturing methods.

1.2 DISSERTATION ORGANIZATION

My dissertation begins with a review of the relevant literature in the design, manufacturing, and power and control strategies for soft robotic actuators and sensors in Chapter 2. In Chapter 3, I report the design of a model thixotropic matrix material for crevice-free EMB3D printing, characterize its enabling rheological behavior, and explore the influence of various printing parameters on overall print fidelity with this technique. In Chapter 4, I use this model matrix material and fugitive and catalytic inks to EMB3D print the Octobot, an entirely soft robot controlled by on-board microfluidic logic and pneumatically powered by regulated monopropellant decomposition reactions. In Chapter 5, I use EMB3D printing and a new sensor ink based on a conductive organic ionogel to create SSAs that possess
proprioceptive, tactile, thermoceptive, and fine-versus-deep touch sensing capabilities. Finally, in Chapter 6, I provide the principal conclusions of my Ph.D. work, and in Chapter 7, I discuss new challenges and opportunities for future related work.
CHAPTER 2

LITERATURE REVIEW

2.1 MOTIVATION AND SCOPE

The emerging field of soft robotics focuses on using soft matter to create robotic systems that exhibit myriad functionalities, which are not possible using conventional rigid components. By virtue of their soft, compliant, physicochemical properties, soft robots have tremendous potential in many applications, including biomedicine, wearable technologies, automation, and well beyond [1–3]. Though the term “soft robot” was not coined until the 2000s [1], researchers first proposed using various soft materials as artificial muscles in the 1950s [4, 5]. Advances in the directed assembly of soft matter via micropatterning, soft lithography, and additive manufacturing have enabled the fabrication of soft robots capable of controlled actuation and movement. Since comprehensive reviews on soft robotics have been published [1–3, 11, 23–25], this chapter focuses on engineering strategies for soft robotic systems, including soft actuator design and fabrication, power- and control-related challenges, and integrating soft sensors. Finally, we highlight several applications for soft robots.

2.2 SOFT ROBOTIC SYSTEMS

Soft matter encompasses a broad range of biological and synthetic materials, including thermoplastic, thermosetting, and elastomeric polymers, liquid crystals and liquid crystalline polymers, complex fluids, hydrogels, and granular media. These “soft” materials possess Young’s moduli ranging from 1 kPa to 1 GPa (Figure 2.1) as well as viscoelasticity, compliance, toughness, and even the ability to reversibly undergo and completely recover from large strains. The properties of soft materials are determined by their chemical composition, structural form, and organization across multiple length scales. Importantly, their physicochemical properties enable a broad array of behaviors, including
Figure 2.1. Young’s modulus of selected engineering, biological, and soft materials. Approximate values of Young’s modulus are indicated for various biological (green font) and synthetic/abiotic (blue font) materials. (Adapted from Refs. 2 and 25).

stimuli-responsive shape-changing behavior, rapid and reversible phase transformations such as gelation, self-healing properties, and electric or ionic conductivity [26, 27].

Soft robots not only possess a soft exterior: they employ soft materials for actuation, sensing, and even computational capabilities that cannot be realized with rigid hardware, such as motor assemblies, electronic components, and batteries [1–3]. Consequently, the complex interplay between soft robotic materials, manufacturing, power, and control strategies pose as many exciting research questions as grand challenges for this nascent field [1–3] (Figure 2.2). The sections below are structured to illustrate these relationships.

2.2.1 Soft Actuators

Soft robotic actuators are capable of large mechanical deformations and ensuing material shape changes in response to applied pressure or other external stimuli. There are numerous criteria to consider for soft actuator designs: generated force and strain, actuation speed, actuator form factor, operating environment, biocompatibility, mechanical properties and performance, power requirements, and control schemes [25]. The last two factors are most crucial to actuator use and performance, as both are highly dependent on a soft actuator’s composition and overall capabilities (Figure 2.2). Many soft material
Figure 2.2. Soft robotics design. Designing soft robotic systems requires consideration of the materials and manufacturing methods employed, necessary power sources, and compatible control strategies.

Actuators [3, 24, 25], including electroactive [6, 12, 13, 28–31], swellable and hygromorphic [4, 32–42], thermally-responsive [43–50], photoresponsive [51–58], magnetoactive [59–65], and biological actuators [66–72], have been explored and demonstrate soft robotic potential. However, most soft robots rely on fluidic elastomer actuators (FEAs) to induce the desired locomotion [2, 73]. Our discussion therefore begins with an overview of FEAs, followed by a brief description of other promising classes of soft robotic actuators.

2.2.1.1 Fluidic Elastomer Actuators

FEAs comprise a broad class of elastomeric composites with functional architectures and mechanical properties that enable directional actuation via inflation with a liquid or gas. FEAs were one of the first examples of soft actuators developed [5]. They arguably remain the most
Figure 2.3. Early designs of fluidic elastomer actuators (from 1950-2000). (A-C) Design and operating principles of McKibben actuators. (A) The McKibben actuator consists of a gas-tight, elastic tube sheathed by an outer braided sleeve. When inflated, the inner tubing expands against the braided sleeve, which acts to restrict longitudinal elongation and drive radial expansion. The result is a contractile actuation. (B) The generated force increases with inflation pressure and can lift a considerable payload. ((A-B) are adapted from Ref. 75.) (C) McKibben actuators were originally designed for orthotic devices for various rehabilitation needs (adapted from Ref. 5). (D-E) Early multi-chamber FEA designs fabricated from micro-molded, fiber-reinforced elastomers. (D) Multiple degrees of freedom were possible through inflating different chambers (adapted from Ref. 81). (E) Early examples of micro-manipulators and walking systems based on early FEAs (adapted from Ref. 80). (F) An early demonstration of a FEA-based endoscope (adapted from Ref. 85).

The first FEA was the braided fluid actuator invented in 1955 by J. L. McKibben for rehabilitative orthoses (Figure 2.3A-C). The McKibben actuator consists of an inflatable piece of elastic tubing fixed at each end with appropriate closures for gas-tight inflation and sheathed with a braided sleeve, mesh, or
fabric as a flexible constraining layer (Figure 2.3A). Upon inflation, the elastic tubing expands (Figure 2.3B), and the outer braided layer restricts inflation such that a force-generating, contractile motion is produced [5] (Figure 2.3C). Many variations of the McKibben actuator have been developed for applications requiring synthetic robotic muscles [75], though the original design is still widely used today, for example, in wearable exosuit devices [76, 77].

Beginning in the late 1980s, FEAs inspired by initial McKibben actuator designs were developed for inflation-powered locomotion of mobile robots. In 1989, a gas-powered, mobile robot for internal pipe inspection was developed that utilized a set of McKibben actuators with various nylon constraining sleeves for stretching and bending actuation modes. Control of stretching and bending via specific inflation sequences enabled the device to travel through pipes with annelid-like locomotion [78]. In the early 1990s, designs of FEAs with multiple degrees of freedom enabled by elastic tube-like architectures with individually addressable, inflatable chambers were reported [79, 80] (Figure 2.3D). Various end effectors, micromanipulators [81, 82], and biomimetic crawling systems were achieved with these tentacle-like actuators [83] (Figure 2.3E). By 1996, advances in digital manufacturing enabled the design of similar micro-actuators [84], which had begun to garner interest for micro- and endoscopic surgery applications [85, 86] (Figure 2.3F). Continued advances in micro-electro-mechanical (MEMS) systems, microfabrication, and soft lithography quickly enabled many innovations in FEA design [87]. While most work focused on actuators for micromanipulation [88, 89], progress in computer-aided design (CAD) and additive manufacturing facilitated the fabrication of larger elastomer-based robotic systems, such as swimming rays [90] (Figure 2.4A-B) and soft articulating surfaces [91]. In the early 2010s, multi-material, multi-network FEAs (Figure 2.4C-D) were reported that produced different gaits in soft tetrapods (Figure 2.4C) as well as soft robots capable of gripping and gently holding objects as fragile as eggs (Figure 2.4D) [74, 92].
FEAs deform upon inflation as a result of asymmetric, directional volumetric expansion of an embedded fluidic network within an elastomeric composite. As the working fluid fills this network, the network increases in volume and expands against the surrounding matrix where the compliance is lower. The FEA can deform to accommodate differences in strain [74, 92]. FEA deformation is highly dependent on the architecture, composition, and mechanical properties of the constituent elastomeric composite. Different types of FEAs have been developed with bending, twisting, torsion, contracting and extending actuation motifs [25] (Figure 2.5).

Many elastomers are appropriate for FEAs, provided they facilitate some degree of volumetric expansion under appropriate operating pressures [11, 74]. Variations of elastomer chemistries have enabled interesting FEA mechanical and functional properties, such as self-healing capabilities [93]. However, given the viscoelasticity and non-linear mechanical properties of typical FEA materials, rationally designing FEAs for specific displacement and force generation is non-trivial. Generally, FEA force generation and displacement both increase with higher bending moment in the FEA, which is
Figure 2.5. Various FEA designs for bending, torsion, and extension/contraction actuation motifs. (A-C) Bending (A), torsion (B), and extension/contraction (C) actuation motifs are shown for composite FEA architectures. The visual key beneath indicates material stiffness and pressure required (schematics adapted from Ref. 25).

generated by inflation. Bending moments typically increase with operating inflation pressure and volume of working fluid. Force generation also increases when the FEA is comprised of stiffer materials and when liquids are used as working fluids instead of gases. These trends and the extent to which an FEA can displace and generate force are all highly dependent on the geometry and composition of the FEA [24, 73, 74, 87, 92, 94–96]. Clearly, there are trade-offs to consider in FEA design for soft robots: softer materials cannot sustain high inflation pressures needed for high force generation and can burst, leading to device failure, while stiffer materials may require operating pressures that cannot be readily supplied with available hardware.

One advantage of soft robots over conventional systems is the ability to enable impressive actuation motifs with relatively simple control inputs (i.e., inflation pressures for FEAs). A number of sophisticated FEAs have been developed, including biomimetic, tentacle-like end effectors [97] (Figure 2.6A), rapid finger-like actuators [96] (Figure 2.6B), and pneumatically actuated wheels with integrated FEAs for rotation [98] (Figure 2.6C). Recent work has investigated the use of open-cell, elastomer foams in FEAs that can be bulk processed for simpler manufacturing [99, 100] (Figure 2.6D). Finally, others have used inflatable pouches [101], pockets [102], and membranes [103] for soft robotic FEAs. These
Figure 2.6. Novel designs of FEA fluidic networks for new soft robotic functionalities. (A) A tentacle-like FEA with multiple degrees of freedom possesses (i) a rigid PDMS core surrounded by an extensible Ecoflex layer in which (ii) several fluidic networks are embedded. (iii) Addressable inflation of these sections gives rise to tentacle-like grabbing of various objects (adapted from Ref. 97). (B) A FEA design capable of actuating rapidly (adapted from Ref. 96). (C) A FEA-actuated car uses (i) FEAs for wheel rotation. (ii) The car is assembled and can (iii) be actuated for rolling over (iv) various terrains (adapted from Ref. 98). (D) FEAs made with (i) open cell foam architectures (ii) bend upon inflation. (iii) Inclusion of strain limiting layers can enable extension. (iv) The open foam architecture of the fluidic FEA network as shown by micro computed tomography (adapted from Ref. 99). (E) An origami-inspired metamaterial is actuated with pocket-like FEAs. (i) Inflating the air pockets induces folding of the structure for (ii) various conformations or actuation states (adapted from Ref. 102).

Actuators, made with various heat stamping methods, have been used for pneumatically addressable hinges in composites comprised of materials stiffer than silicone elastomers for robotic grippers [101] and actutable, origami-inspired metamaterial structures [102] (Figure 2.6E). Eversion of an inflatable
membrane composite steered by addressable pressurization has been used for rapid lengthening and movement of a soft robot that mimics the growth of plants. With this approach, a soft robot can “grow” in length by thousands of percent [103].

While typical FEA$s require a positive pressure from the inflating liquid or gas, some fluidic actuators use a vacuum to drive actuation. For convenience, we also classify vacuum actuated elastomeric composites as a type of FEA. Most notably, jamming transitions in granular media induced under a vacuum have been used for universal object manipulation [104] and stiffening in soft robotics, while recent work has used a vacuum for contractile FEAs. Specifically, vacuum-based approaches can induce compressive buckling in molded elastomers that result in subsequent contraction of the structure [105]. With the proper architecture, discrete buckling events triggered under vacuum can also enable rotational motion for simpler grabbing [106, 107].

Despite their advantages, FEAs typically possess a low power density, generate relatively low force, low actuation speeds, and display inconsistent performance owing to their viscoelastic properties and strain-induced weakening (i.e., the Mullins effect [108, 109]). Device failure via over-pressurization and delamination is also common in FEAs. Tremendous work is needed to further improve FEA design and performance. Advances in new materials and manufacturing methods for FEAs are essential for improved function [11].

2.2.1.2 Stimuli-responsive, Soft Robotic Actuators

Soft actuators have been developed using materials that undergo mechanical deformations and ensuing shape changes in response to solvent or vapor swelling [4, 32–42], applied electric field [6, 12, 13, 28–31], heat [43–50], light [51–58], or magnetic field [59–65], as well as biological activity [66–72]. Many of these actuation strategies are in very early development for soft robotics to be extensively reviewed here.
However, several electroactive polymer actuators (EPAs) and liquid crystalline elastomers exhibit significant potential as soft robotic actuators.

EPAs represent a promising class of actuators for soft robotics given their “plug-and-play” compatibility with conventional robotics power and control hardware [6, 12, 13, 25, 28–31]. Two relevant classes of EPAs for soft robotics are dielectric elastomer actuators (DEAs) and ionic polymer metal composite (IPMC) actuators. EPAs first showed potential as viable artificial muscles and biomimetic actuators in the 1990s when seminal advances in DEAs, including high strains (>100%) and power densities [110, 111], were made by SRI International. DEAs are field-activated EPAs that experience in-plane deformations under electrostatic pressures called Maxwell stresses. Maxwell stresses are produced by Columbic interactions between separated surface charges that are generated by a voltage applied across an elastomeric, dielectric membrane that is coated with deformable, conductive electrodes [6]. DEAs are capable of large force generation, high strain, and high power density. For significant deformations and actuation strains, DEAs generally have voltage requirements of ~100 V per micron of membrane thickness, and with current fabrication methods, this physical constraint means that DEAs require remarkably large voltages – generally tens to hundreds of kilovolts – for operation [6, 12, 30]. Despite these and other notorious challenges, DEAs show great promise as soft robotic actuators [12, 30]. Several DEA-based soft robots have been developed, which crawl [112], walk [113], swim [17, 114], and grab delicate objects [12, 115–117]. Recent advances in DEA manufacturing are enabling completely new functionalities, as demonstrated by the fast multi-DEA soft crawler shown in Figure 2.7A, that exhibits travel speeds of ~2 cm/s and moderate operating voltages of ~3 kV [118].

IPMCs are ionic EPAs that electromechanically actuate as a result of ion transport through a conductive polymer network. IPMCs are typically composed of an anionic polymer membrane with high ionic conductivity, imbibed with mobile cations, and possessing two noble metal electrodes on opposite surfaces [13, 28, 119]. Voltages applied across these metallic electrodes induce migration of mobile cations
Figure 2.7. Stimuli-responsive, soft robotic actuators. (A) New advances in dielectric elastomer actuator (DEA) fabrication are facilitating the design of multi-DEA soft robots. (i, ii) Integration of passive, rigid elements (black lines) enabled high curvature DEA bending for (iii) high speed crawling (adapted from Ref. 118). (B) Ionic polymer metal composites (IPMCs) have recently been used for swimming soft robots. (i) Embedding multiple IPMCs in a soft elastomer matrix to create fins for a (ii) soft robotic swimmer. (iii) Addressable actuation of individual IPMCs creates fin-like actuation (adapted from Ref. 127). (C, i) Principles of LCEA actuation (adapted from Ref. 25). (ii) A famous demonstration of a LCEA lifting a 10g payload (adapted from Ref. 49). (iii) With novel optical alignment methods, “voxelated” LCEA films were patterned. Polarized optical microscopy shows the radial alignment of liquid crystal mesogens (top). The mesogen alignment motif is shown schematically (below). (iv) Heating the LCEA film above its nematic-isotropic transition temperature drives anisotropic, out-of-plane deformation. (v) Molecular structures of the mesogens and main-chain liquid crystal polymers used to create these voxelated LCEAs (adapted from Ref. 50).

towards the cathode. Increased electrostatic interactions at the cathode induce directional expansion of the polymer network that drives actuator bending toward the anode. [28]. Compared to DEAs, IPMCs have notably lower operating voltages of around 1 – 5 V. However, actuation times for IPMCs are much slower due to ion diffusion [13, 28]. While IPMCs have long been identified as an alternative actuator to DEAs, their integration into soft robotics has only recently begun [13, 120, 121]. For example, IPMCs for MEMS, micromanipulation [120, 122, 123], swimming [124], and
untethered crawling [31] systems have emerged. While much work remains in developing novel IPMC assembly methods [125] and materials [126], recent advances have led to the design and assembly of large soft robotic swimmers (Figure 2.7B) [127].

Finally, LCEAs are elastomers capable of undergoing reversible contraction. They offer unique capabilities for muscle-like actuators in soft robotics. Moreover, their chemistries can be readily tailored for various mechanical properties and response to multiple stimuli [46, 47, 128]. LCEAs are composed of liquid crystal mesogens that are directly integrated into or bound as pendant groups to the backbone of their polymer network. If aligned appropriately through various molecular, electrical, magnetic, or mechanical alignment techniques [48], these mesogens can order along a nematic director and maintain this ordered mesophase below their nematic-isotropic transition temperature ($T_{NI}$). In the nematic phase, the polymer network of the LCEA tends to elongate, extending the chains beyond higher entropy random-coil conformations. Increasing their temperature above $T_{NI}$ results in disruption of the liquid crystalline order, and a restoring force from the LCEA will be generated as its polymer network adopts higher entropy, random-coil chain conformations. The force follows along the nematic director, typically generating uniaxial contraction. At temperatures below $T_{NI}$, the mesogens readily associate with each other via the formation of hydrogen bonds and π-π stacking. Thus, cooling the LCEA below the $T_{NI}$ drives re-ordering of the mesogens, extension of the polymer network, and a return to the original configuration (Figure 2.7Ci) [46–48]. LCEAs can be made photoresponsive by integrating azobenzene groups that exhibit cis-trans photoisomerization into the LCE network. When an azobenzene-functionalized LCEA is exposed to ultraviolet (UV) light, the azobenzene group in enters a cis conformation, disrupts the LCEA’s nematic order, and induces shape change.

The muscle-like behavior of LCEAs is clearly shown in Figure 2.7Cii, which highlights a famous experiment in which a particular LCEA is shown lifting a 10 g load upon heating [49]. Most LCEAs only show uniaxial contraction due to inherent limitations of available alignment methods. In 2015,
“voxelated” LCEAs were created with spatially controlled nematic directors via novel optical alignment techniques (Figure 2.7Ci iii). Upon heating, complex, out-of-plane transformations were achieved (Figure 2.7Civ). Vital to this advance was the large actuation strains enabled by a novel main-chain LCEA chemistry (Figure 2.7Cv) [50]. While LCEAs can uniquely provide reversible, inherent contractile actuation with large forces and strains, they are expensive and must be nematically oriented via a controlled alignment process [47, 48].

2.2.2 Molding and Digital Fabrication

Here, we focus on both molding techniques and several light- and ink-based 3D printing methods for fabricating FEA-based soft robots.

2.2.2.1 Molding

Most FEAs are fabricated via multi-step, molding-and-laminating methods inspired by soft lithography and other micro-molding techniques (Figure 2.8). The molding process is not only used to define the overall form and composition of the FEA, but also its internal, embedded fluidic network. One typical procedure for assembling a bending FEA is provided in Figure 2.8A. Bending-style FEAs bend upon inflation (Figure 2.8B-C) as a result of asymmetric, directional volumetric expansion of the embedded fluidic network. As discussed previously, the FEA bends to accommodate differences in strain of the elastomeric composite generated by inflation [74, 92]. In monolithic FEAs, expansion will tend towards regions of the surrounding matrix with thinner features (Figure 2.8Di). Bending is more commonly achieved by introducing inextensible-flexible materials as strain-limiting layers to drive FEA bending while mitigating extension (Figure 2.8Dii) or using multiple materials with disparate extensibilities and stiffness (Figure 2.8Diii). Popular strain-limiting materials include paper [96], fabrics [95, 129], and even Kevlar for puncture-resistant FEAs [130]. Variations of these methods, including lost-
Figure 2.8. Design and manufacturing principles for popular bending FEAs. (A) A typical manufacturing procedure for producing bending FEAs like those shown in (B-C). (B-C) Schematics (B) and photographs (C) of a typical bending FEA before (left) and after (right) inflation (adapted from Ref. 92). (D) Strategies for inducing FEA bending include (i) structural asymmetry (where T₂ > T₁ in the illustration), (ii) the inclusion of strain-limiting layers, such as a fabric, and (iii) composite designs, where Elastomer 1 is stiffer than Elastomer 2 in the illustration (adapted from Prof. M. Wehner, UC Santa Cruz).

wax [131, 132] and rotational casting [133] approaches, and have been used to create the FEA architectures illustrated previously in Figure 2.5 [25]. Molds for these processes have been fabricated from various subtractive and additive manufacturing techniques, including micromachining and soft lithography for small form factor devices, and computer-assisted machining and 3D printing for larger devices [73].

2.2.2.2 Light and Ink-based 3D Printing

Advances in additive manufacturing over the last two decades have revolutionized the science and engineering of advanced soft materials. Many techniques have emerged for digitally fabricating soft
Figure 2.9. Light- and ink-based 3D printing methods used in soft robotic fabrication. Schematics of (A) stereolithography (SLA), (B) selective laser sintering (SLS), (C) PolyJet®, (D) fused deposition modeling (FDM), and (E) direct ink-writing (DIW). (Figure from Ref. 20).

robots. While most advances in 3D printing soft robotic actuators have been for FEA fabrication [134], there is growing progress in 3D printing soft robotic DEAs [135], IPMCs [136, 137], and LCEAs [138, 139].

Stereolithography (SLA) of photocrosslinkable resins [140, 141] (Figure 2.9A) and selective laser sintering (SLS) of polymeric powders [142] (Figure 2.9B), serve as the foundation for nearly all light-based 3D printing methods. While SLS has been used by robotics companies such as Festo to manufacture FEAs [143], most light-based 3D printing methods that have been used for soft robotics fabrication are based on SLA. SLA includes variations such as digital projection lithography (DLP) [144, 145], continuous liquid interface production (CLIP) [146], and two-photon polymerization (2PP) [147]. All SLA-based techniques leverage a basic concept for printing: by selective photopolymerization of a low-viscosity, liquid resin in a layer-by-layer pattern, 3D objects can be formed. Various SLA methods have different capabilities in terms of possible resolution and print speed, requiring consideration of inherent tradeoffs between print resolution, overall build volume, and time. Notably, SLA methods do not readily enable one to pattern multiple materials within a single build sequence, which would require, at minimum, multiple reservoirs containing different, yet chemically compatible, resins [20].
Figure 2.10. Examples of soft robotic actuators 3D printed with stereolithography-based methods. (A) A multi-chambered FEA inspired by antagonistic muscle pairs. (i) Schematic of the internal chambers. (ii-iv) The FEA has four degrees of freedom. The FEA is shown in (ii) a non-inflated state, (iii) when the top left chamber is inflated, and (iv) when the front bottom chamber is inflated (adapted from Ref. 148). (B) A SLA-printed soft FEA gripper. The gripper is shown in (i) inflated and (ii) non-inflated states and (iii) while picking up an object (adapted from Ref. 149). (C) A transparent FEA gripper SLA printed from a photocrosslinkable hydrogel resin can gently catch and release a fish (adapted from Ref. 151).

SLA was first used in the mid-1990s to print multi-chamber, tentacle-like FEAs from various photopolymer thermosets [84]. SLA printing of silicone-based photocrosslinkable resins has more recently been demonstrated to create arbitrarily complex fluidic chambers in FEAs to drive multidirectional actuation upon inflation (Figure 2.10A) [148]. Multi-actuator soft grippers comprised of several FEAs have been printed in a single SLA run (Figure 2.10B) [149]. By introducing thiol-ene click chemistries into silicone resin formulations, FEAs have been printed with intrinsic self-healing abilities [150]. Finally, photocrosslinkable hydrogel resins have been developed for SLA printing of tough,
hydrogel-based FEAs that are completely transparent underwater (Figure 2.10C) [151]. SLA printing, though clearly promising for the design and fabrication of FEAs with complex architectures, is still limited in the multi-material heterogeneity it can provide printed soft robotic actuators. Finally, photopolymer resins still require further development to achieve appropriate FEA mechanical behaviors, such as higher extensibility and softness than is currently available.

Myriad soft materials can be patterned by ink-based 3D printing methods. Printable inks are formulated from molecular, polymeric, or particulate species to achieve the desired flow behavior for either ink droplet- or filament-based methods. Specific parameters include the ink viscosity, surface tension, shear yield stress, and shear elastic and loss moduli, which must be tailored for a given printing method [20]. Three ink-based 3D printing methods, PolyJet™ (an inkjet technique) (Figure 2.9C), fused deposition modeling (FDM) (Figure 2.9D), and direct-ink writing (DIW) (Figure 2.9E) have been used to manufacture soft robots.

PolyJet printing is a droplet-based, 3D printing method that has been used extensively in soft robotics, both for mold production in molding-based fabrication methods as well as direct printing of FEAs. In this approach, photocrosslinkable resins are deposited using inkjet print heads and polymerized upon printing by illumination with an UV light source (Figure 2.9C). These inks require a low viscosity, and ink drop formation is dependent on both material and printing parameters, including ink density ($\rho$), viscosity ($\mu$), surface tension ($\gamma$), characteristic length of the droplet ($L$, which in most cases, is taken as drop diameter), the velocity of the ejected droplet ($v$), and nozzle diameter ($d$). These parameters must be tightly controlled to achieve a specific balance between viscous, surface tension, and inertial forces. The dimensionless $Z$ parameter, given as the inverse of the Ohnesorge number ($Oh$), relates inertial and surface tension forces to viscous forces as:

$$Z = \frac{1}{Oh} = \frac{Re}{\sqrt{We}} = \frac{\sqrt{\rho \gamma L}}{\mu}$$
where $Re$ and $We$ are the Reynolds and Weber numbers, respectively. Generally, ideal droplet formation occurs when $Z$ is between 1 and 10 and the droplet velocity is at least equal to $\sqrt{4\gamma/\rho d}$ [20]. PolyJet and other inkjet-based printing methods have significant advantages due to their high resolution, sophisticated printhead designs (e.g., state-of-the-art multinozzle arrays can deliver more than 100 million drops per second), and multimaterial deposition capabilities [152]. However, the rheological properties of these inks must be carefully controlled and limit the overall material properties that can be printed. It is inherently difficult to inkjet complex fluids, such as concentrated polymer solutions or those that contain filler particles above 100 nm in diameter, without clogging or improper droplet ejection [20].

Unlike droplet-based methods, 3D filament printing allows for a broader range of ink designs, feature sizes, and geometries [153, 154]. In this approach, a viscoelastic ink is deposited as a continuous filament in a layer-wise build sequence. FDM was the earliest embodiment of ink-based 3D printing [153]. In this method, thermoplastic filaments are fed through a hot extrusion head, printed, and solidified upon cooling below their glass-transition temperature (Figure 2.9D). Several types of thermoplastic filaments can be patterned by this approach, including acrylonitrile butadiene styrene (ABS), polylactic acid (PLA), and thermoplastic polyurethane (TPU). These polymer filaments can also be filled with particles, such as carbon black, to enhance their functionality [155].

Direct ink writing (DIW) of viscoelastic materials under ambient conditions has emerged as an important alternative to FDM printing (Figure 2.9E) [154]. Several concentrated polymeric [156–160], fugitive organic (i.e., used as sacrificial materials) [18, 161], and filled epoxy [162] inks have been developed for printing complex 3D architectures. These inks are typically yield-stress fluids that follow the Herschel-Bulkley model [163]:

$$\tau = \tau_y + K\dot{\gamma}^n$$

where $\tau$ is the shear stress, $\tau_y$ is the shear yield stress, $K$ is the consistency, $\dot{\gamma}$ is the shear rate, and $n$ is the flow index ($n < 1$ for shear-thinning fluids). To enable printing, the applied stress within the printhead
must exceed \( \tau_y \) of these inks, enabling their flow through the nozzles. Notably, these inks rapidly recover their requisite mechanical properties, including a sufficiently high \( \tau_y \) and elastic shear modulus, \( G' \), upon exiting the nozzle [154]. In many cases, additional processing steps, e.g., photopolymerization or thermal curing, are required to fully solidify the printed parts. When these steps are decoupled from the printing process, it is challenging to build truly 3D objects, as the underlying printed layers may not fully support subsequent layers. However, it is possible to overcome these limitations by implementing printheads coupled with UV LEDs [164, 165] or heated build chambers.

The complexity and functional performance of 3D printed objects can be enhanced through co-printing multiple materials with a high degree of spatial and compositional precision [166]. Inevitably, multi-material 3D printing methods will be essential to new innovations in soft robotics. Compared to light-based printing methods, ink-based printing approaches like PolyJet, FDM, and DIW readily allow multi-material 3D printing (Figure 2.11). PolyJet and FDM are both capable of printing primary build materials alongside sacrificial materials that support overhanging or spanning features (Figure 2.11A). PolyJet enables voxel-by-voxel patterning of multiple materials, including photopolymer resins whose backbone composition, side group chemistry, and cross-link density can be systematically varied to produce regions with disparate mechanical properties at a higher resolution than FDM printing (Figure 2.11B). However, DIW [154] currently offers the broadest spectrum of printable materials, including structural [158, 159, 162], electrical [19, 167], optical [164, 168] and biological [169].

Multimaterial DIW is achieved either by using multiple (single nozzle) printheads (Figure 2.11C), each of which houses a different ink composition [169] or microfluidic printheads that allow for switching [170], mixing [171], core-shell printing [164, 167], or printing multiple filament arrays in a single pass [172]. Microfluidic switching nozzles can change between two different inks on the fly [170], whereas mixing nozzles can be used to print materials with tunable gradients of mechanical, conductive, or other material properties [171]. Core-shell printheads yield filaments that contain concentrically layered
Figure 2.11. Multi-material 3D printing with fused deposition modeling (FDM), Polyjet, and direct ink writing (DIW). (A) A FDM printed Hilbert cube before (behind) and after (in front) removal of print support material. (B) An art piece by Neri Oxman (MIT Media Lab) printed with Polyjet (scale bar is 10 mm). The inset provides a closer view of the voxel-level control of material heterogeneity afforded by Polyjet. (C) An elastomeric scaffold printed via multimaterial DIW using four silicone inks (scale bar is 2 mm, adapted from Ref. 169). (D) Vascularization of synthetic hydrogels via omnidirectional DIW, the first embodiment of embedded 3D printing. Embedded 3D printing is a free-form, multi-material DIW method that involves (i) the direct extrusion of viscoelastic inks into rheologically tailored matrix materials. In this work, a Newtonian filler fluid with similar composition to the matrix material was added to prevent crevice formation behind the print nozzle. (ii) After printing a fugitive ink (red features) into a photocrosslinkable hydrogel-based matrix, (iii) the matrix is photocrosslinked with UV, the fugitive ink is removed (step not shown), and (iv) a hydrogel with an embedded microvascular network is produced (scale bar in the photograph is 10 mm, adapted from Ref. 18). (A-C adapted from Ref. 20.)

Finally, multinozzle printheads separate a single ink stream into $2^n$ streams, where $n$ defines the number of bifurcating generations within the printhead, allowing one to dramatically reduce the build times [172]. Finally, there is growing interest in directly writing inks within matrix materials via a process known as embedded 3D printing, which enables truly freeform fabrication of soft materials (Figure 2.11D) [18, 19, 173]. These variants of DIW provide unparalleled flexibility in the types and motifs of soft matter forms that can be adapted for soft robotic fabrication.
PolyJet and other multimaterial inkjet printing methods are the predominant ink-based 3D printing method used to create soft robots. With these methods, complex FEAs with bellowed designs, intricate fluidic mechanisms (Figure 2.12A) [174] and appropriate mechanics for traversing treacherous, unstructured terrains (Figure 2.12B) [175] have been printed. PolyJet has also been used to create bodies for jumping soft robots powered by combustion. These bodies are printed with a graded elastic modulus (~1MPa to ~1GPa) by patterning layers composed of multiple photopolymers of varying compliance. This design improved the interface between the robot’s body and the on-board hardware needed for propulsion [176]. Again, the commercial, photopolymer inks used in these printing methods suffer from many mechanical limitations, including creep, long-term performance, and high-cost. These materials also tend to possess a higher stiffness and lower extensibility than materials used in molded FEAs.

There have been some preliminary efforts in fabricating FEAs with ink filament-based 3D printing approaches. FDM printing with commercially available TPU thermoplastics have used to print FEAs capable of generating high forces (Figure 2.12C) [177]. However, these thermoplastics exhibit notably high elastic moduli on the order of 1-10 MPa and poor mechanical performance due to creep. DIW-based methods have produced FEAs with integrated sensors that serve as simple haptic feedback devices [178]. Some preliminary silicone ink designs have enabled some progress in larger DIW-printed FEAs, through designing silicone inks for DIW often compromises their mechanical properties post printing. As such, these inks show low viscosity and stiffness that result in poor print resolution and void-spanning capabilities necessary for complex fluidic architectures (Figure 2.12D). Heated build platforms are required to crosslink printed inks and provide mechanical stability for larger builds [179].

Overall, while these initial demonstrations reveal the power of digital design and additive manufacturing for soft robot fabrication, significant research remains to develop new compatible materials systems and printing methods that truly optimize soft actuator mechanics. Further advances in
Figure 2.12. Examples of soft robots 3D printed via ink-based methods. (A) A fully printed soft robot with FEA-inspired bellows for actuation. The bellows are filled with fluids patterned during the multimaterial, Polyjet-like printing process (adapted from Ref. 174). (B) Polyjet printed, soft robotic actuators enable locomotion over unstructured terrains. (i) The printed FEA includes three chambers that can be individually addressed for multi-degree of freedom actuation. (ii) Four FEAs are integrated into a complete system. (iii) Various actuation schemes enable rotation and translation (adapted from Ref. 175). (C) FDM printed, (i) FEA assemblies capable of high-force actuation. (ii) The gripper can grab objects of various masses. (iii) The maximum payload is approximately 5 kg (adapted from Ref. 177). (D) 3D printing soft robotic FEAs via DIW. (i) In this example, the ink formulations required to 3D print silicones are not mechanically robust. A heated build platform is required to crosslink the builds after printing. Crosslinking facilitates the printing of tall structures (*). The ink (**)) is shown after extrusion to the right. (ii, iii) A DIW-printed soft robot with five FEAs. Photographs show inflation of the front right (ii) and front left (iii) actuators (adapted from Ref. 179).

multimaterial printing, coupled with new resin and ink formulations would enable soft robotic systems with advanced architectural complexity and function.
2.2.3 Power and Control for Soft Robotic Systems

FEAs and EPAs are actuated via inflation with pressurized working fluids and applied electric fields, respectively, that are readily generated and controlled by conventional robotic hardware. By contrast, supplying necessary stimuli for actuators based on swelling/hygromorphic materials, heat-, light- or magnetically-responsive matter, or engineered tissue constructs is far more challenging, hence, the vast popularity of FEAs and EPAs in soft robotics. Below, we discuss power and control strategies for soft robots that employ these types of actuators.

2.2.3.1 Powering Soft Robots

FEAs can be inflated with either liquids or compressible gases, and numerous methods for providing these working fluids at the required operating pressures have been demonstrated. As FEA inflation with pressurized liquids requires large pumping equipment, gases remain the more popular working fluid for FEA actuation. Pressurized gas has been supplied by regulated gas release from air compressors (Figure 2.13) [16, 92, 180], compressed fluid cylinders (including compressed air and CO\textsubscript{2}) [5, 15, 22], hydrogen gas release from heated lanthanum mischmetal alloys [78], methane combustion [176, 181], catalytic decomposition [182], and hydrolysis [183]. Recently, liquid monopropellant fuels have been explored as power alternatives for soft robots to alleviate some of these issues. Hydrogen peroxide, for example, is a compelling fuel choice for soft robotics, as it provides an extremely high energy density (up to 2.7kJ/g) and only requires exposure to a catalyst to decompose into pressurized gas that subsequently drives actuation [22]. Negative pressures, as recent demonstrations have shown [105, 184], can also be used for contractile actuation of certain FEAs, but generating a vacuum for soft robot actuation, particularly in deployable, untethered systems, is far less straightforward.

Voltage requirements for actuating EPAs are highly composition and geometry dependent. For DEA, actuation voltages of approximately 100 V per micron of dielectric membrane thickness are
required [12]. Manufacturing limitations mean that DEAs need hardware for generating high voltage sources for actuation. Until appropriate voltage amplifiers, processing, and materials challenges can be addressed, DEAs will remain more appropriate for soft robots tethered to large power supplies [6, 12, 13, 25, 28–31]. Current IPMCs only need several volts (approximately 1 – 10V) for actuation, however these devices are still limited to small form factors [13]. Finally, while they are not discussed further, we note that LCEAs could also be thermally actuated and controlled with this same robotic hardware if appropriate heating elements, such as Joule heating elements, are integrated into these devices. Some recent progress [185–188] highlights the potential use of advanced, digitally actuable LCEA composites in soft robotics.

2.2.3.2 Design of Autonomous, Untethered Soft Robots

Soft robotic systems are still dependent on traditional, rigid hardware for power and control. Consequently, the field has been left with many obstacles towards the design of autonomous, untethered soft robots (Figure 2.14). These heavy hardware components are a bulky payload for soft robots to carry and are difficult for soft robot bodies to interface with mechanically [176, 189]. Autonomous, untethered systems have been achieved in FEA- [15, 16, 176, 182], DEA- [17], and IPMCs-based [31] soft robots. For
**Figure 2.14. Examples of untethered, autonomous soft robots.** (A) An untethered, soft robotic fish. (i) All hardware is encapsulated within the robot’s body. (ii, iii) The soft robot’s tail is a bimorph FEA that enables rapid, biomimetic swimming maneuvers (adapted from Ref. 15). (B) An untethered, soft robotic quadruped. (i) The overall system is large. (ii) Five FEAs are integrated into the device (shown in non-inflated and inflated states in the left and center images). All hardware is packaged in the center of the robot (right image) (adapted from Ref. 16). (C) An untethered jumping soft robot powered by combustion. (i) The robot’s 3D printed body has a graded modulus from approximately 1 MPa at the bottom of the device to 1 GPa at the interface with on-board hardware. (ii) Integrated FEAs act as legs that position the soft robot for jumping. (iii) The soft robot can jump and land without damage (adapted from Ref. 176). (D) An untethered, soft robotic swimmer powered by DEAs. (i) Schematic of the soft robotic system. (ii) The DEA (labeled in this panel as a “DE muscle”) can actuate to drive fin propulsion. (iii) All power and hardware components are brought on board in the “Epod.” An integrated voltage amplifier is needed for DEA actuation. (iv) The device is capable of steered swimming underwater (adapted from Ref. 17).

The FEA-based robots, swimming (Figure 2.14A) and crawling (Figure 2.14B) systems have been achieved [15, 16]. In the latter demonstration, notably large FEAs were required to successfully carry the hardware [16]. Alternatively, terrestrial jumping systems with smaller form factors than the crawling system were made possible by power via combustion reactions (Figure 2.14C) [176]. DEA-based, untethered systems have only been achieved in swimming soft robots (Figure 2.14D) [17]. Finally, one untethered IPMC-based soft robot has been developed [31], but autonomous soft robots utilizing these actuators are still in
early development. Overall, hardware requirements practically render these untethered soft robots as hybrid-rigid soft systems at best. From many perspectives, this hardware denies soft robots the full benefits and virtues of being soft.

2.2.3.3 Soft Robot Control

Several intriguing ideas have been proposed regarding how to best control soft robots [2]. For example, some researchers have proposed using microfluidic controllers [190] as well as harnessing various mechanical dynamics or deformations, such as snap-through instabilities induced by compressive buckling [191], as exploitable sensory and morphological computational mechanisms in soft robotics [2, 192–194]. However, prevailing success with FEA- and EPA-based soft actuators has convinced many that integrating electronically compatible sensors is still the most straightforward strategy for achieving soft robotic sensing [14].

While some initial efforts have aimed to incorporate flex sensors into soft actuators [17, 132, 195], the rigid nature of conventional sensors and electronic components (e.g., metallic strain gauges, bend and force sensors, Hall effect magnetic sensors, and more) are not ideal for soft robots. Instead, flexible, deformable sensors and electronics based on soft conductive matter, as discussed later in Section 2.2.4, are more appropriate [2, 14, 189]. Unfortunately, closing the control loop in soft robotics remains a grand challenge for the field, as fabricating soft actuators with soft conductive features for sensing is not easily achieved with available design and fabrication methods. Consequently, most soft robots to date employ an open-loop control architecture.

2.2.4 Soft Sensors

Recent progress in the field of soft electronics has led to flexible electronic devices and sensors for myriad applications, including wearable technologies [196], implantable devices [197], and artificial skin
Like traditional robots, soft robots require sensory capabilities that provide proprioceptive, kinesthetic, tactile, exteroceptive, and other modes of feedback for system control. Hence, there is considerable interest in translating decades of innovation in the field of soft electronics to soft robotic applications. To date, soft electronics have relied on creating stretchable devices either through (i) the structural engineering of otherwise rigid, non-stretchable materials (e.g., gold, silicon, and organic semiconductors) into flexible architectures [201, 202] or (ii) the design of composite materials where rigid, non-stretchable components are embedded throughout a stretchable matrix or substrate [203]. These devices require complicated, multi-step, and expensive microfabrication techniques that are not always compatible or easily integrated with soft actuator fabrication methods. Alternatively, intrinsically deformable, conductive soft components have also been explored, including conductive particle-polymer composites, liquid metals, and ionic liquids and gels. These materials are patterned onto, attached to, or encapsulated within an insulating stretchable material [7–10]. Recently, deformable optical sensors have emerged as an alternate strategy.

2.2.4.1 Electrically Conductive Composites

By filling flexible, deformable polymeric networks with electrically conductive particles, percolated conduction paths can be established [204]. These composites are effectively piezoresistive, increasing in resistance, $R$, under deformation (where $R = \rho L/A$, for a composite with resistivity $\rho$, length $L$, and cross-sectional area $A$) (Figure 2.15A). Many types of soft sensors have been developed from conductive particle-polymer composites for strain, pressure, and curvature sensing. The gauge factor, $GF$, of simple strain sensors composed of these composites is given by:

$$GF = \frac{\Delta R / R_0}{\epsilon}$$

where $R_0$ is the initial sensor resistance, and $\epsilon$ is the strain. Particle composition, morphology, concentration, and functional surface chemistry influence the electrical, mechanical, and optical
Figure 2.15. **Soft sensors based on electrically conductive composites.** (A) Schematic overview of the operational principle of soft sensors based on conductive composites (adapted from Ref. 216). (B) An example of a conductive particle-polymer composite used in soft sensors. (i) Overview of the conductive ink formulation. The photographs show the final ink (upper right) and patterned ink features (lower right). (ii) A stretchable composite made with the silver ink undergoing strain to 260%. (iii) Initial conductivity and stretchability as a function of silver flake content. (iv) The ratio of resistance ($R$) to initial value ($R_0$) as a function of strain cycles for strains of 10%, 33%, 50%, and 70% (adapted from Ref. 220).

Properties of these composites. In general, for a single filler, the resulting electric conductivity, $\sigma$, given by:

$$\sigma = \sigma_0 (V_f - V_c)^s$$

where $\sigma = 1/\rho$, $\sigma_0$ is the bulk conductivity of the filler, $V_f$ is the volume fraction of the filler, $V_c$ is the critical volume fraction at the onset of percolation, and $s$ is a power law exponent. Noble metal [205–209] and carbon [204, 210–217] fillers are commonly used to achieve a broad range of electrical and mechanical functionalities. Conductive nanowires, nanotubes, and other 1D particles with high aspect ratio dimensions tend to offer several advantages, namely ease of mobility during network deformation and relaxation. Large gauge factors (~100-500) have been achieved with composites whose filler concentrations are near the filler percolation threshold. In these instances, contributions from electron tunneling between filler particles more greatly influence the observed resistance change [218, 219].
A representative conductive composite design for soft sensor applications [220] is featured in Figure 2.15B. In this example, silver flakes are added to a fluorine rubber solution (DAIJIN Daiel-G801 in the volatile solvent 4-methyl-2-pentanone). An added surfactant (Zonyl FS-300) improves the dispersion of the silver flakes throughout the mixture. The final printable ink can be patterned with other elastomeric matrices (Figure 2.15Bii) for highly stretchable soft sensors. Figure 2.15Biii shows the influence of silver flake content composite conductivity and extensibility, and Figure 2.15Biv provides the ratio of resistance over initial resistance as a function of strain cycles at several different strains for a representative conductive composite formulation. Even for moderate strains of 10%, a clear change in composite conductance results from the rearrangement of the silver filler particles with repeated deformation [220].

Many processing methods have been developed for producing conductive particle-polymer composite sensors, including transfer printing, coating, solution-casting, filtration, and in situ conductor growth techniques [196]. These methods are often highly specific to the particular filler and polymer matrix used. Unlike other soft conductive materials, conductive composites can be tailored for use as inks in various printing methods, including direct ink writing. Figure 2.16 highlights two recent examples of designing conductive composites as inks for 3D printed soft sensors [19, 221]. In the first example, soft sensors were fabricated using embedded 3D printing. Specifically, a conductive carbon grease was directly extruded into uncrosslinked, elastomeric matrix materials, which could be cured after printing, to seamlessly fabricate soft, highly extensible strain sensors (Figure 2.16Ai). By simply changing print speed, the cross-sectional area of the ink filament – and, in turn, the resistance of the strain sensor – could be tuned on-the-fly (Figure 2.16Aii). Mold designs could easily be changed to print inexpensive devices, such as sensorized gloves, with embedded strain sensors (Figure 2.16Aiii) capable of providing resistive feedback (Figure 2.16Aiv). The free-form nature of embedded 3D printing enabled complex sensor architectures to be rapidly printed [19]. Finally, a new “hybrid 3D” printing technique combining DIW
Figure 2.16. 3D printed soft sensors based on electrically conductive composite inks. (A) Soft strain sensors via embedded 3D printing. (i) A photograph of a strain sensor being printed. (ii) Strain sensor resistance changes with print speed. (iii) A printed glove with strain sensors at each joint. (iv) The resistance of each soft strain sensor changes with finger bending. (v) A complex, printed soft sensor before (top) and after (bottom) stretching (adapted from Ref. 19). (B) Hybrid 3D printing, of soft, deformable electronic systems. This technique combines DIW and pick-and-place assembly. (i, ii) Overview of hybrid 3D printed (i) soft sensor arrays and (ii) complete devices with pick-and-placed components. (iii, iv) Soft strain sensors can be printed that (iv) change resistance with strain. (v, vi) Capacitive pressure sensors (v) can be printed with the conductive and soft, packaging inks and (vi) increase in capacitance with applied pressures (adapted from Ref. 221).

and pick-and-place assembly recently enabled the programmable assembly of low-cost, soft electronic systems with arbitrarily customizable form factors and functionalities (Figure 2.16Bi, 2.16Bii). Central to this advance was the development of a silver-TPU composite ink that served as a printable, soft conductive feature for printed sensors or interconnects with other electronic components that could be pick-and-placed into the hybrid 3D printed devices. Both resistive strain sensors (Figure 2.16Biii, 2.16Biv) and pressure-sensitive capacitive sensors (Figure 2.16Bv, 2.16Bvi) could printed with this and another TPU packaging ink [221]. As illustrated in Figure 2.15Biv, the key challenges that emerge for this class of soft sensors stem from the structural dynamics associated with their percolated conduction path, which can manifest as deleterious hysteresis effects for resistive sensors, leading to unreliable, variable sensing [19, 220–222]. Many conductive particle-polymer composites are not appropriate for large deformation
resistive sensing. Designing strain- and pressure-sensitive capacitive sensors with conductive particle-polymer composites is one approach many have taken to avoid these challenges [217, 221]. Moreover, filler addition greatly influences the composite’s mechanical properties, reducing extensibility and flexibility for stiffer fillers and higher filler concentrations [220–222].

2.2.4.2 Liquid Metals

Liquid metals have received significant attention for use as soft, intrinsically deformable conductors. As a highly conductive liquid continuum, they alleviate many challenges associated with conductive particle-polymer composites, particularly conduction hysteresis. Gallium and its alloys, especially Galinstan (GaInSn) and eutectic gallium indium (eGaIn), have been widely used in soft electronics [223]. These alloys possess a viscosity approximately twice that of water, negligible vapor pressure, an electrical conductivity about an order of magnitude less than copper (~3×10^6 S/m), and large surface tension (>500 mN/m). Under ambient conditions, gallium alloys react with oxygen and become passivated by a sub-nanometer thick gallium oxide layer that mechanically stabilizes the liquid [223, 224]. These properties have proved useful for patterning liquid metal conductors by injection into molded cavities, masked deposition, casting, and contact printing methods [225].

A prevailing technique for assembling liquid metal-based sensors is illustrated in Figures 2.17A-B [226]. Using the same multi-step molding and lamination techniques for FEAs (Figure 2.17A), stretchable matrices can be fabricated with embedded micro-channels that can be in-filled with a liquid metal (Figure 2.17B). Deformation of these networks results in a hysteresis-free change in resistance or capacitance, depending on sensor design. Several devices have been developed using these techniques, including highly deformable strain- and pressure-sensitive resistive Figure 2.17C-E [226] and capacitive [227] sensors, curvature sensors for proprioceptive feedback (Figure 2.17F) [228], antennas [229], diodes [230], and more [223, 224]. Alternatively, eGaIn can be directly written in 2D motifs, due to stability from the
Figure 2.17. **Soft sensors based on liquid metals.** (A-E) Design and performance of a soft eGaIn sensor sensitive to pressures and strain. (A-B) Typical fabrication protocol. (A) The process involves the molding and laminating of elastomer layers to create a network of embedded microchannels. (B) The eGaIn can be injected into the channels, as shown in the photograph time series. (C) The final soft sensor with sensitivity to pressure and strain deformations. (D) The elastomeric matrix renders the device highly deformable. (E) Representative readouts are illustrated on the computer screen during applied pressures and strains on the device (figures adapted from Ref. 226). (F) Similar techniques can be used to create (i, ii) devices with embedded liquid metal and void features that (iii, iv) provide curvature sensitivity (figures adapted from Ref. 228). (G) Bulk processing of conductive elastomer-liquid metal composites. (i) Liquid metal droplets can be dispersed in a stretchable elastomer matrix. (ii) Optical micrograph and (iii) micro computed tomography CT views of the dispersed liquid metal droplets. Continuity between the droplets is established after “break-in” cycles. (iv) Photographs of the composites undergoing several hundreds of percent strain (figures adapted from Ref. 232).

oxide layer, and encapsulated in an elastomeric matrix for soft sensor assembly [231]. Finally, akin to the conductive particle-polymer composites discussed previously, eGaIn inclusions have also been introduced in stretchable silicone elastomers for various sensing and thermal management applications in soft robotics (Figure 2.17G) [232–234].

Although liquid metals offer a straightforward solution to intrinsically deformable soft conductors, their material properties pose several challenges. First, their high electrical conductivity is not
always appropriate for sensor applications, where moderate resistances are required. Fabricating liquid metal-based soft sensors is a tedious, low throughput process. It is difficult to create small features, especially when injection is required. Second, the toxicity of gallium alloy-based liquid metals is still not fully understood. Third, gallium oxide layers stabilizing Galinstan and eGaIn can be displaced during large sensor deformations, occluding conduction paths and rendering sensor failure [235]. The liquid nature of these conductors also presents challenges for mechanical interfacing with rigid leads to external readout electronics. Finally, the high surface tension and rapid oxide layer formation can present challenges when printing liquid metal-based sensors via extrusion, inkjet printing [236], or 3D printing methods [225].

2.2.4.3 Ionically Conductive Liquids and Gels

Soft sensors have been designed and fabricated with ionically conductive materials, such as polyelectrolyte solutions, hydrogels, and organic ionic liquids [11, 237, 238]. In addition to resistive [239–243] and capacitive [167, 178, 238, 244] mechanical sensing, soft ionically conductive sensors have been developed for humidity, temperature, pH, and other biological sensing applications [35, 245]. Compared to the aforementioned soft conductors, these materials generally exhibit low toxicity, biocompatibility, tissue-like mechanical properties, and optical transparency. They are also typically cheaper than their nanoparticle polymer composite- and liquid metal-based analogs [245].

Most soft ionic conductors are aqueous in nature. Aqueous-based sensors exhibit poor long-term performance due to water evaporation from the hydrogel network or through encapsulating, gas permeable matrices [238, 246]. Through the addition of glycerol or use of different salts, evaporation can be mitigated [74, 238, 244]. However, water loss is inevitable at elevated temperatures. Aqueous ionic conductors also possess low conductivities (on the order of 1 mS/cm, about six orders of magnitude higher than eGaIn) that are temperature dependent. DC readout electronics are not appropriate for these
conductors, as capacitive charge separation can result in drift. Low operating voltages are also typically required to avoid electrochemical side reactions and hydrolysis. Organic ionic liquids (ILs) have recently been explored as an alternative to aqueous ionic conductors [237]. ILs have previously been used as soft sensors due to their low vapor pressure, non-permeability through elastomeric matrices [247], low hysteresis [243], and appropriate resistivity for sensor applications [240, 241] (i.e., slightly lower than aqueous ionic conductors). They can serve as an alternative dispersion medium to water for hydrogel networks [247]. Disadvantages of ILs include their toxicity concerns and high cost. Some ionic liquids show significantly reduced toxicity. However, advances in ionic liquid chemistries will not only improve their biocompatibility, but their functional and conductive performance as well [237].

By stacking and laminating ionically conductive hydrogel films in planar configurations separated by appropriate dielectric layers, transparent, wearable strain- and pressure-sensitive capacitive sensor arrays have been designed for ionic skin applications Figure 2.18A [238], simple FEAs [248], and deformable tactile interfaces [249]. With similar molding and lamination techniques used for gallium alloy-based sensors, ionic sensors been developed by injecting various ionic liquids into micro-channel networks embedded in elastomer matrices [239–242, 244, 247]. In several examples, both an ionic liquid and gallium alloy were used to create highly resistive strain sensors connected by highly conductive, liquid metal leads for wearable applications [239, 241]. One such glove is shown in Figure 2.18B with corresponding proprioceptive output from the wearer [241]. Ionic liquid injection in micro-patterned, embedded fluidic channels has also produced arrays of capacitive sensors for versatile tactile interfaces [244]. Core-shell printheads have been developed for DIW-based 3D printing of capacitive sensor fibers comprised of aqueous ionic conductors separated by highly stretchable elastomers (Figure 2.18C) [167]. Various hydrogel chemistries and processing methods can be used to improve the performance of these devices, including improved interfacing with encapsulating elastomer matrices [246], other conductive materials like poly(3,4-ethylenedioxythiophene) (PEDOT) (Figure 2.18D) [250], or deformable metal
Figure 2.18. Examples of ionically conductive soft sensors. (A) An example of an “ionic skin” device incorporating four capacitive sensors based on tough, stretchable hydrogels. (i) The device shown mounted on skin. (ii, iii) As illustrated by a similar device, the device is conformal and (iii) stretchable during finger bending. (iv) The sensor layout for the device in (i). (v) Corresponding readouts from a wearer pressing on sensors s #1 through #4 (adapted from Ref. 238). (B) An example of a wearable glove with integrated ionic strain sensors. (i) The resistive strain sensors are connected to more conductive liquid metal interconnects. (ii) The sensors provide proprioceptive feedback on finger bending (adapted from Ref. 241). (C) 3D printed, capacitive ionic sensors with a fiber design for textile integration. (i) A schematic and (ii) photograph of the multi-core-shell nozzle used to 3D print the sensors. (iii) After end caps are fixed to the printed fiber sensors, (iv) the devices can be integrated into textiles for biometric feedback (adapted from Ref. 167). (D) A biocompatible hydrogel device with integrated conductive polymer (PEDOT) features. (i) Overview of the fabrication process and (ii) demonstration of device performance before after autoclaving shows robustness of the PEDOT network (adapted from Ref. 250).

...inclusions for interfacing with off-the-shelf electronic components [251]. Finally, self-healing capabilities have also been integrated into these sensors through novel hydrogel chemistries [252].
2.2.4.4 Deformable Optical Sensors

Recent interest has focused on developing deformable optical sensors for soft robotic sensing. Two prevailing strategies involve the use of deformable waveguides or reflectance for sensing. In the former approach, deformation of elastomeric waveguides results in optical power loss that can be measured with photodetectors and correlated with imposed strains or pressures [253–255]. In the latter, light reflection against a deformable elastomeric membrane is used for high spatial resolution tactile sensing. Specifically, incident light reflected against the membrane while in contact with an object is collected with a camera. With appropriate algorithms, the images can be reconstructed into 3D mappings of contacted objects [256]. While important to highlight in this section, these sensors’ primary disadvantage to soft conductors is their need for conventional electronics hardware, like LEDs or other high-power light sources, photodetectors, and cameras, in close proximity to the elastomeric structures. Furthermore, these sensors rely on off-the-shelf components that can be difficult to co-assemble or directly integrate with soft robots. As such, they also possess limited form factors with minimal customization and design flexibility.

2.2.5 Progress in coupling soft robotic sensors and actuators

Unlike FEAs, DEAs and IPMCs are capable of innately providing capacitive feedback on the state of their actuation [13, 28, 117, 121, 136, 257], and simple closed-loop control demonstrations for these actuators have been reported [30, 121, 258]. However, the device forms and possible sensing motifs with these actuators are greatly limited by current manufacturing methods [259–261]. Alternatively, FEAs provide a more flexible platform for coupling soft robotic sensing and actuation. Several groups have explored the integration of sensors with simple FEAs through the use of conventional, rigid robotic sensors [132, 195, 262], as well as soft sensors based on conductive nanoparticle-polymer composites (Figure 2.19A) [263, 264], liquid metals (Figure 2.19B) [235, 265–267], and aqueous ionic conductors.
Figure 2.19. Examples of soft robotic actuators with integrated soft sensors. (A) The first example of a soft actuator with integrated sensors published in 2006 (Ref. 263). Using the same (i) three-chambered, (ii) tentacle-like actuators shown in Figure 2.3D, the authors (iii) placed fixed three displacement sensors to the exterior of the actuator. Their paper notes the deleterious side effects of dynamic restructuring of conductive composite-based soft sensors (figures adapted from Ref. 263). (B) FEAs with integrated (i) force and (ii) position sensors throughout (iii) the device. (iv) A soft gripper based on four sensorized FEAs is shown without eGaIn (figures adapted from Ref. 266). (C) A prototype of a DIW printed tactile actuator array with integrated soft, ionic capacitive sensors. (i) A user presses on the sensors, which report back the press pattern by inflating small FEA bladders. (ii) The capacitance readout from the sequence is provided as a function of time (figures adapted from Ref. 178). (D) Soft FEA-based gripper with integrated elastomeric waveguides for sensing. (i) One FEA device has three embedded waveguides. (ii) Five sensorized FEAs are integrated into a soft robotic hand. (iii) The authors show that this gripper is capable of various tasks, including biomimetic handling (left), fine tactile sensing of surface roughness (center), and mechanical softness (right) (adapted from Ref. 129).

(Figure 2.19C) [178, 248]. Few researchers have produced soft robotic actuators capable of receiving multiple modes of sensory feedback. Most notably, bending FEAs were recently prepared with multiple strain and pressure sensors for proprioceptive and tactile feedback via three integrated eGaIn soft sensors (Figure 2.19B) [266, 267] and three elastomeric waveguide-based sensors (Figure 2.19D) [129]. In the latter demonstration, the inclusion of rigid fingertips enabled soft robot hands to sense fine surface topologies
and object softness [129]. However, all these devices are difficult to fabricate, as they rely on low-throughput molding and manual assembly procedures. Current manufacturing methods clearly restrain the architecture, material composition, and in turn, overall actuator and sensor functionalities that are necessary for next-generation soft robots requiring closed-loop feedback control.

2.3 Applications

Owing to their soft, compliant nature and dynamic physicochemical properties, soft robots have tremendous potential in applications where robots with exceptionally complex locomotive abilities, high deformability, highly dynamic or biomimetic functionalities, abilities to adapt to undefined or unstructured environments, physical resilience, and – clearly – softness are required [1–3]. While soft robotic technologies have only just begun to be commercialized, many applications for soft robotics seem inevitable. Here, we highlight several emerging applications for soft robots, acknowledging that far more and yet-imagined innovations exist for this new class of robot.

Manipulation and locomotion in undefined environments remain canonical challenges in robotics [2]. Given their compliance, soft robotic actuators have been successfully used as robotic end effectors capable of easily grasping objects with complex or unknown shapes and gently handling delicate objects [2, 74, 80, 97, 104, 117, 268, 269]. Soft Robotics, Inc. (Somerville, MA, USA), for example, is developing soft robotic gripper for handing produce and other food products (Figure 2.20Ai), and others have developed soft grippers for handling animals and other living organisms underwater (Figure 2.20Aii) [268]. Humans can also more safely interact with robots possessing soft end effectors [2]. Finally, soft robotic actuators for locomotion are enabling robots to more easily traverse difficult terrains compared to traditional systems [16, 92, 98, 175], and swim underwater in bioinspired motifs. These collective capabilities have implications for myriad soft robotic systems, ranging from employment in automated manufacturing to personal robotics and security applications.
Figure 2.20. Applications of soft robots. (A) Examples of soft robotic end effectors. (i) A soft gripper from Soft Robotics, Inc. (Somerville, MA) gently picks up a loaf of bread (photo from Soft Robotics, Inc.). (ii) A soft gripper is used to collect fragile living specimen samples underwater (adapted from Ref. 268). (B) Examples of soft robotic systems for medical use. (i) Soft robotic actuators have been designed for replacing or augmenting muscle function in wearable exosuit devices. (ii) A wearable device uses several McKibben actuators (see arrows) for ankle-foot rehabilitation (adapted from Ref. 77). (iii) FEAs and optical strain sensors incorporated into a soft robotic glove orthosis (adapted from Ref. 133). (iv) A soft robotic “heart sleeve” for pumping assistance is shown placed over a porcine heart (adapted from Ref. 274). (C) Soft robots can possess unique abilities over traditional robots. (i) A transparent, hydrogel-FEA based fish swims underwater (adapted from Ref. 151). (ii) A snake-like, FEA-based robot navigates a maze (adapted from Ref. 103). (iii) A photo-responsive LCEA-based “caterpillar” robot can move autonomously (adapted from Ref. 53). Such devices point to a future where soft material composites with robotic capabilities can enable the design of small, hardware-free robots.

The inherent mechanical similarities between the materials comprising soft robots and our own bodies also suggest that soft robots will pave the way for safer, more comfortable, and even more
biocompatible technologies at the human-robot interface. Early work in soft robotics focused on the
development of soft micromanipulators and other prototype devices for surgical applications [39, 85, 86, 88]. More recently, soft robotic actuators acting as artificial muscles (Figure 2.20Bi) have been used in various wearable exosuit (Figure 2.20Bii) [76, 77, 270] and orthotic systems for rehabilitation (Figure 2.20Biii) [2, 133]. Wearable technologies employing soft robotic actuators have clear promise in wearable devices used, for instance, in augmented/virtual reality or smart textiles [271]. New soft tactile interfaces will also be possible with soft robotics [272, 273]. Preliminary work has also shown that soft robotic systems may be useful in implantable devices where artificial muscles are required (Figure 2.20Biv), though much more work is required for these applications [274].

Lastly, the material nature of soft robots points to numerous capabilities that simply cannot be possessed by traditional rigid robots. For example, as advances in soft robotic materials and fabrication methods progress, advances in transparent robots have been made (Figure 2.20Ci) [151]. Soft robots that can deform, reconfigure, and possess complex locomotion motifs have been designed for navigating difficult environments and tight spaces (Figure 2.20Cii) [92, 103]. Soft robotic components for dynamic camouflage [275], display capabilities [248], and stiffness [276–278] have been reported, and new materials and strategies for resilient [16, 130, 176, 181], self-healing [93, 150], repairable [279], biodegradable [280], and even edible [281] soft robots have all been developed. Soft robots will, as a result, be well suited for performance in underwater, extraterrestrial, outer space, physiological, and other harsh environments for traditional robots. Finally, if soft analogs can be substituted for power and control hardware [282] and appropriate fabrication strategies are developed, robots with unprecedentedly small form factors may be achievable. This vision is illustrated by the photoresponsive, LCEA inch-worm shown resting on a human finger tip in Figure 2.20Ciii [53].
2.4 Conclusions

The field of soft robotics has emerged as a result of the convergence of decades of progress in soft matter science, soft actuator design, digital and additive manufacturing, and soft electronics. However, while they stand poised to shape the future of robotics, soft robots remain dependent on conventional, rigid hardware for power and control that denies them the full benefits of being soft. Moreover, it remains difficult to sensorize soft robots with available fabrication methods, leaving soft robotic control loops open. Soft robots derive their unique robotic capabilities from the soft materials comprising them. Hence, creating new soft robots with minimal tethers to traditional hardware and integrated sensory capabilities requires new design paradigms. Towards these goals, new materials and manufacturing strategies are required that allow for the design of soft materials with embodied actuation, sensing, and computational functionalities.
This chapter has been partially adapted from a journal article (1) and a submission-ready manuscript (2):


3.1 INTRODUCTION

Recent advances in soft robotics motivate the need for new fabrication strategies that enable programmable integration of soft matter with disparate mechanical, electrical, and/or chemical properties in arbitrary form factors. To date, most soft robotic systems consist of molded-and-laminated fluidic elastomer actuators (FEA) with embedded fluidic networks [2, 3]. These elastomeric composites actuate when their interconnected pneumatic channels are inflated with gases or incompressible fluids. Unlike other actuators, FEAs are inexpensive to make, simple to operate, and actuate rapidly and reversibly [74, 92, 96, 97]. There are, however, intrinsic challenges in powering, embedding sensors within, and controlling soft robotic systems which hinder progress towards more sophisticated soft robots [2, 3].

3D printing methods, particularly direct-ink writing (DIW), offer several advantages for multi-material fabrication of soft robotic systems with embedded functionality [20]. For example, embedded 3D (EMB3D) printing is rapidly emerging as a novel assembly platform. In this approach, soft functional materials, known as “inks,” are printed in a truly freeform fashion within rheologically tailored, matrix-based reservoirs [18–20, 173, 283]. To date, EMB3D printing has been used to fabricate vascularized hydrogels [18], highly stretchable strain sensors [19], intricate 3D architectures [173], engineered tissue
constructs [283], and, as reported in Chapters 4 and 5, soft robots that exhibit autonomous motion and somatosensory feedback.

The development of functional inks and matrix materials for EMB3D printing has largely been done empirically [18, 19]. It is well known that both the inks and matrix materials should be shear thinning, yield-stress fluids, as described by the Herschel-Bulkley model:

\[
\tau = \tau_y + \eta(\dot{\gamma}) \cdot \dot{\gamma} = \tau_y + K\dot{\gamma}^n \quad \text{for } \tau \geq \tau_y, \\
\dot{\gamma} = 0 \quad \text{for } \tau \leq \tau_y
\]

where \(\tau\) is the shear stress, \(\tau_y\) is the shear yield stress, \(\dot{\gamma}\) is the shear rate, \(\eta\) is the \(\dot{\gamma}\)-dependent apparent viscosity, \(K\) is the consistency, and \(n\) is the flow index (where \(n < 1\) for a shear-thinning fluid). This behavior ensures that functional inks can be readily deposited through fine deposition nozzles and retain their filamentary form. It also facilitates nozzle translation through the matrix during EMB3D printing, while supporting the printed filaments. Both the ink’s shear yield stress, \(\tau_{y,\text{ink}}\), and shear elastic or plateau storage modulus, \(G'_{0,\text{ink}}\), should be on the order of magnitude higher than the matrix material’s shear yield stress, \(\tau_{y,\text{matrix}}\), and shear elastic modulus, \(G'_{0,\text{matrix}}\), respectively. If these conditions are not met, the “print fidelity” will be adversely affected. For example, when \(G'_{0,\text{ink}}\) is too large compared to \(G'_{0,\text{matrix}}\), the translating nozzle can drag the ink within the matrix material. In contrast, if \(G'_{0,\text{ink}}\) is too low with respect to \(G'_{0,\text{matrix}}\), the ink filament can bead up and break apart. If \(\tau_{y,\text{matrix}}\) is too low compared to the viscous stresses associated with nozzle displacement, the translating nozzle will create stress fields that yield and displace the printed features. When, \(\tau_{y,\text{matrix}}\) is too large, deleterious voids or crevices may form within the matrix as the nozzle translates during printing. This necessitates the use of “filler fluids” that possess a low viscosity and thereby infill these defects when placed on top of the matrix reservoir. In general, filler fluids are not only inconvenient to use, but they also introduce compositional heterogeneities to the final part.
A simple dimensional argument suggests that crevices of a depth $h$ will form behind print nozzles traveling through a matrix material of density $\rho$ and yield stress $\tau_{y,\text{matrix}}$ when:

$$\rho gh = \tau_{y,\text{matrix}}$$

where $g$ is the acceleration due to gravity, and $\rho gh$ is the hydrostatic pressure associated with the crevice, $\Delta P$ [173]. Hence, the matrix material will infill until the point $\Delta P$ for some crevice depth $h$ cannot overcome $\tau_{y,\text{matrix}}$, resulting in a crevice in the wake of the print nozzle. While this expression clearly neglects many factors that may contribute to or even mitigate crevice formation – including the matrix material’s non-Newtonian rheology and flow around the nozzle, drag forces acting on the nozzle, and any interfacial contributions from nozzle wetting by the matrix – it does illustrate that $\tau_{y,\text{matrix}}$ should be sufficiently low to mitigate crevice formation, while still supporting the patterned ink within the matrix.

The design of thixotropic yield-stress matrices that possess strain history-dependent, dynamic rheological behavior [284] are one straightforward option for meeting the above for filler fluid-free EMB3D printing (Figure 3.1). This rheological behavior is typically given by a modified, yield-stress fluid model:

$$\tau = \tau_y(\lambda) + \eta(\lambda, \dot{\gamma}) \cdot \dot{\gamma}$$

where $\lambda$ is a dimensionless time- and shear rate-dependent structural parameter, and $\tau_y(\lambda)$ and $\eta(\lambda, \dot{\gamma})$ are $\lambda$-dependent yield stress and apparent viscosity, respectively [284–286]. Many models exist for
thixotropic yield stress fluids and $\lambda$ as described by Equation (3), with most assuming $\lambda$ has values of $0 \leq \lambda(t) \leq 1$ as described by:

$$\frac{d\lambda}{dt} = f(\lambda, \dot{\gamma}) - g(\lambda, \dot{\gamma})$$  \hspace{1cm} (4)

where $f$ and $g$ are functions defining the fluid’s rates of reconstruction (i.e., aging or build-up) and disruption (i.e., deformation or break-down), respectively [285]. The parameter $\lambda$ provides insight to the fluid’s microstructure: $\lambda = 0$ for a completely yielded fluid, and $\lambda = 1$ for a completely structured fluid. One popular thixotropic, yield-stress fluid model is the modified Herschel-Bulkley equation proposed by Tiu and Boger [287]:

$$\tau = \lambda \tau_{y,0} + \lambda K_{y} \dot{\gamma}^n$$  \hspace{1cm} (5)

By tuning the thixotropy of a matrix material such that $\lambda \tau_{y,0} \rightarrow 0$ after matrix deformation and reduction of $\lambda$ via nozzle translation, it follows from Equation (2) that crevice formation would be suppressed in a model thixotropic matrix. Assuming $\lambda$ returns to unity with time, the matrix could fully restructure for future printing of functional features within those regions.

In this chapter, we design and print complex architectures with a soft materials palette appropriate for filler fluid-free, EMB3D printing (Figure 3.1). We first rheologically optimize a model thixotropic matrix material. The matrix material candidates are formulated from fumed silica nanoparticle-filled poly(dimethyl siloxane) (PDMS) blends. Using a model fugitive ink required for patterning fluidic features within the silicone-based matrices, we explore how matrix rheology (as tailored through fumed silica content), nozzle diameter, print speed, and print path selection contribute to overall print fidelity and matrix yielding around translating print nozzles. Our experiments, carried out in collaboration with Prof. Howard Stone’s Complex Fluids Group at Princeton University provide a fundamental understanding of the effects of matrix rheology on EMB3D printing.
3.2 EXPERIMENTAL METHODS

3.2.1 Matrix and Ink Preparation

Model matrices are prepared by blending two commercial PDMS materials, Sylgard 184 (used at 10:1 ratio of base to catalyst) and SE 1700 (used at 4:1 ratio of base to catalyst) (Dow Corning, USA) at various ratios in a planetary mixer at 2000 rpm for 3 min with degassing at 2200 rpm for 2 min. The model ink for the printing demonstration is prepared from an aqueous suspension of Pluronic F127 (Pluronic, Sigma-Aldrich) at 25 wt% in deionized, ultra-filtrated (DIUF) water. Pluronic F127 is a triblock copolymer of poly(ethylene oxide)-b-poly(propylene oxide)-b-poly(ethylene oxide) and is a commonly used fugitive ink for printing vascular features [18, 169]. A 1:1 mass ratio of red and blue food coloring is added to the Pluronic ink at 0.7 wt% to facilitate visualization. The ink is loaded at 2°C into a 3 cc syringe barrel (EFD Nordson), fixed with a cap, and degassed by centrifugation for 5 min before printing. The ink gels as warms to room temperature during the degassing step.

For matrix yielding experiments, blue SilcPig dye (Smooth-On, Inc.) is added at 1 wt% using the same mix-and-degas cycle for matrix preparation, and this dyed matrix is also loaded into a 3 cc syringe barrel (EFD Nordson), fixed with a cap, and degassed by centrifugation for 5 min before serving as a model ink for EMB3D printing.

3.2.2 Rheological Measurements

All rheological measurements are performed with a controlled-stress rheometer (DHR-3, TA Instruments, New Castle, DE, USA) equipped with either a 40 or 60 mm diameter, 2° cone and plate geometry. 60 mm diameter cone geometries are used for the 0 and 33 wt% SE 1700 matrix materials, while the 40 mm diameter cone is used for the others. In all experiments, the matrix materials are equilibrated for 10 min, a time that is well beyond the recovery time of these thixotropic matrices. Shear storage ($G'$) and loss ($G''$) moduli are measured as a function of shear stress at a frequency of 1Hz.
Silicone matrices of varying composition are characterized by subsequent steady-state flow sweep and flow ramp tests to qualitatively determine their thixotropic behavior [284–286]. In these “flow-sweep-flow-ramp” experiments, flow sweeps from low \(10^2 \text{s}^{-1}\) to high \(10^3 \text{s}^{-1}\) shear rates are conducted and immediately followed by flow ramps from high to low shear rates. In addition, three-phase modulus recovery tests are performed to quantitatively characterize their thixotropic behavior after applying a shear stress that exceeds the equilibrium yield stress, \(\tau_{y,0}\) [284–286]. In these experiments, \(G'(t)\) and \(G''(t)\) are measured during three phases of applied shear stresses (at 1 Hz frequency): 1 Pa for 3 min; 100 Pa for 1, 10, or 100s; and 1 Pa for 30 min. We defined their thixotropic recovery time, \(t_{\text{recover}}\), as the instant where \(G''(t) = G'(t)\) during the third restructuring phase of the modulus recovery test, or when:

\[
\frac{G''(t)}{G'(t)} = \tan(\delta) = 1.
\]

3.2.3 Embedded 3D Printing

Printing demonstrations are carried out in matrix materials with SE 1700 fractions of 33 wt\%, 50 wt\%, and 66 wt\% using a custom-designed, multi-material 3D printer (ABG 10000, Aerotech Inc., Pittsburg, PA, USA) with four independently addressable z-axis stages that house separate ink reservoirs [169]. All G-Code for printing is generated from Python-based software (MeCode, developed by J. Minardi). Before printing, matrix materials are poured into either plastic Petri dishes or machined Delrin molds and degassed under vacuum. The Pluronic ink is extruded through 12.7 mm long, 25 gauge print nozzles with inner and outer diameters of 0.25 mm and 0.52 mm, respectively (EFD Nordson). Two print speeds, 2 mm/s and 0.5 mm/s, are used for these experiments. At these speeds, the Pluronic ink is extruded with pressures of 30 and 25 psi, respectively.
3.2.4 Imaging and Videography

Photographs are taken using a digital SLR camera (Canon EOS 5D Mark II, Canon USA Inc) and a digital zoom microscope (VHX-2000, Keyence, Japan). All photographs are cropped using Affinity Designer vector graphics editor (www.affinity.serif.com/en-us/designer, Serif Europe Ltd).

3.3 RESULTS AND DISCUSSION

3.3.1 Matrix Design and Rheology

We designed thixotropic PDMS matrices from blends of SE 1700 and Sylgard 184 for EMB3D printing of soft robotic systems. SE 1700 is a stiff, silicone elastomer paste that contains fumed silica nanoparticles at approximately 20 wt%, and Sylgard 184 is used to dilute SE 1700 to achieve the desired rheological response for EMB3D printing. Specifically, we aimed to create a matrix material that behaves as shown in Figure 3.2. When the matrix is at rest (Figure 3.2i), the fumed silica fillers within the matrix will form a percolated network, giving rise to an equilibrium, at-rest shear yield stress, $\tau_{y,0}$ (from (5), $\lambda = 1$). As the nozzle travels through this matrix during EMB3D printing, the matrix yields, and the

![Diagram](https://via.placeholder.com/150)

Figure 3.2. Schematic illustration of a thixotropic, fumed silica-filled matrix material for crevice-free EMB3D printing. While at rest, the percolated network of fumed silica nanoparticles provides the matrix material’s required rheological properties for EMB3D printing. During nozzle translation, disruption of this network (ii) fluidizes the matrix (iii) and crevice formation is prevented. The fumed silica nanoparticles in the fluidized matrix (iv) restructure with time (v) to their equilibrium microstructure (vi).
Figure 3.3. Rheological properties of the matrix materials. (A) Apparent viscosity versus shear rate and (B) shear storage \(G'\), closed symbols) and loss \(G''\) open symbols) moduli versus shear stress for matrix materials with varying SE 1700 content. (C) Shear stress versus shear rate plots from subsequent flow sweep (closed symbols) and flow ramp (open symbols) tests on matrix materials with varying SE 1700 content. Herschel-Bulkley fits on the flow sweep and flow ramp data are shown as solid and dashed red lines, respectively (see Table 3.1). (D) Subsequent flow sweep (closed symbols) and flow ramp (open symbols) experiments on matrix materials with 50 wt% SE 1700 with different maximum shear rates (1, 10, and 100 s\(^{-1}\)) reveal that even slight disruption result in matrix fluidization.

Percolated filler network is disrupted, decreasing the apparent yield stress of the matrix material, \(\tau_{y,l}\) (from (5), \(\tau_{y,l} = \lambda \tau_{y,0}\,\) and \(\lambda < 1\)) (Figure 3.2ii). Complete disruption of the fumed silica microstructure will eliminate the yield stress of the matrix material, and \(\tau_{y,l} \rightarrow 0\) Pa (from (5), \(\lambda = 0\)) (Figure 3.2iii). When held in a quiescent state after perturbation (Figure 3.2iv), the percolated fumed silica network will slowly restructure (Figure 3.2v) to its equilibrium microstructure, and \(\tau_{y,l} \rightarrow \tau_{y,0}\) (Figure 3.2vi).

The rheological behavior of these matrix materials is dependent on their fumed silica content, i.e., the relative amount of SE 1700. We see from flow and amplitude sweeps that with increasing SE 1700
content, the matrix materials exhibit more pronounced shear thinning behavior (Figure 3.3A) and increasing \( \tau_{y,0} \) and stiffness (i.e., plateau \( G' \), or \( G'_0 \)) (Figure 3.3B). From the oscillatory amplitude sweeps, we find that matrix materials with 0 and 33 wt% SE 1700 do not possess a yield stress, and \( G'' > G' \). Matrix formulations with 50, 66, and 100 wt% SE 1700 possess \( \tau_{y,0} \) of approximately 25 Pa, 75 Pa, and 160 Pa, respectively. Flow-sweep-flow-ramp experiments qualitatively provide insight to the thixotropic nature of these matrices: in the steady-state flow sweep, the matrix is sheared from \( 10^2 \) s\(^{-1} \) to \( 10^2 \) s\(^{-1} \), during which the matrices yield; once the sweep has finished, the yielded material is immediately probed from \( 10^2 \) s\(^{-1} \) to \( 10^2 \) s\(^{-1} \). Measurements in the flow ramp portion of these experiments are collected without waiting for the flow to reach steady state. Instead, we rely on the inertia of the geometry to shear the yielded matrix and probe its yielded rheological behavior. By plotting shear stress versus shear rate for the flow-sweep-flow-ramp tests, their thixotropic behavior is revealed by the separation of the flow sweep and flow ramp curves (Figure 3.3C). The 0 wt% and 33 wt% SE 1700 matrices do not possess a yield stress or obvious thixotropy, while the 100 wt% SE 1700 matrix shows no thixotropic behavior due to its high fumed silica content. However, thixotropic behavior is observed for the 50 wt% and 66 wt% SE 1700 matrices. By fitting each of these curves to the Herschel-Bulkley model (1), we see the clear changes in yield stress of the 50 wt% and 66 wt% SE 1700 matrices after yielding (see Table 3.1, where Herschel-Bulkley parameters have subscripts 0 or \( t \) to indicate that \( \lambda = 1 \) or \( \lambda < 1 \), respectively).

<table>
<thead>
<tr>
<th>Matrix material</th>
<th>( \tau_{y,0} ) (Pa)</th>
<th>( K_0 ) (Pa ( \cdot ) s(^{n_0} ))</th>
<th>( n_0 )</th>
<th>( R^2 )</th>
<th>( \tau_{y,t} ) (Pa)</th>
<th>( K_t ) (Pa ( \cdot ) s(^{n_t} ))</th>
<th>( n_t )</th>
<th>( R^2 )</th>
</tr>
</thead>
<tbody>
<tr>
<td>100 wt% SE 1700</td>
<td>382</td>
<td>69.8</td>
<td>0.56</td>
<td>0.9982</td>
<td>365</td>
<td>79.8</td>
<td>0.54</td>
<td>0.9909</td>
</tr>
<tr>
<td>67 wt% SE 1700</td>
<td>80.1</td>
<td>10.4</td>
<td>0.90</td>
<td>0.9909</td>
<td>31.8</td>
<td>21.9</td>
<td>0.75</td>
<td>0.9983</td>
</tr>
<tr>
<td>50 wt% SE 1700</td>
<td>22.0</td>
<td>11.0</td>
<td>0.89</td>
<td>0.9998</td>
<td>0.4</td>
<td>12.6</td>
<td>0.85</td>
<td>0.9999</td>
</tr>
<tr>
<td>33 wt% SE 1700</td>
<td>0.0</td>
<td>5.8</td>
<td>0.93</td>
<td>0.9999</td>
<td>0.0</td>
<td>4.8</td>
<td>0.98</td>
<td>0.9999</td>
</tr>
<tr>
<td>0 wt% SE 1700</td>
<td>0.0</td>
<td>3.6</td>
<td>1.01</td>
<td>1.0000</td>
<td>0.0</td>
<td>3.8</td>
<td>1.00</td>
<td>1.0000</td>
</tr>
</tbody>
</table>
Figure 3.4. Storage modulus recovery of the 50 wt% SE 1700 matrix after yielding. (A) Storage modulus ($G'$) as a function of time during the three-phase thixotropy tests with deformation phases of 1, 10, and 100 s. (B) $G''/G' = \tan(\delta)$ as a function of time for each recovery phase in (A). Vertical dashed lines indicate the thixotropic recovery times, or the time at which $\tan(\delta) = 1$.

Preventing crevice formation in EMB3D printing matrices requires $\tau_{y,t} \to 0$ Pa upon nozzle translation to facilitate matrix fluidization and in-fill of the yielded matrix behind the nozzle. Table 3.1 indicates that the 50 wt% SE 1700 matrix is the only material that meets these requirements. We further explored the thixotropic nature of the 50 wt% SE 1700 matrix with additional flow-sweep-flow-ramp studies where the highest shear rate used was changed to be 1, 10, or 100 s$^{-1}$ (Figure 3.3D). Even with the 1 s$^{-1}$ study in which generated shear stresses are near $\tau_{y,0,r}$ we see clear fluidization of the matrix and $\tau_{y,t} \to 0$ Pa.

Plots of $G'$ over time obtained from three-phase thixotropy tests quantitatively illustrate how the 50 wt% SE 1700 matrix recovers after yielding (Figure 3.4). After a 3-min probe phase to analyze initial modulus, a shear stress of 100 Pa is applied for varying times during a deformation phase, resulting in temporary fluidization of the matrix. During the recovery phase, the modulus increases over time. Figure 3.4A shows a plot of the ratio $G''/G'$, given as $\tan(\delta)$, with time after each of the recovery phases measured in Figure 3.4B. We approximate the recovery of matrix yield stress as the onset of fumed silica filler percolation in the recovering matrix and approximate the time required as $t_{\text{recover}}$, or the moment...
\[\tan(\delta) = 1.\] The measured values of \(t_{\text{recover}}\) for applications of 100 Pa lasting 1, 10, and 100 s are marked in Figure 3.4B by the vertical dashed lines. We clearly see that larger deformation periods require longer restructuring times. However, because the momentary deformation incurred by nozzle translation through a discrete volume of matrix material during EMB3D printing happens within a time period shorter than 1 s, the true recovery time of the 50 wt% SE 1700 matrix is less than the \(t_{\text{recover}} \sim 200\) s required for the matrix to recover after being yielded by a 100-Pa shear stress for 1 s.

### 3.3.2 Embedded 3D Printing

A simple 3D form is EMB3D printed to illustrate the implications of matrix material composition and printing parameters on the fidelity of the features patterned within these matrices. For this demonstration, we used one ink (25 wt% Pluronic), one print nozzle \((d = 0.52\) mm), two print speeds \((V = 0.5\) mm/s and 2 mm/s), and three elastomeric matrices that represent a subset of those studied, i.e. 33 wt%, 50 wt%, and 66 wt% SE 1700 formulations. This simple 3D form (Figure 3.5A) is printed within the three matrices using the four unique print paths shown in Figures 3.5B-3.5D. The images of the patterned features reveal how matrix yielding impacts their fidelity (Figure 3.5E). Comparing these results, we find that matrix composition plays a critical role in enabling patterns that most closely resemble the 3D form in Figure 3.5A. The features printed with the 33 wt% SE 1700 matrix exhibit the most pronounced deviation from the intended form, while those printed in the 66 wt% SE 1700 matrix most closely followed it (Figure 3.5E). As the least thixotropic of the three matrices, the 66 wt% SE 1700 matrix gave rise to small crevices and raised peaks during EMB3D printing, and the sample had to be viewed from underneath to better visualize the results (Figure 3.6).

The results in Figure 3.5E also indicate that print path selection influences pattern fidelity. Independent of matrix composition, we find that the printed 3D form exhibits the closest resemblance to the desired form using Print Path #4, with the greatest deviations observed using Print Paths #1 and #2. In
Figure 3.5. Matrix composition and print path effects on pattern fidelity. (A) Top-down (top), side-on (middle), and 3D (bottom) views of a simple form that is EMB3D printed. The blue, green, and red arrows to the left represent the X-, Y-, and Z-axes, respectively. (B-D) Schematic illustrations of Print Paths #1 through #4. (B) For Paths #1 and #2, the form is printed in halves: (i) the left half of the form is printed, the nozzle is raised out of the matrix and (ii) brought to the next start location, and (iii) the right half of the form is printed. Paths #1 and #2 differ in that in Path #2 a 5-minute pause in printing is inserted before step (ii). (C) For Path #3, the form is printed in four segments (i) with each segment terminating at the vertex. (ii) Printing continues in a clockwise fashion until (iii) the last segment is printed. (D) For Path #4, the form is printed again in halves, but in a fashion to minimize ink displacement. (i) The bottom left and top right segments are first printed in one continuous filament, (ii) the tip is brought to the next location, and (iii) the top left and bottom right segments are printed in one continuous filament. (E) The results of printing the form with each print path in 33 wt% (left), 50 wt% (middle), and 66 wt% (right) SE 1700 matrices are shown for two print speeds, $V = 2$ mm/s (top row) and 0.5 mm/s (bottom row). (Nozzle $d = 0.52$ mm; scale bars are 3 mm.)
Figure 3.6. Underside view of the EMB3D printed forms in the 66 wt% SE 1700 matrix. The top and bottom rows of the printed forms in each set of results corresponds to print speeds of 2 mm/s and 0.5 mm/s, respectively (outer diameter of nozzle $d = 0.52$ mm, scale bar = 3 mm). With the samples flipped, the right most edge of this image would correspond to the left most edge of the image in Figure 3.5E.

Paths #1 and #2, the embedded form is printed in halves with or without a 5-minute delay between each half, respectively (Figure 3.5B), and matrix material yielding in front of the nozzle causes the deposited ink to be dragged downwards when the nozzle is translated away from the vertex. At slower print speeds, the separation distance between the halves at the intended vertex slightly decreases. Since thixotropic matrices recover their yield stress over time after yielding, Path #2 is used to explore the effects of the 5-minute recovery period on the final print results. However, no obvious difference in the printing fidelity was observed between Paths #1 and #2 (Figure 3.5E). In Path #3 (Figure 3.5C), the 3D form is printed in four continuous segments in a clockwise fashion, with each segment terminating in the center of the form at the intended vertex. Once a segment is printed, the nozzle is translated upwards in the Z-direction and out of the matrix material. While the desired form is still not achieved with Path #3, it does have improved fidelity compared to structures printed using Paths #1 and #2. Slight discontinuities at the vertex arise due to modest lifting of the printed ink features when the nozzle is removed from the matrix material or neighboring segments are printed. Finally, in Path #4 (Figure 3.5D), the form is printed in two diagonal halves. In the 50 wt% and 66 wt% matrix formulations, Path #4 clearly provides patterns that most closely resemble the intended form. The vertex is slightly off center due to ink displacement by
the translating nozzle, but this displacement is small and could be diminished by slightly increasing the applied printing pressure. Selection of matrix material (i.e., selection of appropriate matrix material rheology), print speed, and print path all contribute to the final results in EMB3D printing.

3.3.3 Matrix Material Yielding During Embedded 3D Printing

Print path selection and optimization is essential in EMB3D printing because the displacement of previously printed ink, as seen in Figures 3.5E and 3.6, results from matrix deformation around the translating print nozzle. To better understand how matrix composition, print speed, and nozzle diameter influence matrix deformation, another printing demonstration is conducted in which nozzles are translated through selectively dyed matrix materials (Figure 3.7). Specifically, the matrix is dyed blue and used as an ink to print lines 3 mm above the bottom of printing molds in matrix materials of identical SE 1700 content. After a 10 min pause to allow full recovery of the thixotropic matrices, the same nozzle is brought back into the matrix, 1 mm below the initial print height, and translated perpendicularly through the dyed matrix material features at various speeds (Figure 3.7A). For these experiments, three matrix materials (33, 50, and 66 wt% SE 1700), two nozzle diameters (0.31 and 0.72 mm), and five print speeds (0.125, 0.25, 0.5, 1, 2, 4, and 8 mm/s) were explored. Distortions in the dyed features are clearly observed in all conditions due to matrix displacement around the translating nozzle (Figure 3.7B). Overall, the highest distortions are observed for the 33 wt% SE 1700 matrix, and the extent of distortion in that matrix (and all others) generally increases with print speed and nozzle diameter.

Performing these experiments with other matrix materials, smaller/larger nozzle diameters, and higher/lower print speeds presented several printing- and imaging-related challenges. For example, matrix distortion was exceedingly large in experiments with matrix materials of SE 1700 content less than 33 wt%, while experiments with stiffer matrix materials produced distortions and crevices during printing. Furthermore, these experiments are somewhat limiting in the quantitative insights they could
Figure 3.7. Matrix yielding during EMB3D printing. (A) After a dyed matrix material is printed in a non-dyed matrix of equal SE 1700 content (top), a nozzle is translated through the matrix (middle) such that it displaces the selectively dyed matrix material feature (bottom). (B) Images of the displaced selectively dyed matrix material for various matrices, print speeds, and nozzle diameters. In the far right image, a bubble formed by a collapsed crevice in the 66 wt% SE 1700 matrix is in view. (Scale bars are 2 mm “O.D.” stands for outer diameter and is equivalent to d.)

provide overall. To develop a deeper understanding of these effects, we worked with Prof. Howard A. Stone’s Complex Fluids Group at Princeton University and used particle image velocimetry (PIV) as a tool for studying matrix yielding during EMB3D printing. Our primary findings are summarized in Figure 3.8. Specifically, we find that matrix yielding is mitigated when the matrix rheology and print conditions give rise to a high Oldroyd number, Od:

\[ Od = \frac{\tau_0 d^n}{K V^n} \]  

(7)
Figure 3.8. PIV results from an external study reveal the relationship between $Od$ and the dimensions of fluidized matrix material regions around translating nozzles during EMB3D printing. (a) Shear rate field with representative width $W$ and length $L$ measurements in the 50 wt% SE 1700 matrix with a nozzle diameter $d = 0.52$ mm and print speed $V = 1.5$ mm/s. The nozzle and print direction are represented by the grey circle and arrow, respectively. The width $W$ represents the maximum disturbed distance on both sides of the nozzle. The length $L$ represents the maximum disturbed distance in front of the nozzle. (b) Non-dimensional width ($W/d$) versus $Od$ for various matrix compositions, nozzle diameters, and print speeds. Red symbols represent 33 wt%, magenta represent 50 wt%, cyan represent 66 wt% and blue represent 80 wt% SE 1700. Squares represent $V = 0.25$ mm/s, triangles $V = 0.5$ mm/s, and circles $V = 1.5$ mm/s. No outlines represent $d = 0.52$ mm, grey outlines $d = 0.42$ mm, and black outlines $d = 0.31$ mm. (c) Non-dimensional length ($L/r$) versus $Od$ for various matrix compositions, nozzle diameters, and print speeds. The matrix composition, print speed and nozzle diameter correspond to the same symbol color, shape and outline parameters as shown in (b). This figure and the data it contains are adapted from a manuscript by Grosskopf, et al. (2018).

where $K$ and $n$ are the matrix material consistency and flow index, respectively, as calculated from fits of flow curve data to the Herschel-Bulkley yield stress fluid model (1). These findings are in good agreement with the printing demos shown in Figures 3.5 to 3.7, namely, that matrix displacement and yielding around translating nozzles is reduced when these matrices have a higher SE 1700 content (which
produces higher $\tau_y$ and $K$ and lower $n$) and the combined nozzle diameter ($d$) and print speed ($V$) produce a lower effective shear rate ($V/d$).

As the distortions observed for the 66 wt% SE 1700 matrix reveal (Figure 3.5E), choosing a matrix material that increases $Od$ for highest fidelity printing may not ultimately be appropriate for EMB3D printing. In fact, it was not feasible to print within matrices with more than 66 wt% SE 1700 due to the formation of crevices and simple difficulty of pouring these stiffer matrices into the printing molds. As such, suitable matrix materials for EMB3D printing must not only possess a high $Od$, but as also exhibit some thixotropic behavior that allows the matrix to “self heal” during the printing process. Therefore, the most suitable matrix material contains 50 wt% SE 1700, as it possesses high $\tau_{ymatrix}$, $Od > 1$ for the range of print speeds and nozzle diameters studied, and thixotropic behavior, which allows crevices to self heal behind the translating nozzle. Using this matrix, we created several complex architectures to illustrate this material’s suitability for EMB3D printing (Figure 3.9).

![Figure 3.9. EMB3D printed architectures.](image) Optical image of a printed array of high aspect ratio spirals (left) and a body centered cubic lattice (right), using the 25 wt% Pluronic ink (dark purple features) in a 50 wt% SE 1700 matrix with $d = 0.72$ mm and $V = 2$ mm/s. Acrylic cubes (5.1 cm edge length) contain the matrix during printing.
3.4 Conclusions

We have investigated the effects of matrix composition, rheology, and printing parameters on EMB3D printing. The ideal matrix material is a thixotropic, yield-stress fluid that enables features to be printed without forming crevices or other adverse defects. Our printing experiments clearly highlight the complex interplay between matrix rheology and printing parameters on feature fidelity, while the results from the PIV experiments corroborate these findings. Overall, maximizing Od is key to reducing the zone of yielded matrix material around the nozzle during EMB3D printing. For the specific matrix materials studied, the most direct means of tuning Od is by varying the SE 1700 content. In addition, optimization of the print path to minimize matrix yielding near previously printed features is essential. With this fundamental understanding in hand, a broad array of potential matrix materials can be systematically screened for use in EMB3D printing of soft robotics and other applications.
CHAPTER 4

EMBEDDED THREE-DIMENSIONAL PRINTING OF ENTIRELY SOFT, AUTONOMOUS ROBOTS

This chapter has been adapted from the publication:


4.1 INTRODUCTION

Soft robotics is a nascent field that aims to provide safer, more robust robots that interact with humans and adapt to natural environments better than their rigid counterparts [2, 24]. Unlike conventional robots composed of rigid materials, soft robots based on hydrogels [36, 288], electroactive polymers [30], granular media [104], and elastomers [84, 289] exhibit elastic moduli ranging from 100 kPa to 1 GPa [2], are physically resilient [16, 130], and have the ability to passively adapt to their environment [2, 24, 80]. Molded and laminated elastomers with embedded pneumatic networks are widely used materials in soft robotics [2, 84, 290]. Actuation of these elastomeric composites occurs when interconnected channels comprising the pneumatic network are inflated with incompressible fluids or gases supplied via tethered pressure sources [2]. To date, robotic end effectors with bioinspired [148] and rapid [96] actuation, deployable crawlers [16, 92] and swimmers [15] with complex body motions, and robust jumpers [176, 181] have been developed based on this design strategy. However, in each case, these robots are either tethered to or carry rigid systems for power and control, yielding hybrid soft-rigid systems [15, 16, 176, 182].

Creating a new class of fully soft, autonomous robots [190] is a grand challenge, as it requires soft analogs of the control and power hardware currently used. Recently, monopropellant fuels have been suggested as a promising fuel source for pneumatically actuated soft robots [22, 182]. Their rapid decomposition into gas upon exposure to a catalyst offers a strategy for powering soft robotic systems
Figure 4.1. Entirely soft, autonomous robot assembly. (A) A pre-fabricated soft controller with microfluidic logic is (B) loaded into a mold. (C) Matrix materials are poured into the mold for (D, E) EMB3D printing of fugitive and catalytic inks (scale bar in (E) represents 10 mm). (F) After matrix curing, the printed fugitive ink “auto-evacuates” yielding open channels (scale bar represents 2 mm). (G) The Octobot is removed from the mold and inverted to reveal an autonomous, fully soft robot, controlled via the embedded microfluidic soft controller and powered by monopropellant decomposition (scale bar represents 10 mm). Fluorescent dyes have been added in (E) and (G) to assist in visualization of internal features.

that obviates the need for batteries or external power sources. In this chapter, we report a method for creating a completely soft, pneumatic robot, the “Octobot,” with eight arms that are powered by monopropellant decomposition. To accomplish this, we use microfluidic logic [21] as a soft controller and multi-material, embedded 3D (EMB3D) printing method to fabricate pneumatic networks within a molded elastomeric, robot body (Figure 4.1). Our hybrid assembly approach allows one to seamlessly integrate soft lithography, molding, and 3D printing to rapidly and programmably fabricate a range of materials and functional elements in the form factors that are required for autonomous, untethered operation of a soft robot.

4.2 EXPERIMENTAL METHODS

4.2.1 Soft Controller Fabrication

Soft controllers are fabricated from Sylgard 184 poly(dimethyl siloxane) (PDMS, Dow Corning Corp., Auburn, MI, USA) using soft lithography molding and bonding techniques. First, a mold was
patterned on a silicon wafer using SU-8 negative photoresist (Microchem, Corp. Westborough, MA, USA). SU-8 3050 photoresist was used to achieve 100 mm film thickness. Baking, exposing, and developing steps were performed in accordance with product specifications in the product datasheet. The completed wafer is placed in a petri dish to form a competed mold assembly.

Soft controllers consist of an upper mold, a lower mold, and an intermediate thin film. The upper and lower molds are made on one wafer to ease fabrication. PDMS is poured into the mold assembly to a height of one millimeter. Separately, PDMS is spun coat onto a wafer at 1500 rpm for 60 s for a film thickness of 35 µm. After curing at 90°C for 20 minutes, PDMS forms are removed from the molds, and holes are punched at all inlets and outlets. The upper layer is bonded to the wafer-adhered thin film after exposing to oxygen plasma at 35 Watts for 20 s in a Deiner Pico plasma system (Deiner Electronic GmbH). Holes are punched in the thin film, masks are placed as described by Mosadegh, et al., [291] and the lower layer is bonded to the thin film using the plasma recipe above.

4.2.2 Preparation of the Inks, Matrix Materials, and Fugitive Plug Material

Two inks, a “fugitive ink” and “catalytic ink,” are formulated for EMB3D printing. The fugitive ink is prepared by adding 27 wt% gel of Pluronic F127 to ice-cold, deionized, ultra-filtrated (DIUF) water, followed by mixing in a planetary mixer for 5 min at 2000 rpm, and storing at 4°C. The fugitive ink is not used until the Pluronic F127 completely dissolves in solution. The ink is prepared for printing by loading the solution at 4°C in a 3 cc syringe barrel (EFD Nordson, East Providence, RI, USA) and centrifuged at 3000 rpm for 5 min to degas. For EMB3D printing, the fugitive ink’s barrel is fitted with a stainless steel nozzle (0.15 mm inner diameter, EFD Nordson). The fugitive ink is extruded at 65 psi during printing using an 800 Ultra fluid dispensing system (EFD Inc., East Providence, R.I., USA), and features are printed using speeds of 1.5, and 10 mm/s for all mesofluidic features and vent orifice features, respectively.
The catalytic ink is prepared by dissolving a synthesized, diacrylated Pluronic (F127-DA) at 30wt% concentration with a solution of Irgacure 2959 (at 0.5wt%, BASF) in DIUF water at 4°C. The F127-DA is synthesized under an inert nitrogen atmosphere by first adding 400 mL of dry toluene (Sigma, St. Louis, MO, USA) to a three-neck flask fixed to a condenser with circulating cold water and magnetically stirred at 300 rpm. 70 g of Pluronic F127 (Sigma) is then dissolved in the toluene after heating the solvent to 60°C. After the solution is allowed to cool to room temperature, triethylamine (5.6 g, Sigma) is added to the solution, followed by the drop-wise addition of acryloyl chloride (5 g, Sigma) with continued stirring, both at a molar ratio of 10:1 with the Pluronic F127. The reaction mixture is stirred overnight and maintained in the inert atmosphere. The diacrylated Pluronic F127 (F127-DA) product is then filtered from the yellow triethylammonium hydrocholoride byproduct and precipitated from the filtered solution with hexane (Sigma) at a 1:1 volume ratio. The F127-DA is obtained through a second filtration step and allowed to dry in a chemical hood for at least 24 h. This protocol is adapted from Wu, et al [18]. For each gram of the synthesized F127-DA solution (30 wt%), 100 mg of PEG-DA is added, and this solution is mixed in a planetary mixer for 1 min at 2000 rpm and degassed for 3 min at 2200 rpm. This mixture is then stored in the dark at 4°C. Finally, 5 w/w% Pt black (Sigma) is added to this base solution at 4°C and mixed in a planetary mixer for 5 min at 2000 rpm. The Pt-filled F127-DA physically gels during mixing, facilitating loading into a UV-blocking 3cc syringe barrel (EFD Nordson) for printing. Note, this catalytic ink is freshly prepared for each print session, as the Pt black slowly crosslinked the acrylate moieties present in the ink. For EMB3D printing, the syringe barrel housing this ink is fitted with a stainless steel nozzle (0.33 mm inner diameter, EFD Nordson). The catalytic ink is extruded at 55 psi during printing using an 800 Ultra fluid dispensing system (EFD Inc., East Providence, R.I., USA), and catalytic features are printed using speeds of 1.5 mm/s. After printing, the catalytic ink is crosslinked for 15 min at 18 mW/cm² under a UV source (Omnicure EXFO).
Two matrix materials are developed for fabricating fully soft robots. The first matrix, referred to as the “body matrix,” is prepared by blending two commercially available PDMS formulations: Sylgard 184 and SE 1700 (Dow Corning). SE 1700 is a silicone elastomer paste that contains fumed silica nanoparticles. Sylgard 184 PDMS is used to dilute SE 1700 to achieve the desired rheological response for embedded 3D printing. As discussed in Chapter 3, we find that the optimal body matrix is composed of a 1:1 mass ratio of SE 1700 (4:1 ratio of base to hardener) and Sylgard 184 (10:1 ratio of base to hardener). This matrix is prepared by mixing the blend in a planetary mixer at 2000 rpm for 3 min with degassing at 2200 rpm for 2 min. The second matrix, referred to as the “fuel reservoir matrix,” is prepared by mixing Part A Ecoflex 00-30 to Part B Ecoflex 00-30 (with 1.2 w/w% Slo-Jo Platinum Silicone Cure Retarder and 1.2 w/w% Thivex, Smooth-On Inc., Macungie, PA, USA) in a 1:1 ratio. The matrix is prepared in a planetary mixer at 2000 rpm for 1.5 min with degassing at 2200 rpm for 1 min.

Lastly, the “fugitive plug” material used to prevent ingress of the body matrix material into the soft controller is prepared prior to printing by first synthesizing and then mixing a diacrylated Pluronic material (F127-DA) (at 30 wt% in a 0.5 wt% solution of Irgacure 2959 in DI water) with F127 (at 30 wt% in DI water) at a mass ratio of 1:4. The fugitive plug is stored in the dark at 4°C in a syringe. When used, the fugitive plug material is allowed to physically gel before it is crosslinked for 3 min at 6 mW/cm² under a UV source.

4.2.3 Rheological Characterization of Inks and Matrix Materials

All flow and oscillatory amplitude sweeps are carried out using a controlled-stress rheometer (DHR-3, TA Instruments, New Castle, DE, USA) equipped with a 40 mm diameter, 2° cone and plate geometry. In all experiments, the fugitive and catalytic inks are equilibrated at room temperature for 1 min before testing; the fuel reservoir and body matrix materials are equilibrated for 20 min and 10 min, respectively, to simulate the times at which Octobot printing began with each material. Amplitude
sweeps for measuring shear storage moduli, $G'$, as a function of shear stress are conducted at a frequency of 1Hz.

### 4.2.4 Octobot Mold Fabrication

Octobot molds are fabricated inside a CNC machined Delrin® mold equipped with two locating pins to mount the soft controller. Their desired shape is modeled in Solidworks. A negative mold is created in Parasolid format for file transfer. MasterCAM is used to develop all machining tool paths and to export final G-code for final fabrication. Blanks of 105 mm length are cut from black acetal (Delrin®), stock size 1x3 inch (McMaster Carr, Santa Fe Springs, CA, USA). Acetal is used due to its dimensional stability, and thick stock is chosen to prevent warping during machining and repeated Octobot curing cycles. Octobot molds are produced by CNC milling on a HAAS OM-2A vertical machining center (HAAS Automation Inc, Oxnard, CA, USA). 1 mm dowel pins are pressed into drilled holes for soft controller mounting.

### 4.2.5 Octobot Assembly

A complete illustration of the multi-step assembly process outlined below is provided in Figure 4.2. To fabricate an Octobot, we first micro-mold [21, 291] the soft controller that houses the microfluidic logic necessary for controlling fuel decomposition. To prepare a mold (Figure 4.2A) for EMB3D printing, Ecoflex 30 (Smooth-On, Inc.) is first prepared with 1 wt% Slo-Jo and 0.25 wt% Thivex (both with respect to Part A) by mixing in a Thinky planetary mixer for 1.5 min with a 1 min degas cycle. This uncured Ecoflex 30 is cast into the actuator layers of the Octobot mold and degassed in a vacuum chamber for 3 min. A glass slide is used to remove excess material and create smooth surfaces that will ultimately become the extensible layers of the actuators. The molds are then placed in a 90°C oven for 30 min to cure the Ecoflex, removed, and trimmed of excess material as necessary (Figure 4.2B). The soft controller,
Figure 4.2. Workflow for EMB3D printing an Octobot. (A) An EMB3D printing mold is machined from Delrin®. (B) The hyperelastic layers needed for actuation are cast and crosslinked in the mold’s actuator regions. (C) A soft controller protected with a polyimide tape mask is loaded onto the EMB3D printing mold’s pins. (D) The fuel reservoir matrix material is carefully loaded into the fuel reservoir area of the mold and degassed under vacuum. (E) Liquefied fugitive plug material is manually loaded into the soft controller via the inlets and briefly degassed. (F) The protective tape is removed after the fugitive plug material physically gels, and the fugitive plug is photocrosslinked. (G) The body matrix material is cast into the mold and degassed. (H) Any excess body matrix material is removed with a squeegee step, EMB3D printing begins, and the entire mold and EMB3D printed materials are placed in a 90°C oven to crosslink. (I) After two hours, the crosslinked Octobot is removed from its mold and kept at 90°C for a total of four days to ensure complete auto-evacuation of the aqueous fugitive inks. (J) Before operation, excess body matrix material is removed via laser cutting. (K) The final Octobot, shown here in a close-up view, is prepared for operation.

protected temporarily with a polyimide mask, is then placed into this mold with the prepared hyperelastic layers needed for actuation, loaded onto the press-fit pins placed in the mold, and the polyimide (Kapton) tape is left adhered (Figure 4.2C).
A custom-designed, multi-material 3D printer (ABG 10000, Aerotech Inc., Pittsburg, PA, USA) with four independently z-axis addressable ink reservoirs is used to for EMB3D printing the fugitive and catalytic inks within the Octobot matrices \[169\]. All G-Code for printing is generated from Python-based software (MeCode, developed by J. Minardi). Registration coordinates and print heights are then taken from the cured Ecoflex layers in the actuators and in all inlets of the soft controller; these are essential for EMB3D printing and provided to the custom print software. The fuel reservoir and body matrix materials are then prepared as described previously. While the body matrix material is mixing, the fuel reservoir matrix is deposited in the fuel reservoir region of the printing mold (Figure 4.2D). It is then degassed for three minutes to ensure no trapped gas is present. Excess bubbles in the uncured fuel reservoir matrix are removed with a pipettor. Non-gelled, chilled fugitive plug is then filled throughout the soft controller via injection through the inlets (Figure 4.2E). While the fugitive plug is still in the liquid state, it is briefly degassed in a vacuum chamber. The fugitive plug material is then allowed to physically crosslink, excess gel is scraped from the top of the tape, the tape is removed, and the fugitive plug is photocrosslinked with a UV source at 6 mW/cm\(^2\) for 3 min (Figure 4.2F). After the gels are crosslinked, the body matrix is cast within the mold, covering the fuel reservoir matrix and the fugitive plug-filled soft controller (Figure 4.2G) and degassed for 1-3 min \textit{in situ}. Again, excess bubbles are removed with a pipettor, excess material is scraped off and away form the mold with a glass slide, and EMB3D printing of the fugitive and catalytic inks begins (Figure 4.2H). After printing, the entire mold is cured at 18 mW/cm\(^2\) for 15 min to crosslink the catalytic ink. The mold is then transferred to a 90°C oven, where the matrix materials are cured. The Octobot is removed from the mold and kept at 90°C for 4 days to facilitate “auto-evacuation” of the aqueous fugitive inks, during which water evaporates from the inks and diffuses through the matrix, leaving behind an open network of channels that are interfaced with the soft controller (Figure 4.2I). After auto-evacuation, Octobot fabrication is completed upon removal of excess matrix material (Figure 4.2J) by release cutting from the surrounding matrix material using a CO\(_2\) laser (Universal Laser
Systems, Scottsdale, AZ, USA). The ready Octobot is cleaned with isopropyl alcohol and water (Figure 4.2K).

Before operation, Sylgard 184 PDMS (Dow Corning Corp. Auburn, MI, USA) is poured into the Octobot’s open cavity above the soft controller to a height of 1.5 mm and cured at 90°C for 20 min. A 1 mm biopsy punch (Militex Inc, York, PA, USA) is used to punch holes through the newly poured PDMS layer and into the fuel inlets. Dyed water is injected into these holes to inflate the fuel tanks, flow through the system, and insure proper device function. Holes are punched in the downstream orifice features to allow the water to vent from the system.

### 4.2.6 Actuator Characterization

Stand-alone actuators with “two” and “four bladder” designs are EMB3D printed using special actuator characterization molds machined from Delrin®. First, Ecoflex 30 (Smooth-On, Inc.) is first prepared with 1 wt% Slo-Jo and 0.25 wt% Thivex (both with respect to Part A) by mixing in a Thinky planetary mixer for 1.5 min with a 1 min degas cycle. This uncured Ecoflex 30 is cast into hyperelastic layers of the actuator characterization molds and degassed in a vacuum chamber for 3 min. A glass slide is used to remove excess material and create smooth surfaces, and the molds are placed in a 90°C oven for 30 min to cure the Ecoflex. The body matrix material is prepared, poured into the actuator characterization mold prepared with hyperelastic layers, and the actuators are printed using the fugitive ink.

To prepare them for characterization, the actuators are first released from mold assembly and then a 1 mm hole is created with a biopsy punch (Militex Inc. York, PA, USA), which serves as the air inlet. Each actuator is pressurized slightly to ensure inflation and then tested for angular displacement (i.e., the actuator is allowed to deflect unconstrained and the total displacement angle is measured) and blocked force (i.e., the actuator is constrained from deflection and resultant force is measured). For each
actuator, break in testing consists of five cycles, in which actuator air pressure is slowly (~30 s) ramped up to the pressure set point, then slowly (~30 s) ramped down to ambient. Pressure set point for the first cycle is $P_0$, and set point for all following cycles is $P_1$ (Table 4.1). Data acquisition consists of five additional cycles for each actuator, in which air pressure is cycled as above to pressure set point $P_1$.

<table>
<thead>
<tr>
<th>Hyperelastic layer thickness, $h$ (µm)</th>
<th>Two bladder actuators</th>
<th>Four bladder actuators</th>
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<tbody>
<tr>
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<td>$P_0$ (bar)</td>
<td>$P_1$ (bar)</td>
</tr>
<tr>
<td>500</td>
<td>0.35</td>
<td>0.3</td>
</tr>
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<td>750</td>
<td>0.4</td>
<td>0.35</td>
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<tr>
<td>1000</td>
<td>0.45</td>
<td>0.4</td>
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<tr>
<td>1250</td>
<td>0.5</td>
<td>0.45</td>
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<tr>
<td>1500</td>
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For characterization of angular displacement, actuators are plumbed with regulated compressed air, and mounted vertically between a matte black background and a Sony NEX3 digital camera for video data acquisition. Actuators are pressurized with five break-in cycles as described above, followed by five data acquisition cycles. As above, the first break in cycle is to $P_0$, and all subsequent break in and data acquisition cycles are to $P_1$. Video data is analyzed using ImageJ image analysis platform (NIH.gov) to obtain bend angle versus pressure for each actuator.

For blocked force characterization, individual actuators are mounted on a fixed platform beside an Instron model 5544 materials testing frame (Illinois Tool Works Inc., Norwood, MA, USA). The actuator is lowered until just above the force sensor portion of the testing frame, and the actuator is plumbed with regulated compressed air. Actuators typically behave differently upon initial few actuations versus subsequent actuations due to the Mullins effect [108]. Each actuator therefore receives
five “break in” cycles prior to data acquisition. Air pressure and actuator force data is recorded on the Instron testing frame data acquisition system at 100 ms intervals.

4.2.7 Untethered, Autonomous Soft Robot Operation

The Octobot is loaded into an acrylic tank outfitted with a backlight to highlight colored fuel as it flows through the system. Aqueous hydrogen peroxide (90 wt%, HTP grade, Peroxychem, Philadelphia, PA, USA) is diluted to 50 wt%, and samples dyed red and blue are filled into two syringes prepared with this liquid fuel mixture. The syringes are loaded onto a syringe pump, and connected to the Octobot via 1mm diameter silicone rubber tubing. Water is flowed into the acrylic tank to wash away dye in the Octobot exhaust stream and drained into a nearby sink. The syringe pump flows fuel at a rate of 3 mL/min (each syringe) into the Octobot for 10 s. The silicone rubber tubing is removed with tweezers from the Octobot, which is allowed to operate untethered. The Octobot alternates actuation until fuel pressure is insufficient to switch the oscillator and alternating actuation ceases.

4.2.8 Imaging and Videography

Photographs and supporting videos are acquired with a digital SLR camera (Canon EOS 5D Mark II, Canon USA Inc) and a 4K video (Blackmagic Production 4K, Blackmagic Design, Melbourne, Australia). Photos are cropped using Inkscape vector graphics editor (www.inkscape.org), and video sequences are clipped from raw footage and exported using iMovie (Apple Corp, Cupertino, CA, USA). All print parameter measurements and images of EMB3D printed features in Octobots are taken with a digital zoom microscope (VHX-2000, Keyence, Japan) and cropped using Affinity Designer vector graphics editor (www.affinity.serif.com/en-us/designer). Their mean values and standard deviations are determined from three samples printed at each print speed of interest.
4.3 RESULTS AND DISCUSSION

4.3.1 Octobot Design

Central to the integrated design of the autonomous, entirely soft Octobot is the incorporation of a soft, microfluidic controller (Figure 4.3A) that regulates the on-board timing and location of fuel decomposition (Figure 4.3B). This control system is roughly divided into four distinct sections: upstream (liquid fuel storage), oscillator (liquid fuel regulation), reaction chamber (decomposition into pressurized gas), and downstream (gas distribution for actuation and venting) (Figure 4.3C). Upstream, monopropellant fuel is infused via a syringe pump into each of two fuel reservoirs printed into the hyperelastic matrix, which expand elastically and force fuel into the oscillator. Upstream check valves in the soft controller prevent fuel from flowing back out the fuel inlets. The fuel reservoirs expand elastically, forcing fuel into the oscillator. The oscillator includes a system of pinch and check valves based on prior designs [21], which convert pressurized fuel inflow into alternating fuel outflow. With one channel temporarily occluded, fuel from the other channel flows from the soft controller’s outlets into the Pt laden reaction chambers, where it rapidly decomposes. The geometry of the microfluidic soft controller is designed to operate at a fuel flow rate of ~40 µL/min [21][21]. Downstream, the resulting pressurized gas, prevented from returning to the soft controller via downstream check valves, flows into one of the downstream mesofluidic networks comprised of four actuators and one orifice. The supplied pressure deflects the actuators and exhausts to atmosphere through the vent orifice. These subcomponents operate based on the interaction and timing of the local pressures, similar in concept to an electrical oscillator (Figure 4.3D). Upon successful venting, the fuel flow into one reaction chamber stops and flow to the other begins, initiating a similar sequence in the other downstream catalytic chamber and actuator network (Figure 4.3E).
Figure 4.3. Octobot control logic and design. A system of check valves and switch valves within the soft controller regulates fluid flow into and through the system. (A) A schematic illustration shows the soft controller within an Octobot. (B) The soft controller’s oscillator causes an Octobot to alternate between blue (top) and red (bottom) actuation states. The photos of the soft controller show the switch valves’ conformation for each state. The monopropellant fuel is dyed blue and red to show states, and the arrows indicate direction of liquid fuel flow. Scale bar is 5 mm. (C) A schematic and (D) qualitative electrical analogy of the Octobot system are provided, where check valves, fuel tanks, oscillator, reaction chambers, actuators, and vent orifices are akin to diodes, supply capacitors, electrical oscillator, amplifiers, capacitors, and pull down resistors, respectively. (E) Conceptual curves show key variables as a function of time. (1) Nominal pressure drives fuel through system at a decreasing rate. (2) Pinch valves in the oscillator convert upstream flow into alternating flow between red and blue channels. Flow rate and switching frequency are functions of upstream pressure and downstream impedance. (3) When upstream pressure is too low, oscillation is not possible, so both sides flow at reduced rate. (4) Catalyst decomposes fuel, yielding pressurized gas, which flows downstream to the actuators and the vent orifices concurrently. (5) Actuators deform based on pressure. Vents must be sufficiently small to allow full actuation, yet sufficiently large to allow timely venting.

To provide an on-board power source, we use an aqueous hydrogen peroxide solution as the fuel due to its high energy density (1.44 kJ/g as compared to 0.1-0.2 kJ/g for batteries) as well as its benign
decomposition byproducts. As the fuel decomposes in the presence of the platinum catalyst, the following reaction occurs $2\text{H}_2\text{O}_2$ (l) $\rightarrow$ $2\text{H}_2\text{O}$ (l,g) + $\text{O}_2$(g), which results in a volumetric expansion of approximately 240 times at ambient pressure [22]. Since this monopropellant liquid fuel can be handled in small volumes and decomposes at the point of use, we can use microfluidic logic to directly handle the fuel, eliminating the need for external valves [16] to control gas at high pressure and flow rate.

### 4.3.2 Matrix Materials and Inks

By combining micro-molding with EMB3D printing, we rapidly patterned the required mesofluidic networks by extruding sacrificial inks through fine nozzles that are embedded within the uncured elastomer matrices and seamlessly integrated them with the microfluidic features of the soft controller. To self-heal crevices that form within the “body matrix” as the nozzle is translated during the printing process, we used an elastomeric matrix material comprised of fumed silica nanoparticles that exhibits thixotropic behavior [284]. Specifically, we use the optimized matrix material formulation based on 50 wt% SE 1700 (see Chapter 3). Briefly, when completely restructured or at rest, this matrix behaves like a Herschel-Bulkley fluid, i.e., it exhibits both shear-thinning behavior and a yield stress. These properties ensure that the extruded inks remain in place within the matrix [18, 19]. However, upon yielding, the body matrix readily flows into any crevices formed. The body matrix restructures with time, ultimately recovering its original viscoelasticity, which ensures that EMB3D printing can be repeated later in the same matrix region. We also created a “fuel reservoir” elastomeric matrix, into which fuel reservoir channels are printed. Both the body and fuel reservoir matrices are cured within the mold after printing is completed.

To create the fuel reservoirs, catalytic reaction chambers, actuator networks, and vent orifices, two hydrogel-based inks (fugitive and catalytic) are printed into the molded matrix materials (Figure 4.4). These printed features are interfaced with each other as well as the soft controller through the use of
“fugitive plugs” introduced at the controller’s inlets prior to filling the mold with the matrix materials.

The fugitive ink is composed of an aqueous, poly(ethylene oxide)-b-poly(propylene oxide)-b-poly(ethylene oxide) triblock copolymer (Pluronic F127) gel [18, 169]. The catalytic ink contains platinum particles suspended in a mixture of Pluronic F127-diacrylate (F127-DA) and poly(ethylene glycol) diacrylate (PEG-DA) that is photocrosslinked after printing.

Flow (Figure 4.5A) and amplitude sweeps (Figure 4.5B, where only the storage moduli, $G'$, are shown for clarity) reveal the shear thinning and yield-stress behavior, respectively, of the viscoplastic
Figure 4.5. **Rheological behavior of the inks and matrix materials.** Plots of (A) apparent viscosity versus shear rate and (B) storage modulus, $G'$, versus shear stress for the fugitive ink (red), catalytic ink (black), body matrix material (blue), and fuel reservoir matrix material (green).

matrix materials and viscoelastic inks. Shear yield stresses, $\tau_y$, of the material components, determined form the amplitude sweeps in Figure 4.5B as the cross-over point at which $G' = G''$, are approximately 450 Pa for catalytic ink, 320 Pa for the catalytic ink, 35 Pa for the body matrix, and 25 Pa for the fuel reservoir matrix. The plateau storage moduli, $G''$, are approximately 21 kPa for the catalytic ink, 16 kPa for the fugitive ink, 3.1 kPa for the fuel reservoir matrix, and 1.7 kPa for body matrix. With $\tau_y$ and $G''$ of the inks approximately an order of magnitude greater than those of the matrix materials, the components of the materials palette display appropriate rheological behavior for EMB3D printing following empirically identified trends in previous works [18, 19] as well as the guidelines established in Chapter 3.

### 4.3.3 Printing and Auto-Evacuation of the Fugitive and Catalytic Inks

The printed features produced from both inks can be changed “on-the-fly” by simply varying the print speed (Figure 4.6), which is important for printing features both as narrow as the vent orifices and as wide as the soft controller interconnects. Typically, this fugitive ink must be removed or “evacuated” after printing to yield open channels [18, 169]. However, we find that the fugitive ink composed of pure Pluronic F127 can be auto-evacuated by heating the printed features within the crosslinked, silicone-
Figure 4.6. **Print parameter characterization of the fugitive and catalytic inks.** (A) Optical images of cross-sections from fugitive ink traces printed at various print speeds demonstrate that trace dimensions can be changed “on-the-fly” (scale bars are 100 μm). (B) Optical image of a cross-section of a reaction chamber printed with the catalytic ink containing a Pt-laden plug (scale bar is 500 μm). (C) Scanning electron micrograph of the Pt-laden plug shows catalyst availability (scale bar is 25 μm). (D) A plot of cross-sectional trace width versus print speed for fugitive and catalytic inks printed at 65 psi (450 kPa) and 50 psi (345 kPa), respectively, highlight the decrease in trace width with increasing print speed (data points represent \( n = 3 \) measurements, error bars indicate standard deviation).

Figure 4.7. **Auto-evacuation of the fugitive and catalytic inks.** Photographs of an octobot’s reaction chambers with upstream portions of the actuator networks (top) and a one-pad actuator (bottom) at various times, \( t \), reveal the auto-evacuation of the fugitive and catalytic inks, which leaves behind open channels that serve as mesofluidic features.

Based matrices at 90°C [292] (Figure 4.7). As water evaporation ensues, the triblock copolymer species either form a thin coating at the matrix-open channel interface or they may partially diffuse into the
matrix [293]. The fugitive plugs within the soft controller’s inlets also undergo this auto-evacuation process, facilitating connectivity between the microfluidic logic and all printed mesofluidic components (Figure 4.8). By contrast, the catalytic ink is crosslinked in place after printing yielding a Pt-laden plug within the matrix (Figure 4.6B).

4.3.4 Free Displacement and Blocked Force Characterization of the Actuators

The actuators, which consist of printed bladders in contact with a lower modulus, hyperelastic elastomer layer (Figure 4.9A), are designed to inflate asymmetrically to generate angular displacement (Figure 4.9B). Their maximum working pressure and displacement are tuned based on the thickness of the hyperelastic layer, $h$ (Figure 4.9C). If this layer is too thin, as we observed when $h = 500 \mu m$, it ruptures prematurely. However, while the maximum displacement angle, $\theta$, and generated force (Figure 4.9D) increase with $h$, so too does the required working pressure. As a compromise, we kept $h$ as 1000 $\mu m$, as it affords consistent performance at the lowest working pressure. Note, these actuators produce
Figure 4.9. Octobot actuator characterization. (A) Two-bladder actuator design in which traces (i) are printed in contact with the hyperelastic layer (ii) inside of the body matrix material (iii), and the difference in modulus of (ii) and (iii) result in bending upon inflation. The thickness, \( h \), of the hyperelastic layer (ii) is modified to change actuator characteristics. The body matrix material possesses a height of 800 mm. (B) The actuator tip angle \( \theta \) changes upon inflation (scale bar represents 10 mm). (C) Mean displacement angle, \( \theta \), versus inflation pressure taken from three representative actuators with varying hyperelastic layer heights, \( h \), in units of \( \mu m \). Error bars indicate 95% confidence interval. (D) Blocked force versus pressure curves for two-bladder actuators of varying \( h \) in units of \( \mu m \). The lines in the plot are third degree polynomial fits of data collected from five inflation-deflation cycles of representative actuators. The shaded regions indicate 95% confidence interval.

0.04 N of force (Figure 4.9D). Thus, in an ideal system, two actuators are theoretically sufficient to lift the 7 g Octobot. While the actuators could be redesigned for increased force generation (one option demonstrated in Figure 4.10), any new soft controller/actuator designs or system scaling would require careful balancing of fuel supply, flow rate, and actuator requirements.

4.3.5 Autonomous, Untethered Octobot Operation

For autonomous Octobot operation and timely cycling between actuation states, a system-level balance between supply gas flow, actuation pressure, and exhaust rate is required. One important first parameter that is held constant through system design is the fuel concentration. While higher fuel concentrations would provide increased gas production and energy density for more robust actuation, our fuel contains 50 wt% H2O2. Above this concentration, fuel decomposition resulted in combustion of the organic materials within the catalytic reaction chambers printed from the catalytic ink (Figure 4.11) and the ultimate failure of the system. Upon injection of 0.5 mL of fuel via syringe pump, each of the
Figure 4.10. Extended actuator characterization. (A) Schematic of four-bladder actuator design in which traces (i) are printed in contact with the hyperelastic layer (ii) inside of the body matrix material (iii), similar to the actuators shown in Figure 4.9. The thickness, $h$, of the hyperelastic layer (ii) is modified to change actuator characteristics. The body matrix material possesses a height of 800 mm. (B) The four-bladder actuator before (top) and during (bottom) inflation. The tip angle $\theta$ changes upon inflation (scale bar represents 5 mm). (C) Mean displacement angle, $\theta$, versus inflation pressure taken from three representative actuators with varying hyperelastic layer heights, $h$, in units of $\mu$m. Error bars indicate 95% confidence interval. (D) Blocked force versus pressure curves for four-bladder actuators of varying $h$ in units of $\mu$m. The lines in the plot are third degree polynomial fits of data collected from five inflation-deflation cycles of representative actuators. The shaded regions indicate 95% confidence interval.

Figure 4.11. Demonstration of printed catalyst integrity with different fuel concentrations. Catalytic ink was EMB3D printed to form Pt laden plugs in the shapes of squares (indicated with arrows). (A) These features were dropped into H$_2$O$_2$ at 50% (left) and 90% (right). (B) High concentration H$_2$O$_2$ fuels resulted in combustion of the Pt-laden plugs formed with the catalytic ink. (C) After fuel decomposition, (D) the catalyst in the 50% H$_2$O$_2$ fuel was recovered.
Octobot’s two fuel reservoirs elastically expand to a pressure of approximately 50 kPa. At this operating pressure of 50 kPa and fuel concentration of 50 wt%, a volumetric expansion of gas of 160 times from the liquid state is expected [22]. Therefore, with the geometry of the microfluidic soft controller designed to operate at a fuel flow rate of ~40 µL/min and a theoretical maximum oscillation rate of 5.5 switches per minute, pressurized gas can theoretically be produced at a rate of ~6.4 mL/min [21]. Under these operating conditions, the theoretical runtime of 12.5 min could be achieved using a system with a fuel capacity of 1 mL. Using an actuator hyperelastic elastomer layer thickness of 1000 µm, we then tailored the diameter of the vent orifices by modulating print speed. Orifices roughly 75 µm in width allowed proper actuator displacement with timely subsequent venting. The ability to rapidly pattern and adjust the geometry of these features “on-the-fly” via EMB3D printing allowed us to iterate through more than 30 designs and nearly 300 Octobots to converge on an appropriate system-level architecture.

Through this iterative process, we created Octobots with embedded components that work together in concert for autonomous Octobot operation. Fuel is infused via a syringe pump into each of the two fuel reservoirs embedded in the hyperelastic fuel reservoir matrix, the fuel lines connecting the infusing syringes and the fuel inlets of the soft controller are removed, and fuel is forced from the pressurized reservoirs into the oscillator which autonomously alternates between the red and blue actuation states. The resulting Octobots operated autonomously (Figure 4.12), cycling between actuation states for four to eight minutes (Figure 4.13). While this is less than the predicted theoretical runtime, the soft controller does alternate actuation states as expected. We believe that downstream impedances arising from decomposition-actuation-venting cycles as well as the decreasing flow rate of fuel into the soft controller with time are responsible for the departure from theoretical performance [21].

These and other issues can be addressed by integrating more sophisticated microfluidic circuits as soft controllers. A major strength of our integrated design and fabrication approach is its ability to incorporate a wide variety of existing and novel microfluidic devices. Thus, future soft robot designers
Figure 4.12. **Untethered, autonomous Octobot operation.** The soft controller’s oscillator causes an Octobot to alternate between actuation states. The monopropellant fuel is dyed to show states. Stills from top-down (top) and face-on (bottom) operation videos show an Octobot autonomously alternating between blue (“1”) and red (“2”) actuation states. Arrows indicate inflated, bending actuators. Scale bars are 10 mm.

Figure 4.13. **Autonomous switching between actuation states during Octobot operation.** (A) Switching in the soft controller was tracked with time according to the Octobot operation video from which the top row of stills in Figure 4.12 were taken. (B) The corresponding inflation of actuators associated with the blue and red actuation states are also reported with time.

can leverage the extensive work reported by the microfluidics community to develop more sophisticated control systems and individual logic devices. For example, in addition to the current microfluidic controller, pneumatic and hydraulic oscillator systems [21, 294–298] have been developed to generate any number of output signals and varying oscillation frequencies. Microfluidic circuits and logic operators
have also been developed [299–301], including fluidic versions of electric logic gates, e.g. NAND/NOR
AND/OR, XOR/XNOR [302–304], flip-flops, and gain valves [305–307]. By integrating these advances, one
can envision new entirely soft robots with more complex actuation strategies, advanced control schemes,
and new form factors across a broad range of length scales.

4.4 CONCLUSIONS

In summary, we have demonstrated the first untethered operation of a robot composed solely of
soft materials. The coupling of monopropellant fuels and microfluidic logic allowed us to power, control,
and realize autonomous operation of these pneumatically actuated systems without conventional robotic
hardware. Through our hybrid assembly approach, we constructed both the robot body and embedded
the necessary components for fuel storage, catalytic decomposition, and actuation to enable system-level
function in a rapid manner. The Octobot is a minimal system designed to demonstrate our integrated
design and fabrication strategy, which may serve as a foundation for a new generation of entirely soft,
autonomous robots.
CHAPTER 5

SOFT SOMATOSENSITIVE ACTUATORS VIA EMBEDDED 3D PRINTING

This chapter has been adapted from a manuscript prepared for *Advanced Materials*:


5.1 INTRODUCTION

Humans possess manual dexterity, motor skills, and other physical abilities that rely on feedback provided by the specialized receptors and afferent neurons that comprise our somatosensory system. The emerging field of soft robotics has strived to replicate these capabilities [2, 3] for myriad applications, including universal [104] and compliant end effectors for robotic handling [129, 266, 269], wearable devices for rehabilitation and performance enhancement [76, 129, 270], robust systems that operate in extreme environments [15–17, 175], and autonomous soft robots [16, 17, 182, 308]. However, to realize their full potential, next-generation soft robots require integrated soft sensors that provide discrete, somatosensory feedback motifs, including proprioceptive, haptic, and thermoceptive sensing [2, 3, 309].

Molded and laminated elastomers with embedded pneumatic networks are widely used materials for soft robots [2, 3, 309]. Recent efforts have focused on introducing sensing capabilities within these fluidic elastomer actuators (FEAs) [129, 178, 235, 248, 263–266]. However, to date, most embodiments provide only a single mode of feedback that is achieved by integrating conventional rigid sensors [132, 195, 310] or soft sensors composed of conductive liquid metals [235, 265–267], aqueous ionic conductors [178, 248], or conductive nanoparticle-filled polymer composites [263, 264]. Quite recently, FEAs that provide multiple modes of sensory feedback have been reported [129, 266, 267]. Most notably, FEAs with proprioception and exteroceptive contact sensing have been achieved by either incorporating multiple liquid metal sensors [266, 267] or elastomeric waveguides that provide feedback through optical
power loss during waveguide deformation [129]. However, their fabrication requires multiple molding and lamination steps that constrain their overall complexity and sensing capabilities. Importantly, designs that rely on ionically conductive sensing pose additional challenges, including sensor failure due to water evaporation [238, 239, 246], while those based on conductive nanoparticle-filled polymers exhibit hysteresis as their particle network is disrupted during large, repeated deformations [204, 220].

In this chapter, we report a method for creating soft robotic actuators innervated with a complex network of sensors, referred to as soft somatosensitive actuators (SSAs), via multimaterial, embedded 3D (EMB3D) printing [18–20, 308]. This manufacturing approach enables the seamless integration of multiple ionically conductive and fluidic features within elastomeric matrices to create SSAs with the desired actuation and bioinspired sensing capabilities. By assembling three SSAs into a soft robotic gripper, we specifically demonstrate proprioceptive and haptic feedback enabled by embedded curvature, inflation, and contact sensors. Harnessing the temperature-dependent ionic conductivity [311, 312] of the SSAs’ contact sensors coupled with our free-form fabrication process, we also created SSAs with temperature, and deep-versus-fine touch contact sensing, respectively, which have not yet been realized by other soft robotic actuators.

5.2 EXPERIMENTAL METHODS

5.2.1 Materials

Three matrix (dorsal, actuator, and anterior) materials and two (sensor and fugitive) inks are used to create soft somatosensitive actuators (SSAs) by EMB3D printing. All matrix materials are formulated from commercially available two-part, platinum-cure silicone elastomers and compatible cure retarder (Slo-Jo) and thickener (Thivex) (all from Smooth-On Inc., USA). The dorsal matrix is prepared by mixing Parts A and B of Ecoflex 00-10 in a 1:1 ratio with 1.2 w/w% (with respect to Part B) Slo-Jo Platinum Silicone Cure Retarder and 0.6 w/w% (with respect to Part B) Thivex in a planetary mixer at 2000 rpm for
1 min followed by a 2 min degas cycle at 2200 rpm (all from Smooth-On Inc., Macungie, PA, USA). The actuator matrix is prepared by mixing Parts A and B of SortaClear 40 in a 10:1 ratio with 2 w/w% (with respect to Part A) Slo-Jo Platinum Silicone Cure Retarder and 0.15 w/w% (with respect to Part A) Thivex in a planetary mixer at 2000 rpm for 2 min followed by a 7 min degas cycle at 2200 rpm (all from Smooth-On Inc., Macungie, PA, USA). Before Part A SortaClear is used, it is degassed under vacuum for 1 hr, aliquotted as necessary into the mixing containers, and degassed again for 5 min at 2200 rpm in the planetary mixer. The anterior matrix is prepared by mixing Parts A and B of Ecoflex 00-30 in a 1:1 ratio with 1.5 w/w% (with respect to Part B) Slo-Jo Platinum Silicone Cure Retarder and 1.2 w/w% (with respect to Part B) Thivex in a planetary mixer at 2000 rpm for 1 min followed by a 2 min degas cycle at 2200 rpm (all from Smooth-On Inc., Macungie, PA, USA).

The sensor ink is prepared before printing by combining 1-ethyl-3-methylimidazolium ethyl sulfate (EMIM-ES, ≥95%, Sigma-Aldrich, USA, stored under an inert N₂ environment) and Aerosil 380 fumed silica (Evonik, USA) at 6 wt% and mixing in a planetary mixer for 15 min at 2000 rpm with a 1 min degas cycle at 2200 rpm. The resulting ionogel is carefully loaded into a capped 10 cc syringe barrel (EFD Nordson) and degassed by a centrifugation cycle of 5 min at 1000 rpm, 5 min at 3000 rpm, and another 5 min at 3000 rpm after any large trapped air bubbles are carefully aspirated out of the ink with a pipette. The fugitive ink (25 wt% Pluronic F127) is prepared by adding Pluronic F127 to ice-cold deionized, ultrafiltrated (DIUF) water and mixing in a planetary mixer (Thinky ARE-310 mixer, USA) for 5 min at 2000 rpm. The ink is stored at 4°C until the aqueous Pluronic F127 is in the liquid state. Before EMB3D printing, the fugitive ink is loaded at 4°C into a capped 10 cc syringe barrel (EFD Nordson, East Providence, RI, USA) and degassed by centrifugation at 3000 rpm for 10 min. As the fugitive ink warms to room temperature during this degassing step, it undergoes a fluid-to-gel transition. The syringe barrels that house each ink are fitted with 1.5 inch- and 1 inch-long stainless steel nozzles, respectively (0.25 mm inner diameter, 0.52 mm outer diameter, EFD Nordson, USA).
5.2.2 Rheological Characterization

The rheological properties of the matrix materials and inks are measured using a stress-controlled rheometer (DHR-3, TA Instruments, New Castle, DE, USA) equipped with a 40 mm diameter, 2” cone geometry. All measurements are carried out at room temperature (21°C) after a stationary equilibration phase of 10 min or 1 min for the matrix materials and inks, respectively. The 10 min equilibration time was selected to simulate the time between matrix material preparation and ultimate EMB3D printing and allow these thixotropic materials to reach steady-state rheological behavior. For oscillatory strain sweeps, the shear storage ($G'$) and loss ($G''$) moduli were measured at a frequency of 1Hz.

To characterize the thixotropic behavior of the matrix materials, we carried out flow sweep and ramp tests as well as three-phase modulus recovery tests to determine the time required for each material to recover its initial equilibrium yield stress yield stress, $\tau_{y,0}$. For the subsequent flow sweep and flow ramp tests, steady-state flow sweeps are conducted from low ($10^{-2}$ s$^{-1}$) to high ($10^{2}$ s$^{-1}$) shear rates and immediately followed by ramp sweeps from high ($10^{2}$ s$^{-1}$) to low ($10^{-2}$ s$^{-1}$) shear rates. In the three-phase modulus recovery trials, $G'$ and $G''$ are measured during three different phases of applied shear stresses at a frequency of 1 Hz. In the first phase, a shear stress well below the respective matrix yield stress is applied for 3 min to probe the quiescent, steady-state moduli of the matrix materials. In the second phase, a shear stress correlating to the stress generated in the respective matrix material at a shear rate of $V/D$, where $V$ is the maximum print speed that a nozzle of diameter $D$ travels in that matrix material. Note, this shear stress is applied for 1 s, the shortest experimental time possible for this apparatus. The third and final phase is conducted to probe the recovery of the initial rheological behavior of the matrix materials. The same respective probing stresses used in the first phase of the experiment are applied during this third phase. In each phase, shear stress is applied at a frequency of 1 Hz. We define their thixotropic
recovery time, $t_{\text{recover}}$, as the instant where $G''(t) = G'(t)$ during the third restructuring phase of the modulus recovery test, or when:

$$\frac{G''(t)}{G'(t)} = \tan(\delta) = 1.$$ 

The parameters used for the thixotropy tests measurements provided in Table 5.1.

<table>
<thead>
<tr>
<th>Matrix material</th>
<th>V/D (s$^{-1}$)</th>
<th>Approximate shear stress at V/D (Pa)</th>
<th>Shear stress applied in first phase (Pa)</th>
<th>Shear stress applied in second phase (Pa)</th>
<th>Shear stress applied in third phase (Pa)</th>
</tr>
</thead>
<tbody>
<tr>
<td>Dorsal matrix</td>
<td>11.5</td>
<td>430</td>
<td>1.0</td>
<td>430</td>
<td>1.0</td>
</tr>
<tr>
<td>Actuator matrix</td>
<td>7.7</td>
<td>420</td>
<td>1.0</td>
<td>420</td>
<td>1.0</td>
</tr>
<tr>
<td>Anterior matrix</td>
<td>7.7</td>
<td>130</td>
<td>0.1</td>
<td>130</td>
<td>0.1</td>
</tr>
</tbody>
</table>

### 5.2.3 SSA Mold Fabrication

A layered mold assembly is designed to allow precise height control of individual matrix layers as well as alignment during EMB3D printing. All mold materials are purchased from McMaster-Carr Supply Company. Parts are machined on a Haas OM-2A vertical machining center (Haas Automation Inc, USA), and all laser cutting is performed with a VersaLaser PLS6.75 CO$_2$ laser (Universal Laser Systems). A black acetal (Delrin) base layer is machined containing a cavity in the shape of the dorsal matrix layer and holes for mounting screws and alignment pins. This first mold layer is machined from acetal for its dimensional stability and machinability. 2.54 cm thick stock is selected as the layer thickness to prevent warping during repeated thermal cycling. The second mold layer of clear polycarbonate is machined in the shape of the actuator matrix and includes holes for mounting screws and alignment pins. Polycarbonate is used to allow visual inspection during fabrication and for its relative dimensional stability during thermal cycling. The third mold layer of clear acrylic is laser cut in the geometry of the
anterior matrix with holes for mounting screws and alignment pins. The clear acrylic allows for visual
inspection during EMB3D printing. The thicknesses of these final two mold layers define the height of
their respective matrix layers. During fabrication, all three layers were securely screwed together using
commercial fasteners to further increase dimensional stability of individual layers and the assembly.

5.2.4 SSA Fabrication

A custom-designed, multimaterial 3D printer (ABG 10000, Aerotech Inc., Pittsburg, PA, USA)
equipped with four independently addressable z-axis stages is used to fabricate the SSAs [169, 308]. All
G-Code for EMB3D printing is generated from Python-based software developed in-house [308]. The
complete EMB3D printing tool path for a SSA is illustrated in Figure 5.1.

Printing begins by loading the dorsal matrix material into the first layer of the mold assembly.
Any air bubbles are carefully removed by aspiration with a pipettor and excess matrix material is
squeegeed off with a glass slide (note: this process is repeated for each subsequent layer). This mold layer
is loaded onto the printer and the curvature sensor is printed within the dorsal matrix (printing time ~1
min). The second mold layer is then added and fastened with screws to the mold assembly while the
actuator matrix is prepared. The actuator matrix material is carefully loaded into the mold assembly and
any air bubbles are removed by aspiration with a pipettor. Several features are then printed within the
actuator matrix, including the SSA pneumatic network and spacers, the curvature sensor inlets, inflation
sensor, and inflation sensor inlets. This printing step takes approximately 40 min, after which the mold
assembly is retrieved from the printer. The third mold layer is added and fastened to the middle mold
layer with screws to the mold assembly while the anterior matrix material is prepared. The anterior
matrix material is carefully loaded into the mold assembly. Using the sensor ink, the contact sensor inlets
are first printed into the actuator matrix with vertical traces brought up in the Z-direction into the
Figure 5.1. Complete workflow for embedded 3D (EMB3D) printing of a soft, somatosensitive actuator. (A-L) The workflow is shown from a side-view cut-through perspective. All sensing and actuation features are printed with the sensor (red) and fugitive (dark blue) inks, respectively. (A) The first mold layer is filled with the dorsal matrix, and (B) the curvature sensor is printed. (C) The second mold layer is fixed to the first mold layer and filled with the actuator matrix. (D) Two curvature sensor inlets are printed (only one shown), beginning first in the curvature sensor, moving up through the actuator matrix, and terminating with horizontal leads. (E) Eight bladder spacers are printed in the actuator matrix such that they make contact with the dorsal matrix material. (F) Top segments of the inflation sensor (18 total, only nine shown) are then printed in the actuator matrix on either side of a bladder spacer. (G) The actuator bladders (nine) are printed above these segments of the inflation sensor. (H) Around each bladder, vertical segments of the inflation sensor are printed (18 total, only nine shown). (I) The inflation sensor is completed by selectively printing horizontal segments to create the inlets (two are printed, only one shown) as well as connections with the vertical segments. The inflation sensor is only connected around three bladders, but other sensor ink features must wrap around the other bladders to ensure homogenous bladder inflation. (J) A single segment of fugitive ink is used to connect all bladders and create an actuator inlet. (K) The third mold layer is fixed to the mold assembly and filled with the anterior matrix. (L) Contact sensor inlets are printed, beginning in the actuator matrix and extending into the anterior matrix (two printed, only one shown). Finally, the contact sensor and its distal meander are printed in the anterior matrix such that there is continuity between the sensor and its inlets that extend into the actuator matrix.
anterior matrix. The contact sensor is printed into the anterior matrix, making contact with the inlets that terminate in the actuator matrix (print time ~3 min).

After the printing process is completed, the SSA is crosslinked at room temperature for at least 12 h and at 90°C for 2 h. The SSA is removed from the mold assembly. A 6.35 mm-long, 22 gauge syringe tip (EFD Nordson) is pushed into the back end of the SSA through the actuator matrix and into the actuator inlet. The SSA is kept at 4°C for 1 h to liquefy the fugitive ink. Using a syringe, 10 mL of ice-cold DIUF water is slowly filled into the SSA actuator to remove the fugitive ink. These water infill and aspiration steps are repeated a total of three times, after which the SSA is dried in a 90°C oven for 2 h. Electric leads are prepared by soldering 26 gauge, silicone-coated wire to nickel-plated pins sized for insertion into the EMB3D printed SSA sensors. The pins are inserted through the actuator matrix into the sensor inlets, and the wires are connected to the readout electronics.

5.2.5  Readout Electronics and Resistance Measurements

Based on prior strategies for measuring the resistance, \( R \), of ionically conductive soft sensors [239], we use the read-out electronics shown in Figure 5.2 to measure \( R \) for each EMB3D printed sensor. To measure sensor resistance, a relaxation oscillator using a low-voltage inverting operational amplifier is used to pass an oscillatory voltage in the form of a square wave over a sensor, identified as \( R_s \) in Figure 5.2A. The square wave voltage \( (V_{out,1}) \) alternates between \( V_{max} = +1.2 \text{V} \) and \( V_{min} = -1.2 \text{V} \) at a period, \( T \), given by:

\[
T = 2R_2C_1 \ln \left( \frac{1+B}{1-B} \right)
\]

where \( B = R_1/(R_1 + R_2) \), \( C_1 \), \( R_1 \), and \( R_2 \) are the other passive components in the relaxation oscillator, and the resistance change in the ionic sensor, \( \Delta R \), is given by:

\[
\Delta R = R_s - R_{s,0} = \frac{\Delta T}{2R_sC_1 \ln \left( \frac{1+B}{1-B} \right)}
\]
**Figure 5.2. Readout hardware.** (A) Schematic illustration of the readout hardware used to acquire feedback from each printed sensor. The relaxation oscillator circuit, contained in the blue square, is responsible for passing an alternating voltage across the sensor, which is indicated by the red variable resistor, $R_s$, and equals $R_{s,0}$ when the sensor is not deformed. The output signal, $V_{\text{out,1}}$, has the form of a square wave with voltages alternating between +1.2 V and -1.2V. The period of $V_{\text{out,1}}$ is dependent on the resistance of the sensor, as given by Equation 3 (Methods). The elements in the green region of the schematic process $V_{\text{out,1}}$ according to Equation 4 (Methods) to provide a resistance measurement for the sensor, $R_s$. (B) Schematic views of representative $V_{\text{out,1}}$ signals are provided in the oscilloscope, where the top waveform is $V_{\text{out,1}}$ when $R_s = R_{s,0}$ and the bottom is $V_{\text{out,1}}$ when $R_s > R_{s,0}$. For our sensors, we selected $C_1 = 10$ nF, $R_1 = 680$ Ω, and $R_2 = 9.9$ kΩ, which gave us oscillation frequencies in $V_{\text{out,1}}$ between 600 and 1000 Hz for non-deformed sensors.

where $\Delta T = T - T_0$. For our sensors, we selected $C_1 = 10$ nF, $R_1 = 680$ Ω, and $R_2 = 9.9$ kΩ, which gave us oscillation frequencies in $V_{\text{out,1}}$ between 600 and 1000 Hz for non-deformed sensors. The operating principle behind the readout electronics is illustrated in Figure 5.2B, where the schematic signal illustrates the form of $V_{\text{out,1}}$ when the sensor is at rest (top waveform) and when the sensor is deformed such that its resistance increases (bottom waveform). To measure $R_s$, a NI USB-6212 16-bit data acquisition unit (National Instruments, Austin, TX) connected to a computer is used to measure $V_{\text{out,1}}$ for each sensor with a MATLAB script that samples $V_{\text{out,1}}$ at a frequency of 130 kHz. The MATLAB script computes the period of the waveform, and $\Delta R$ is determined by Eq. (1), where $R_0$ is defined as the initial $R$ at the beginning of the measurement trial.
5.2.6 SSA Characterization

To study the influence of relative humidity of sensor resistance, 21 strain sensors were printed into 3 mm-deep molds filled with the anterior matrix material, cured in a 90°C oven overnight, and had their resistances measured. For these sensors, $R_0 = 1186±134$ kΩ ($n = 21$) at Day 0. Batches of 7 sensors are kept at one of three different conditions: on a laboratory benchtop with relative humidity (RH) of $53±2\%$, in a dry box with RH = $27±2\%$, and in a tissue culture incubator at a temperature of $37°C$ and RH = $85\%$. The $R$ of each sensor is measured at Days 1, 2, 4, 6, 8, 14, and 20. After the Day 20 measurement, the sensors are kept in a 90°C oven, and $R$ measurements were taken on Day 21 and Day 25 of the experiment, corresponding to 1 and 5 days in the 90°C environment. Before taking any measurements, the sensors are kept on a laboratory benchtop for 30 min.

Before any data acquisition or general use, SSAs undergo three break-in cycles to remove any influences on actuator or sensor performance arising from the Mullins effect. A break-in cycle for a SSA involved inflating to 14 kPa for 20s, deflating for 20s, inflating to a 28 kPa, and so on, increasing the inflation pressure in 14 kPa increments until a final inflation pressure of 152 kPa was used. Inflation is achieved using pressure regulation from 800 Ultra fluid dispensing systems (EFD Inc., East Providence, R.I., USA).

During free and blocked displacement characterization, SSAs are inflated over the pressure range of 0 to 152 kPa. SSAs are inflated for 30 s at a given inflation pressure, resistance measurements are recorded for each sensor (taken as the mean $R$ over a 3 s sampling interval), and the inflation pressure is increased/decreased by 14 kPa. For free displacement characterization, photographs are taken during each measurement, and the displacement angle of the SSA tip is measured in ImageJ image analysis software (NIH.gov). For blocked displacement characterization, SSAs are inflated against a mass balance. Force generated by the SSA is calculated as the product of the mass recorded on the mass balance and
acceleration due to gravity. Figure 5.3A-B show the free and blocked displacement characterization setups.

For contact sensor characterization, a translation stage (i.e., a z-axis on the multi-material printer) is used to generate a contact pressure against the contact sensor’s distal meander while the SSA is laid on the mass balance upside-down (see Figure 5.3C). An acrylic rectangle is laser cut with dimensions covering the area of the contact sensor’s distal meander and glued to a Luer lock assembly that is fixed to our translation stage. A ~2 mm thick piece of cured anterior matrix with the same dimensions as the acrylic rectangle separates the acrylic rectangle and the SSA to eliminate any stress concentration effects. Using the stage, the acrylic part is pressed into the contact sensor at 0.2 mm increments, generating contact pressures (calculated as the product of the measured mass and acceleration due to gravity divided by the area of the acrylic rectangle). Resistance measurements are recorded at each increment (taken as the mean $R$ over a 3 s sampling interval). Identical methods are used to characterize the fine and deep contact sensors, which used rigid acrylic parts sized to the areal dimensions of each sensor.
To characterize the temperature-induced changes in sensor $R$, a strain sensor is placed on the Peltier plate of the DHR-3 rheometer (set at 20°C). The temperature is kept constant for 60 s before $R$ is recorded (taken as the mean $R$ over a 3 s sampling interval). Without removing the sensor, the temperature is changed to 5°C, $R$ is recorded, and this process continues in 5°C increments over the temperature range of 5 to 95°C in 5°C increments.

5.2.7 Gripper Assembly Design and Fabrication

To form the soft robotic gripper, three individual SSAs are mounted in a triangle shaped configuration and tethered to compressed air regulators (EFD Inc., East Providence, R.I., USA) and readout electronics. Actuator mounts are laser cut from acrylic on a VersaLaser PLS6.75 CO$_2$ laser (Universal Laser Systems, USA) and manually formed using a heat gun and hand tools. The mount adapter is CNC machined using a Haas OM-2A vertical machining center (Haas Automation Inc., USA). Three SSAs are first fastened into actuator mounts and then onto the mount adapter. All tethers to the compressed air regulators and readout electronics are threaded through the hollow tube on which the mount adapter was held. All hardware and raw materials are purchased from McMaster-Carr Supply Co., USA.

5.2.8 Imaging and Videography

Photographs and supporting videos are taken with a digital SLR camera (Canon EOS 5D Mark II, Canon USA Inc). Supporting micrographs are taken with a digital zoom microscope (VHX-2000, Keyence, Japan). An IR camera (FLIR T621, FLIR Systems Inc.) is used to record thermal videos during gripper grab tests. Video sequences are clipped from raw footage and exported using iMovie (Apple Corp, USA), and all photographs and still shots taken from videos are cropped using Affinity Designer vector graphics editor (www.affinity.serif.com/en-us/designer, Serif Europe Ltd).
5.3 RESULTS AND DISCUSSION

5.3.1 SSA Design and Assembly

The SSAs are fabricated by directly writing the sensing and fluidic networks within molded elastomeric matrices via multimaterial, embedded 3D (EMB3D) printing (Figure 5.4). Specifically, conductive ionogel and fugitive inks are printed within three elastomeric matrices – the dorsal, actuator, and anterior matrix materials – to define the sensor and pneumatic networks required for actuation, respectively. The key steps for EMB3D printing of SSAs are highlighted in Figure 5.4A. Briefly, the dorsal, actuator, and anterior matrix materials are loaded sequentially in the mold and the following features are printed: (i) the curvature sensor in the dorsal matrix, (ii) the FEA features (including actuator spacers and bladder network) and inflation sensor in the actuator matrix, and (iii) the contact sensor in the anterior

![Figure 5.4. Fabrication of soft somatosensitive actuator (SSA) innervated with multiple soft sensors.](image)

(A, i) The curvature sensor is printed within the dorsal matrix (Layer #1), the actuator features and inflation sensor are printed within the actuator matrix (Layer #2), and the contact sensor is printed in the anterior matrix (Layer #3). (B) Schematic illustrations and (C) images of the final SSA. The images in (C) are taken under black light exposure, and the fugitive (blue) and sensor (red) inks have been fluorescently dyed to facilitate visualization (scale bars are 10 mm).
Figure 5.5. **Soft somatosensory actuators.** All photographs are taken under black light exposure. Fluorescently dyed fugitive (blue) and sensor (red) inks are used to aid visualization of the internal SSA features. (A-C) Photographs of various steps during EMB3D printing. (A) After all features are printed within the second mold layer and before adding the third mold layer, (B) excess actuator matrix material is carefully removed. (C) The anterior matrix material is then added, and the contact sensor is printed. (D-H) Photographs of the SSA removed from the mold assembly after matrix material curing. (D) Top-down and (E) end-on views highlight the internal features of the SSAs. (F) A close-up of the contact sensor’s distal meander. Scale bars in (A)-(E) are approximately 10 mm; scale bar in (F) is 1 mm.

matrix. All sensor leads terminate in the actuator matrix. After printing, the matrix materials are cured and the fully fabricated SSA (Figure 5.4B-C, Figure 5.5) is removed from the mold assembly. Next, the fugitive ink is evacuated from the SSA leaving behind an open network of pneumatic channels, and electrical leads are inserted into the sensors. Each SSA bends freely in a semi-circular motif when inflated, and the inflation, curvature, and contact sensors directly embedded within the body of the SSA are designed to deform, and consequently increase in resistance, when the SSA inflates, bends, and makes contact with external bodies, respectively (Figure 5.6).
5.3.2 Matrix Materials and Inks

As established in previous chapters, each elastomeric matrix must possess the requisite rheological properties for EMB3D printing as well as the appropriate mechanical properties upon curing. In particular, the matrices should exhibit shear thinning, yield-stress, and thixotropic behavior prior to curing. These properties facilitate the matrices’ flow into each mold layer as well as the integration of functional features via EMB3D printing [308]. Results from subsequent flow sweep and flow ramp (flow-sweep-flow-ramp) experiments performed with each matrix are shown in Figure 5.7A-C. Fits of these data to the Herschel-Bulkley equation are provided in Table 5.2 and reveal that the dorsal, actuator, and anterior matrices are all shear-thinning, yield stress fluids and exhibit thixotropy. According to these fits, the dorsal, actuator, and anterior matrices possess τ_{yy} of approximately 48 Pa, 72 Pa, and 24 Pa, respectively. Compared to previous fumed silica-filled PDMS matrices explored in Chapters 3 and 4, these matrices exhibit significantly higher consistency, K_0, and shear-thinning exponent n_0. Additionally, τ_{yy} determined for each matrix from the flow ramp data are higher than for the previous matrices. This
Figure 5.7. **Matrix thixotropy.** (A–C) Log-log plots of the shear stress as a function of shear rate for (A) the dorsal matrix, (B) actuator matrix, and (C) anterior matrix from flow sweep (filled circles) and subsequent flow ramp (open circles) measurements. (D) Log-log plots of storage modulus, $G'$, as a function of time for each matrix material from three-phase modulus recovery experiments. (E) The tangent of the phase angle, $\tan(\delta)$, as a function of time is plotted for the modulus recovery phase of (D). The thixotropic recovery time for each matrix material is indicated as the time at which $\tan(\delta) = 1$.

Table 5.2. **Herschel-Bulkley fits of data from subsequent flow sweep and flow ramp experiments.**

<table>
<thead>
<tr>
<th>Matrix material</th>
<th>$\tau_{y,0}$ (Pa)</th>
<th>$K_0$ (Pa $\cdot s^{n_0}$)</th>
<th>$n_0$</th>
<th>$R^2$</th>
<th>$\tau_{y,t}$ (Pa)</th>
<th>$K_\tau$ (Pa $\cdot s^{n_\tau}$)</th>
<th>$n_\tau$</th>
<th>$R^2$</th>
</tr>
</thead>
<tbody>
<tr>
<td>Dorsal matrix</td>
<td>47.9</td>
<td>84.3</td>
<td>0.62</td>
<td>0.9982</td>
<td>33.0</td>
<td>32.7</td>
<td>0.83</td>
<td>0.9999</td>
</tr>
<tr>
<td>Actuator matrix</td>
<td>71.7</td>
<td>142</td>
<td>0.48</td>
<td>0.9835</td>
<td>31.6</td>
<td>33.9</td>
<td>0.85</td>
<td>0.9986</td>
</tr>
<tr>
<td>Anterior matrix</td>
<td>24.2</td>
<td>28.0</td>
<td>0.63</td>
<td>0.9987</td>
<td>17.8</td>
<td>11.7</td>
<td>0.82</td>
<td>0.9994</td>
</tr>
</tbody>
</table>

arises due to the dorsal, actuator, and anterior matrices’ shorter pot life, and the matrices thicken slightly over the time required for performing the flow-sweep-flow-ramp tests.
Plots of $G'$ over time obtained from three-phase thixotropy tests on the three matrices provide more quantitative insight into how each matrix recovers after yielding (Figure 5.7D). After a 3-min probe phase to analyze initial modulus, appropriate shear stresses (see Table 5.1 and Experimental Methods) are applied for 1 s during a deformation phase to simulate temporary fluidization the matrix by a translating nozzle. The matrices’ moduli increase over time during the final recovery phase. Figure 5.7E shows a plot of the ratio $G''/G'$, given as $\tan(\delta)$, with time after each of the recovery phases measured in Figure 5.7D. By approximating the recovery of matrix yield stress as the moment $\tan(\delta) = 1$, we can determine values of $t_{\text{recovery}}$ for each matrix. $t_{\text{recovery}}$ for the matrices are marked in Figure 5.7E by the vertical dashed lines. We find $t_{\text{recovery}}$ is approximately 220 s, 14 s, and 204 s for the dorsal, actuator, and anterior matrix materials, respectively. As discussed in Chapter 3, this temporary fluidization enables for crevice-free EMB3D printing. Finally, upon curing, the dorsal matrix consists of a highly extensible elastomer (Shore-00 Hardness of 10) that exhibits minimal impedance on SSA bending. The actuator matrix exhibits the highest durometer (Shore-A Harness of 40) to enable appropriate force generation, while the anterior matrix consists of an elastomer with an intermediate durometer (Shore-00 Hardness of 30).

The sensor ink is a conductive ionogel composed of the organic ionic liquid 1-ethyl-3-methylimidazolium ethyl sulfate (EMIM-ES) filled with fumed silica particles, which serve as a rheology modifier. With their low vapor pressure, non-permeability through elastomeric matrices, and appropriate resistivity for sensing applications, organic ionic liquids have been used previously as sensors in soft robots [240, 241]. To introduce those sensors, an ionic liquid is injected into channels within elastomeric structures produced by multi-step molding and lamination methods [240, 241]. Since EMIM-ES is a Newtonian fluid, we added fumed silica nanoparticles to create a conductive ionogel that exhibits shear-thinning behavior (Figure 5.8A) and a shear yield stress, $\tau_y$, that increases with filler concentration (Figure 5.8B). While the neat EMIM-ES and EMIM-ES filled with 2 wt% fumed silica show no yield stress,
Figure 5.8. Rheological behavior of inks and matrix materials. (A) Log-log plots of apparent viscosity as a function of shear rate and (B) storage modulus, $G'$, as a function of shear stress for suspensions of fumed silica in 1-ethyl-3-methylimidazolium ethyl sulfate at various weight percents of fumed silica (wt%). (C) Log-log plots of apparent viscosity as a function of shear rate and (D) $G'$ as a function of shear stress for all inks and matrix materials used for EMB3D printing the soft actuators.

the 3, 4, 5, 6, and 7 wt% fumed silica inks exhibit shear yield stresses of approximately 7 Pa, 16 Pa, 70 Pa, 150 Pa, and 320 Pa, respectively. Also, the respective plateau moduli, $G'_o$, are 0.1 kPa, 0.5 kPa, 3 kPa, 6 kPa, and 13 kPa. The optimal ionogel for EMB3D printing contains 6 wt% fumed silica particles. The fugitive ink used to print the pneumatic features within these SSAs is composed of an aqueous gel of Pluronic F127, a poly(ethylene oxide)-b-poly(propylene oxide)-b-poly(ethylene oxide) triblock copolymer [308].

Flow (Figure 5.8C) and amplitude sweeps (Figure 5.8D, where only $G'$ are shown for clarity), reveal the shear thinning and yield-stress behavior of the viscoplastic matrix materials and viscoelastic inks. The shear yield stresses of the material components, as determined from the amplitude sweeps in Figure 5.8D
as the cross-over point at which $G' = G''$, are approximately 260 Pa for fugitive ink, 150 Pa for the sensor ink, 30 Pa for the dorsal matrix, 100 Pa for the actuator matrix, and 14 Pa for the anterior matrix. The plateau storage moduli, $G'_0$, are approximately 16 kPa for the fugitive ink, 6 kPa for the sensor ink, 2 kPa for the dorsal matrix, 3 kPa for the actuator matrix, and 0.7 kPa for anterior matrix. Following trends established in earlier chapters, the rheological behavior of the inks and matrices facilitated EMB3D printing of SSAs.

5.3.3 SSA Characterization

Each sensor is composed of a resistive strain gauge whose electrical resistance is given by $R = \rho L / A$, where $\rho$ is the resistivity, $L$ is length, and $A$ is the cross-sectional area of the printed ionogel features. Low-voltage AC readout electronics are used to measure $R$ without complications arising from capacitive charge separation or electrochemical reactions [239]. The change in resistance, $\Delta R$, during operation is given by $\Delta R = R - R_0$, where $R_0$ is the initial resistance. We first probed the effect of relative humidity on $R$ by creating a model sensor embedded within the anterior matrix (Figure 5.9), since the hygroscopic sensor ink uptakes water that permeates through the crosslinked matrices. We observed decreases in $R$ over time that can be attributed partially to increases in cross-sectional area, $A$, due to swelling by water uptake (Figure 5.9C) and the concomitant decrease in $\rho$ in these ionogel-water mixtures [313]. After four days, we find that the sensors reach a steady $R$ value that changes negligibly over time.

Next, we characterized $\Delta R$ for the embedded curvature, inflation, and contact sensors during free and blocked displacement as well as the $\Delta R$ of the contact sensor during the application of a contact pressure. When inflated, the bladders within the SSAs compress the inflation sensor (i.e., $A$ decreases) and elongate the curvature sensor (i.e., $L$ increases) during bending. As shown in Figure 5.10A, for a freely bending SSA, $\Delta R_{\text{curvature}}$, $\Delta R_{\text{inflation}}$, and displacement angle, $\theta$, all increase with inflation pressure, while $\Delta R_{\text{contact}}$ negligibly changes. During blocked displacement, the SSA cannot bend freely
Figure 5.9. Sensor stability. (A) Resistance, $R$, and (B) relative resistance change with respect to the resistance on Day 0, $\Delta R/R_{Day0}$, are shown at different times for printed sensors at three different relative humidity (RH) conditions ($n = 6$ sensors per condition). After Day 20, the sensors are placed in a 90°C oven for five days, with $R$ and $\Delta R/R_{Day0}$ taken at $t = 1$ day and $t = 5$ days. (C) Bright field micrographs of representative sensors from each RH condition are shown on $t = 20$ days (top) and after $t = 1$ day at 90°C (bottom). (Scale bars are all 2 mm.)

when inflated against a rigid obstruction. Correspondingly, $\Delta R_{curvature}$, $\Delta R_{inflation}$, and $\Delta R_{contact}$ all increase, but not in an identical manner as in free displacement: at each respective inflation pressure, $\Delta R_{curvature}$ is noticeably less due to obstructed bending, $\Delta R_{inflation}$ is slightly lower, and $\Delta R_{contact}$ is only slightly positive compared to free displacement values. Using a mass balance as our obstruction, we find increasing inflation pressure drives increased force generation (Figure 5.10B). The representative SSA characterized in Figure 5.10 is capable of generating blocked forces of 0.38 +/- 0.01 N at an inflation pressure of 152 kPa. Finally, $\Delta R_{contact}$ noticeably changes when contact pressures are directly applied to the distal meander of the contact sensor (Figure 5.10C). Similar to observations reported for liquid metal-
Figure 5.10. Sensor characterization. (A) Resistance change, $\Delta R$, for the curvature, inflation, and contact sensors and displacement angle, $\theta$, as a function of inflation pressure during free displacement. (B) $\Delta R$ for each sensor and force generated as a function of inflation pressure during blocked displacement. (C) $\Delta R_{\text{contact}}$ as a function of applied contact pressure. For (A-C), data points and shaded regions represent mean values and standard deviations, respectively, ($n=3$).

based soft sensors [226], we find that $\Delta R_{\text{inflation}}$ and $\Delta R_{\text{contact}}$ are non-linearly related to inflation and contact pressures, respectively, while $\Delta R_{\text{curvature}}$ increases linearly with inflation pressure.

A slight hysteresis in $\Delta R_{\text{curvature}}$ and $\Delta R_{\text{inflation}}$ is observed in the $\Delta R$ readouts over inflation-deflation cycles. We believe this arises from the viscoelastic nature of the elastomeric matrices within the SSA, since it is also observed in liquid metal-based soft sensors [226, 314]. This hypothesis is further supported by the sensors’ $\Delta R$ recorded during dynamic free (Figure 5.11A-B) and blocked (Figure 5.11C-D) displacement trials, in which the SSA undergoes repeated deflation-inflation cycles at 20s intervals and inflation pressure increases at 14 kPa increments. The readout data in Figures 5.11B and 5.11D agree well with the data reported in Figures 5.10A and 5.10B, respectively, and $\Delta R$ for each sensor nearly returns to zero once the SSA is deflated. In the dynamic blocked displacement trial, the SSA is placed on a rigid acrylic rod such that the rod edge provides contact pressures that produce clear increases in $\Delta R_{\text{contact}}$ at higher inflation pressures (Figure 5.11D).

We explored readouts from three simple external manipulation motifs on non-inflated SSAs (Figure 5.11E). Figure 5.11F shows $\Delta R$ for each sensor with time during three upward bends, three downward bends, and three flicks. $\Delta R_{\text{inflation}}$ is negligible, since the SSA is not inflated. During
Figure 5.11. Performance of soft somatosensitive actuators. (A) Images of a SSA at 0 kPa (top) and 152 kPa (bottom) during a dynamic free displacement test, in which the SSA experiences periods of no inflation (0 kPa for 20 s) to increasing inflation pressure (held for 20 s) in increments of 14 kPa to 152 kPa. (B) $\Delta R$ of each sensor is plotted as a function of time. (C) Still images of a SSA inflating against an acrylic rod at 0 kPa (top) and 152 kPa (bottom) during a dynamic blocked displacement test, in which the SSA is actuated via the same inflation sequence as in (A) and (B). (E) $\Delta R$ for each sensor is plotted as a function of time. (E) Upward bend (left), downward bend (middle), and flick-style (right) manipulations carried out on a non-inflated SSA. (F) $\Delta R$ for each sensor is plotted as a function of time for a SSA undergoing a sequence of three upward bends, three downward bends, and three flicks. All scale bars are 20 mm.

upward bending, the contact and curvature sensors lay above and beneath the neutral axis of the SSA, respectively. Hence, the contact sensor elongates, while the curvature sensor is compressed, i.e., $\Delta R_{\text{contact}} > 0$ and $\Delta R_{\text{curvature}} < 0$. During downward bending, these opposite responses are observed, i.e., $\Delta R_{\text{contact}} < 0$ and $\Delta R_{\text{curvature}} > 0$. During a flick-type deformation, $\Delta R_{\text{contact}}$ and $\Delta R_{\text{curvature}}$ mimic the response of a damped harmonic oscillator (Figure 5.12A). Interestingly, this same behavior (Figure 5.11F) is also observed for an inflated SSA (Figure 5.12B). The above data illustrate that the curvature, inflation, and contact sensors embedded within these SSAs provide the desired somatosensory feedback. The curvature sensor correlates with actuator displacement, the inflation sensor indicates whether or not the displacement is intentional, and finally the contact sensor indicates when it is in contact with an object.
Figure 5.12. External SSA manipulation. (A) A plot of $\Delta R$ for the curvature, inflation, and contact sensors as a function of time taken from Figure 5.12F between 40s and 43s. (B) $\Delta R$ for each sensor is reported as a function of time for a SSA undergoing three upward bends, three downward bends, and three flicks in a sequential manner after being inflated to 83 kPa.

5.3.4 Somatosensory Feedback from a Soft Robotic Gripper

To fully demonstrate the utility of this collective somatosensory feedback in a soft robotic system, we integrated three SSAs in a mounting assembly to produce a soft robotic gripper. For these demos, in which the gripper grabs various balls, all three SSAs are inflated identically, and the sensory feedback is collected only from the SSA in the center of the image shown in Figures 5.13 and 5.14. Figures 5.13A and 5.13B contain images and a corresponding plot of $\Delta R$ with time recorded from all sensors, respectively, during a simple object manipulation event. Starting with a non-inflated gripper, the ball is first used to bend the center SSA in an upward then downward manner to simulate interaction with an incoming object. The gripper is then inflated at a modest pressure of 55 kPa to grab the ball. At this inflation pressure, only an obvious increase in $\Delta R_{\text{curvature}}$ was anticipated, though $\Delta R_{\text{inflation}}$ and $\Delta R_{\text{contact}}$ slightly increased. Tugging the ball induces slight variations in $\Delta R_{\text{curvature}}$. At $\sim 22$ s, the inflation pressure is increased to 152 kPa, leading to a concomitant rise in $\Delta R$ of each sensor. Now that the ball is tightly grabbed, clear change are observed in $\Delta R_{\text{curvature}}$ and $\Delta R_{\text{contact}}$ when the ball is tugged again. The ball is pulled completely from the gripper at $t = \sim 34$ s while the inflation pressure is maintained. Without the
Figure 5.13. Interaction with a soft robotic gripper comprised of three SSAs. (A) Images of an interaction sequence between a ball and a soft robotic gripper comprised of SSAs (scale bar is 20 mm). (B) $\Delta R$ of each sensor as a function of time during the interaction sequence shown in (A). (Note: The non-inflated gripper ($t = 0$s) had one SSA bent upwards ($t = 5$s) and downwards ($t = 6$s) before the ball is inserted into the gripper. The gripper is then inflated to 55 kPa ($t = 11$s) to hold the ball ($t = 14$s) even when manually tugged ($t = 16$s). The inflation pressure is increased to 152 kPa for a stronger grip ($t = 23$s). After additional tugging ($t = 26$s), the ball is removed from the gripper ($t = 34$s), which remains inflated at 152 kPa ($t = 37$s). The gripper is deflated at $t \sim 43$s.)

Ball, the SSAs are able to more freely displace, increasing $\Delta R_{curvature}$ and decreasing $\Delta R_{contact}$ to approximately 0 k$\Omega$. We clearly observe the kinesthetic nature of the inflation sensor by removing the ball from the inflated gripper, as $\Delta R_{inflation}$ remains somewhat constant. $R$ for all sensors returned to approximately $R_0$ once the SSAs were deflated.

Contact pressures generated by grabbing this first ball are rarely sufficient to create $\Delta R_{contact} > \sim 100$ k$\Omega$. This is a consequence of the overall design as well as the materials used in these SSAs. By grabbing objects with different textures that create higher contact pressures, we demonstrate that the current SSAs can produce $\Delta R_{contact} > 100$ k$\Omega$ (Figure 5.14A). A plot of $\Delta R$ versus time recorded for all sensors during the study is provided in Figure 5.13B. The grabbing sequence involves applying an inflation pressure of 83 kPa for 15s, followed by an increase of the inflation pressure to 165 kPa for 30s. Note that $\Delta R_{contact}$ increases at this inflation pressure even without an object was in its grasp. When grabbing the smooth object, $\Delta R_{contact}$ is approximately 40 k$\Omega$ at 165 kPa, $\sim 30\%$ higher than when
Figure 5.14. Somatosensory feedback from soft robotic grippers. (A) Still images show the gripper holding nothing (left), a smooth ball (middle) and a spiked ball (right) (scale bar is 20 mm). (B) $\Delta R$ for each sensor is plotted as a function of time. (C) Thermal images of a gripper holding a room temperature (RT) ball, a hot ball ($\sim$60°C), and a cold ball ($\sim$0°C) (scale bar is 20 mm). (D) $\Delta R_{\text{curvature}}$ and $\Delta R_{\text{contact}}$ plotted as a function of time. (E) Still images showing a gripper comprised of SSAs with embedded fine and deep contact sensors holding a foam ball at 97 kPa (P1), 124 kPa (P2), and 152 kPa (P3) (scale bar is 10 mm). (F) The $\Delta R_{\text{curvature}}$, $\Delta R_{\text{fine}}$, and $\Delta R_{\text{deep}}$, are plotted as function of time for the sequence shown in (E).

grabbing nothing, $\Delta R_{\text{contact}}$ reached 300–500 kΩ when grabbing the spiked object, whose texture generated higher contact pressures.

Next, we explored temperature sensing using these SSAs. The ionogel conductivity as a function of temperature is given by the Vogel-Tamman-Fulcher (VTF) equation [311, 313]:

$$\sigma = \rho^{-1} = \alpha \cdot \exp \left( -\frac{\beta}{T - T_0} \right)$$

where $\sigma$ is conductivity, $T$ is temperature, and $\alpha, \beta,$ and $T_0$ are fitting parameters. Hence, the printed sensor $R$ should decrease with increasing $T$ according to:

$$R = \left( \frac{L}{A} \right) \left( \alpha \cdot \exp \left( -\frac{\beta}{T - T_0} \right) \right)^{-1} = \alpha^* \cdot \exp \left( \frac{\beta}{T - T_0} \right)$$

We first validated this behavior over $T = 5^\circ$C–95°C (Figure 5.15A), and then used the soft robotic gripper to grab three identical plastic balls held at room temperature (RT), $\sim$60°C (hot), or $\sim$0°C (in dry ice, cold)
Figure 5.15. Temperature and fine-versus-deep touch sensing. (A) Sensor resistance, $R$, as a function of Peltier temperature. Data points represent the mean $R$ measured and shading indicates standard deviation ($n = 3$). $R$ decreases with increasing temperature according to Equation 4, for which a fit is shown. The fit parameters $\alpha'$, $\beta$, and $T_0$ are $\alpha' = 0.404$ k$\Omega$, $\beta = 610$ K, and $T_0 = 148$ K ($R^2 = 0.9992$). (B) Schematic illustration of fine and deep contact sensors in a SSA. (C) Resistance change, $\Delta R$, as a function of contact pressure for the fine and deep contact sensors. (D) Stills show manual pressing of the fine contact (left), deep contact (middle), and both sensors (right). (E) The corresponding readouts of $\Delta R$ from the fine and deep contact sensors are plotted as a function of with time.

by inflating the gripper to 83 kPa for 15s then at 165 kPa for 30s (Figure 5.14C). $\Delta R$ is plotted as a function of time in Figure 5.14D for these conditions. When grabbing the RT ball at 165 kPa, $\Delta R_{\text{contact}}$ is approximately 9 k$\Omega$ greater than that for when the gripper holds nothing at 165 kPa. When grabbing the hot ball, $\Delta R_{\text{contact}}$ decreases noticeably, even becoming negative, due to the local increase in the contact sensor’s conductivity where the distal meander made contact with the ball. Finally, when grabbing the cold ball, a clear increase in $\Delta R_{\text{contact}}$ is observed that exceeds the value of $\Delta R_{\text{contact}}$ for the same ball held.
at RT. When the gripper releases the hot and cold balls, $\Delta R_{\text{contact}}$ does not immediately return to the value of 0 k$\Omega$ measured at RT.

As a final demonstration, we created SSAs with both fine and deep contact sensors, which are printed with identical widths of the original contact sensor, but with shorter lengths and at different heights within the anterior matrix (the fine contact sensor is positioned closer to the surface of the anterior matrix) (Figure 5.15B). Importantly, in this dual design, $\Delta R$ of the fine contact sensor in response to an applied contact pressure is higher than that observed for the deep contact sensor (Figure 5.15C). The fine and deep contact sensors also have different receptive fields, akin to human fingers (see Figure 5.15D-E). To illustrate this, a soft foam ball is grabbed at three different inflation pressures, 97 kPa (P1), 124 kPa (P2), and 152 kPa (P3) (Figure 5.14E). $\Delta R$ for the fine ($\Delta R_{\text{fine}}$) and deep ($\Delta R_{\text{deep}}$) contact sensors are provided in (Figure 5.14F) for these conditions, alongside a control experiment in which the gripper grabs nothing. For each inflation pressure, $\Delta R_{\text{fine}}$ is negative or $\sim$0 k$\Omega$ when the gripper grabs nothing and increases noticeably when grabbing the ball. The deep contact sensor is less sensitive, but $\Delta R_{\text{deep}}$ is still approximately 8 k$\Omega$, 10 k$\Omega$, and 12 k$\Omega$ greater when grabbing the ball at P1, P2, and P3, respectively, than when grabbing nothing. The ability to integrate multiple contact sensors sensitive to different modes of contact may facilitate handling of extremely delicate objects.

5.4 Conclusions

In summary, we have shown that our EMB3D printing method and accompanying materials palette enables the construction of soft robotic actuators with conductive features that emulate the human somatosensory system. By embedding multiple sensor networks within a molded elastomeric matrix, we created soft robotic actuators with haptic, proprioceptive, and thermoceptive sensing. The sensors consist of a conductive ionogel that is both stable long-term and hysteresis-free, while the pervasive pneumatic channels that drive their actuation when inflated are patterned using a fugitive ink. Our approach
represents a foundational advance that may find potential application in soft robotic, wearable, and haptic devices [272] requiring embedded soft sensing for closed-loop control.
CHAPTER 6

CONCLUSIONS

My Ph.D. dissertation has led to the development of new functional inks and elastomeric matrix materials specifically tailored for embedded 3D (EMB3D) printing of autonomous and somatosensitive soft robots. Fugitive, catalytic, and sensor inks can be rapidly and arbitrarily printed in the viscoplastic matrices to create embedded features for actuation or fuel transport, fuel decomposition, and sensor feedback, respectively. Using EMB3D printing, I created the Octobot, which is an entirely soft robot that has no dependency on external power and control hardware, as well as soft somatosensitive actuators (SSAs) that consist of fluidic elastomer actuators that are innervated with soft sensors for multiple somatosensory feedback motifs. The primary findings from my Ph.D. research are highlighted below.

1. **Viscoplastic matrices enable crevice-free EMB3D printing**

   Matrix materials with viscoplastic behaviors have been developed and characterized for EMB3D printing [18, 19]. Several matrix materials – the Octobot’s body matrix and the SSAs’ dorsal, actuator, and anterior matrices – showed clear thixotropic behavior and enabled us to print the respective functional features within these matrices. The body matrix’s thixotropic behavior was tailored through enrichment of a PDMS blend with fumed silica nanoparticle fillers that form a percolated microstructural network. The SSAs’ matrices were formulated from viscous, off-the-shelf silicone elastomers. Their thixotropy was achieved using commercial available, proprietary additives that impart thixotropic behavior by increasing intermolecular bond formations throughout the silicone blend. During nozzle translation, the matrix materials are locally yielded, which leads to their temporarily fluid nature. These materials exhibit “self healing,” i.e., they flow behind the nozzle such that no crevices are formed. After restructuring over time, the matrix materials return to their equilibrium rheological behavior.
2. **Maximizing Oldroyd number is key to achieving high fidelity EM3D printing**

With printing demos, matrix yielding studies, and particle image velocimetry (PIV) experiments conducted with our collaborators in Prof. Howard Stone’s Complex Fluids Group at Princeton University, we identified that maximizing the dimensionless Oldroyd number, $Od$, is crucial to successful EMB3D printing. We found that $Od$ is most notably influenced by matrix material rheology, with higher $Od$ generally observed for matrix materials with higher shear yield stress and consistency index, and lower flow index. $Od$ also increases when the effective shear rate, $V/d$, decreases, where $V$ and $d$ are the print speed and nozzle diameter, respectively. Consequently, highest print fidelity is achieved by selecting a print path under high $Od$ printing conditions (i.e., $Od > 1$) that minimizes the exposure of previously printed features to the yielding, fluidized matrix material region around a translating nozzle.

3. **Microfluidic logic and monopropellant decomposition serve as soft analogs to conventional robotic control and power hardware for entirely soft, autonomous robots**

Our embedded soft controller with microfluidic logic and on-board decomposition of a monopropellant fuel supply were key features of our entirely soft Octobots that led to the elimination of traditional electronic controllers and power sources, respectively. Central to this design strategy was identifying that a liquid monopropellant fuel could serve as an “operand” on which a soft microfluidic controller could operate and, in turn, temporally regulate for selective decomposition. The soft controller was inspired by a previous microfluidic oscillator design [21]. Additional check valves and a new layout were incorporated to facilitate fuel injection into the Octobot without leaking into the ambient environment, prevent flow of gas from the decomposed fuel back upstream into the soft controller, and integrate with the overall Octobot form. With a high energy density (1.44 kJ/g at 50wt%, as compared to 0.1–0.2 kJ/g for batteries) and benign byproducts (oxygen, water, and heat) [22], hydrogen peroxide ($\text{H}_2\text{O}_2$) served as a useful monopropellant fuel that enabled us to inflate the Octobot’s eight fluidic
elastomer actuators. This fuel simply needed to be exposed to a catalyst (i.e., in our Pt-laden, catalytic reaction chambers) for decomposition into pressurized gas, which obviates any need for battery-operated hardware, such as the pump and valve setups needed to regulate pressurized working fluid sources or convective mixing and ignition systems necessary for inflating FEAs via combustion reactions [22].

4. Multi-material, EMB3D printing facilitates material assembly across multiple materials and length scales

By integrating soft lithography and molding with our EMB3D printing method, we realized basic autonomy in a completely soft, untethered robot. The microscale features (~$10^1$ – $10^3$ µm) of our embedded soft controller were patterned with soft lithography, and our printing molds defined the macroscale features (~$10^2$ – $10^5$ µm) of the elastomeric robots. EMB3D printing enabled us to rapidly pattern the discrete, complex, multifunctional networks with micro- and mesoscale features (~$5\times10^1$ – $2\times10^3$ µm) that served as the Octobots’ fuel reservoirs, interconnects with the soft controller, actuator networks, catalytic reaction chambers, and vent orifices. Fundamentally, EMB3D printing offers a rapid, seamless approach to multimaterial fabrication compared to other multimaterial 3D printing methods (see Chapter 2). While six different materials comprise the Octobot, only two of these had to be printed (i.e., the fugitive and catalytic inks). Two of these were molded and used as matrix materials, and the other two materials were used to create the Octobot actuators’ hyperelastic layers and soft controller.

The auto-evacuation of the fugitive ink and the design of our fugitive plug materials that were filled and cross-linked within the soft controller were keys to successfully creating our Octobots. By relying on auto-evacuation (i.e., the permeation of the water in the fugitive ink through the crosslinked body matrix), no additional steps were required to remove our fugitive ink as in previous works [18, 169], greatly simplifying the Octobot assembly process. Finally, the crosslinked fugitive plugs prevented matrix material ingress into the soft controllers, which otherwise resulted in often catastrophic blockages.
As a mixture of Pluronic F127 and F127-DA, the fugitive plug was only weakly crosslinked and could auto-evacuate along with the Pluronic F127 fugitive ink.

5. **Ionogel-based soft sensors exhibit reliable performance and long-term stability**

Typical approaches to 3D printed soft sensors involve the design of printable suspensions of conductive particles. The rheological properties of these suspensions are tuned through particle morphology, concentration, and the addition of polymeric additives, carrier fluids, and/or solvents [8, 19, 220]. While this is an intuitive strategy to conductive ink design, we took an inverse approach to designing hysteresis-free sensor inks for the SSAs by aiming to rheologically tailor a conductive fluid with non-conductive rheology modifying additives. Since liquid metals pose many challenges and toxicity concerns, we developed organic ionogels as a conductive ink. While other groups have used nanoparticles or triblock copolymers to create viscoelastic, yield-stress ionogels from organic ionic liquids, we employed fumed silica as an inexpensive rheology modifier [315, 316]. Specifically, an ionogel sensor ink was designed using 1-ethyl-3-methylimidazolium ethyl sulfate as the organic ionic liquid, and 6 wt% fumed silica provided the ink with a shear yield stress of ~150 Pa and a plateau storage modulus of ~6 kPa, values that are appropriate for EMB3D printing in the SSA matrix materials. Based on humidity experiments and dynamic actuation studies, we found that these inks displayed long-term stability without hysteresis. Even after five days at elevated temperatures of ~90°C where aqueous ionic conductors would lose water content due to evaporation, our printed sensors are stable.

6. **Somatosensory actuators that provide proprioceptive, kinesthetic, and tactile feedback**

Using EMB3D printing coupled with our materials palette, we fabricated fluidic elastomer actuators that were innervated with three types of soft sensors in a single manufacturing step. Specifically, curvature, inflation, and contact sensors were printed to enable multiple, fundamental
modes of somatosensory feedback. As these SSAs move, the curvature sensor is the primary indicator of how and to what extent this motion is occurring. The inflation sensor indicates whether or not SSA displacement is intentional and to what extent the SSA could be displacing. Finally, the contact sensors, sensitive to contact pressures applied to the sensor’s distal meander, obviously change in resistance when they make contact with an object. Hence, resistance changes in the curvature, inflation, and contact sensors provide the SSAs with proprioceptive, kinesthetic, and tactile feedback, respectively.

7. **EMB3D printed ionogel sensors enable thermoceptive and fine-and-deep touch sensing**

Beyond fundamental proprioception, kinesthesia, and tactile feedback, we explored new modes of somatosensory feedback for soft robotic grippers. According to the Vogel-Tamman-Fulcher (VTF) equation, the resistances of our printed contact sensors should decrease in resistance with increasing temperatures. We showed that this is true with simple strain sensors in contact with a substrate of temperature varied from 5˚C to 95˚C. Across this temperature range, the sensors resistance changed ~200 kΩ in good agreement with the VTF equation \[311\]. By grabbing three spherical objects at room temperature, ~0˚C, and ~60˚C with our SSA-based gripper, we saw clear indication of thermoceptive sensing from the SSAs’ contact sensors.

Finally, making use of the free-form patterning nature of EMB3D printing, we printed SSAs with two distinct contact sensors – i.e., fine and deep contact sensors – with different areal dimensions and at different heights within the anterior matrix material. The fine contact sensor was narrower than the deep contact sensor and was positioned closer to the surface of the anterior matrix than the deep contact sensor. The fine and deep touch sensors had different receptive fields, akin to our own bodies’ various specialized touch receptors \[200\]. Due to the differences in their forms and print heights within the SSA anterior matrix \[317\], characterization of the fine and deep contact sensors’ resistance change with respect to applied pressures clearly showed that the fine contact sensor was more sensitive to contact pressure.
Fundamentally, these results show that multiple contact sensors sensitive to fine and deep touch could be utilized for monitoring successful and robust grasps, slip detection, and well beyond.
CHAPTER 7

CHALLENGES AND OPPORTUNITIES

My dissertation findings present new challenges and opportunities for future research efforts, alike, for EMB3D printing as a soft matter assembly method, next-generation entirely soft robots, and other sensorized soft robotic systems. These are highlighted below.

1. **New matrices, hardware, and tool-path optimization strategies for EMB3D printing**

   Our new insights into EMB3D printing open new opportunities for material, hardware, and software innovations for this free-form manufacturing method. In this work, only off-the-shelf products were utilized for matrix material formulations. New materials and enabling chemistries will expand the effective pot-life of our thixotropic silicone matrices, facilitate new matrix curing methods, and provide new matrix material properties altogether. One promising approach to designing new silicone matrices that provide high 0d conditions and crevice free printing would be through the design of curable suspensions of granular microgels comprised of crosslinked silicone [173].

   Current inconveniences in EMB3D printing workflows include the need to manually register print nozzles to various mold or soft controller features as well to pour in and degas matrix materials. New hardware and software advances that automate these steps will dramatically improve the simplicity of the EMB3D printing method and could pave the way to completely automated print processes.

   EMB3D printing fundamentally allows one to freely print in three dimensions within an arbitrarily large reservoir volume. At present, current slicing algorithms and tool-path generators for ordinary layer-by-layer 3D printing methods are not appropriate for this new printing approach, and GCode commands for EMB3D printing must be manually generated. New slicing and tool-path optimization algorithms specifically designed for EMB3D printing will greatly improve the translation of digital object files to appropriate GCode for printing.
Finally, we anticipate interesting opportunities for using multi-axis robotic arms as translation stages towards optimizing print fidelity in this new printing method. With tool-path design so crucial to achieving desired print results in EMB3D printing, six-axis robotic arms could use highly optimized tool-paths for nozzle translation that are superior to those allowed by our 3-axis printer stages and minimize exposure of previously printed inks to yielding matrix materials [318, 319].

2. More sophisticated soft microfluidic controllers

Our integrated design and fabrication approach for entirely soft robots enables a wide variety of existing and novel microfluidic devices to be seamlessly introduced integrated into the assembly process. Thus, future entirely soft robots can be designed with more sophisticated microfluidic control systems needed for more advanced actuation and gaiting schemes. Specific improvements to system performance that could come with such advances will include higher frequency actuation and venting cycles, multi-degree-of-freedom actuators, more complicated gait cycles enabled by planned actuator motions, and even mechanisms for incorporating true closed-loop feedback control based on external mechanical stimuli from the environment.

3. Powering entirely soft robots via monopropellant decomposition

The Octobots rely on Pt-laden features printed with our catalytic ink for fuel decomposition and ensuing production of pressurized gas. To maintain reaction chambers with high network integrity, photocrosslinkable additives (i.e., Pluronic F127-DA, and PEG-DA) were used. At H\textsubscript{2}O\textsubscript{2} fuel concentrations above ~50 wt\%, we observed combustion of the Pluronic F127-DA and PEG-DA and chose to cap our fuel concentration at ~50 wt\%. With fuels of higher H\textsubscript{2}O\textsubscript{2} concentration, we may have realized several benefits for the Octobots, including faster actuation, larger actuator displacements, and increased switching frequencies between the two actuator networks. New catalytic ink designs that bypass the use
of organic components that combust at high fuel concentrations will open new opportunities for future entirely soft robots. Finally, another limitation of using monopropellants to power entirely soft robots is that they must be refueled. This is a clear hindrance to designing entirely soft, autonomous robots for deployment, but it begs an exciting question: could an entirely soft robot produce its own fuel, akin to all living organisms, by scavenging chemical precursors or solar energy that could participate in the on-board synthesis of new H$_2$O$_2$ or an alternative but suitable fuel?

4. *Design strategies for next-generation entirely soft robots*

While the Octobot is a minimal system serving as a first example of an untethered, completely soft robot, future developments in microfluidic logic design, actuator design, and fabrication will facilitate the creation of more sophisticated completely soft robots. Improvements in EMB3D printing fidelity and materials designs could be used to replace soft lithography altogether, and soft controllers could be directly EMB3D printed in soft robots. With appropriate resins, light-based 3D printing approaches such as two-photon polymerization could be used to print compact, highly intricate soft controllers or even sub-centimeter entirely soft robotic systems, with catalysts and microfluidic controllers directly integrated. Through elaboration on the design strategies we’ve presented here, one can imagine many opportunities for entirely soft robots. Without the need for conventional hardware, it will be exciting to explore scaling limitations in entirely soft robots with form factors far smaller than the Octobot prototype.

Following our strategies, new entirely soft robotic systems will require careful balancing of fuel supply, fuel flow rate, downstream fuel decomposition and expansion of gaseous byproducts, venting, and actuator requirements. Topology optimization and computational fluid dynamics will be essential to rationally designing next-generation entirely soft robots and streamlining the system-level design process. However, we expect that forthcoming advances in soft electroactive polymer and liquid crystal
elastomer actuators, printable soft energy storage and generation materials, and additive manufacturing will enable entirely new integrated design and fabrication approaches for entirely soft robots that utilize alternatives to soft microfluidic controllers and power via chemical reactions.

5. *Improved feedback and control for soft somatosensitive actuators*

EMB3D printing enables the fabrication of multi-material, soft robotic actuators with embedded soft sensors with hysteresis-free conduction in a single manufacturing step. With the current soft sensors innervating our SSAs, the soft robotic actuators we have created possess multiple, fundamental modes of somatosensory feedback that will find utility for creating autonomous soft robotic systems controlled via closed-loop feedback strategies. At present, we have only printed four sensors in the SSAs, but any number of sensors could be introduced and distributed throughout these actuators. Immediate avenues for investigation in the current SSA design include (i) the introduction of capacitive sensor designs, sensors sensitive to other modes of deformation, and arrays of contact sensors for higher resolution tactile sensing, (ii) optimization of the fluidic actuator’s architecture (i.e., bladder spacing, bladder number, and more), and (iii) improved mechanical interfacing between the soft sensors and rigid leads to read-out electronics. Future SSA designs will benefit from additional inks, such as inks for EMB3D printing rigid features that improve tactile sensitivity and other conductive inks for printing high conductivity leads to the resistive sensors. New control algorithms should be established for closed-loop feedback control of next-generation SSAs, and machine learning could be harnessed to simplify these control algorithms.

6. *Applications*

If more sophisticated soft controllers could provide complex sensory capabilities, future entirely soft robots like the Octobot could find use in search and rescue missions where environments are harsh or difficult to traverse or navigate, utility in space and marine robotic applications, wearable robotic systems
for rehabilitation or augmented performance, microscale robotic applications, energy harvesting or communications applications, and more. One major advantage of future entirely soft robots would be their low cost. Based on materials cost analyses of our current Octobot, these robots could be produced at < $1 (USD) for materials per device. This could enable entire swarms of soft robots to be cheaply manufactured and deployed for scenarios where the robot is not expected to return or be reused.

Our EMB3D printing methods for creating soft somatosensory actuators address the more immediate need of creating sensorized soft robots. Next-generation soft robots require integrated soft sensors capable of providing discrete, somatosensory feedback motifs, including proprioceptive, haptic, and thermoceptive sensing, to realize their full potential in automation, manufacturing, personal robotics, autonomous deployable systems, and all other applications reviewed throughout this Dissertation. Advances in machine learning and computation will be essential for developing control strategies for sensorized soft robots and could, among other capabilities, utilize their embedded, distributed soft sensors for a variety of tasks, including object [132], texture [129], and gesture recognition [272], environmental sensing, and more [2, 3].
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