Soft Actuator Materials for Electrically Driven Haptic Interfaces

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Haptics involves human touch sensing and tactile feedback and plays a crucial role in physical interactions of humans with their environment. There is an ever-increasing interest in development of haptic technologies, due to their role in various applications such as robotics, virtual and augmented reality, healthcare, and smart electronics. Electrically driven actuation mechanisms for soft materials like dielectric elastomer actuators (DEAs), electrohydraulic soft actuators (ESAs), ionic polymer–metal composites (IPMCs), and liquid crystal elastomers (LCEs) hold the potential for the development of the next generation of haptic feedback devices due to a variety of advantages such as light weight and compact design, untethered activation and control, large actuation strains, and distributed and localized actuation. Herein, a detailed look is taken at the advancement in material designs for these electrically driven soft actuators. A detailed analysis of the different strategies for improving the electromechanical performance of existing material systems is presented. Approaches adopted to synthesize novel material systems are explained. Advancements in compliant electrode materials for the electrically driven soft actuators are also described. The conclusion reflects on the main challenges in the field and provides perspectives on recent advancements expected to have a significant impact.

1. Introduction

Haptics encompasses human touch sensing and feedback via kinesthetic (force/position) and cutaneous (pressure) receptors, associated with perception and manipulation. It includes real and simulated tactile interactions among humans, machines, and objects in real, remote, or virtual environments. Amid different perception modalities in humans, tactile feedback plays a significant role in physical interactions with the environment. The sense of touch enriches our life by providing information about contact with different objects and helps us navigate unstructured environments (tactile exploration). The nervous system, musculoskeletal system, and skin enable tactile sensing and feedback. Among these, the skin plays a key role in tactile feedback. The receptors on the skin perceive different physical information, like hardness (rigid/soft), temperature (warm/cold), roughness (smooth/rough), and friction (sticky/slippy), and provide feedback to the central nervous system. Haptic perception is comparable with vision and hearing in terms of complexity and enables the dexterity of the human hand in grasping and manipulation. Tactile feedback allows sorting of physical data faster than vision and hearing. There is an ever-increasing interest in the development of haptic technologies, motivated by their strategic and critical role in an assortment of applications, such as robotics, virtual and augmented reality (AR), healthcare, and smart electronics.

To this end, several tactile displays and tactile feedback devices have been developed. These tactile feedback devices are based on different technologies such as electrostatics, piezoelectrics, Peltier elements, surface acoustic waves, air jets, pneumatics, electrorheological and magnetorheological fluids, microfluidics, and electroactive polymers. However, most of the current generation of tactile feedback devices aim to simulate the perception of texture change via sensory manipulation; certain technologies aim to create actual topographical changes.

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The intricacy of tactile sensing in humans is a result of a complex overlay of biological structures, resulting in extraordinary performance in detecting distributed forces and displacements, with high spatiotemporal resolution and a wide dynamic range.
There is a huge gap between expected and actual performance of the current generation of tactile displays. This can be attributed to a number of factors. 1) Majority of the existing haptic feedback devices make use of rigid components. The biomechanical properties of the skin, and in particular its compliance (Figure 1B), are key for its haptic capabilities. However, these stiff engineered interfaces prevent the very nature of the material to accommodate the contact and transmit tactile information efficiently. 2) Furthermore, the devices require cumbersome and careful assembly of several hundreds of these rigid components to produce desired tactile feedback, leading to fabrication complexity and higher costs. 3) Realization of natural tactile experiences, pertaining to human sense of touch, remains a challenge as most of the current generation of haptic feedback devices provide only limited unidimensional feedback in a limited range.

An emerging field of research, soft robotics, promises to mitigate these concerns. Inspired by the concepts of embodied intelligence and morphological computation, soft robots make use of compliant materials to optimize the interaction with the environment, while reducing the computational cost of the control system. Quite often, the topology, morphology, and material properties of soft robots are bioinspired, with mechanical properties similar to that of biological tissues, such as the skin. Furthermore, they can respond to external stimuli like electricity, pressure, humidity, pH, temperature, and light to produce tangible responses in shape or volume. These compliant materials have contributed to the emergence of tactile feedback devices based on soft materials. The significant impact of utilizing soft materials is that it provides a perfect mechanical compliance with the human skin and conforms to the shape and matches the dexterity of human fingers. In addition, they also hold the potential to simplify the design and assembly process. Different strategies like pneumatics, fluidics, varying magnetic fields, phase transitions, thermally driven mechanisms, electrically driven mechanisms, and light-driven mechanisms have been pursued for achieving desired actuation in soft materials (Figure 1Q).

Among these, electrically driven actuation mechanisms hold a special place. This can be attributed to a variety of advantages offered by them such as light weight and compact design, unthethered activation and control, large actuation strains, and distributed and localized actuation. These advantages make them highly suitable for meeting the demanding performance required from the current generation of tactile feedback devices.

This Review focuses on electrically driven actuation mechanisms for soft materials that have the potential to be utilized in the field of haptics with an emphasis on the various enabling material classes. First, we review the fundamentals of human touch system to understand the basics of tactile sensing in humans and then discuss the significant role of haptics in various technological fields. Next, a wide range of actuation principles behind the electrically driven actuation mechanisms for soft materials are reviewed. Thereafter, we take a detailed look at the advancement in material designs for these electrically driven soft actuation mechanisms followed by a thorough analysis of the different strategies for improving the electromechanical performance of existing material systems along with the approaches taken to synthesize new material systems. A detailed report on the significant advancements in electrode materials for the electrically driven soft actuators has also been described.

We conclude by providing perspectives on recent advancements expected to have significant impact on the field of electrically driven actuation mechanisms and haptic feedback devices.

2. Fundamentals of Haptics

Sense of touch allows us to interact with different objects in our environment and discern various physical properties of the object like shape, size, temperature, and surface texture. It allows us to maintain stable grasp of objects and also allows us to roll objects between our fingers without dropping it. Apart from our interaction with objects, sense of touch also allows us to interact with the environment and helps in maintaining spatial orientation. Before designing any haptic technology or identifying an existing technology for tactile feedback, it is imperative to understand the “sense of touch” in humans. The following section provides a brief insight into physiology of human touch, spatiotemporal thresholds of mechanoreceptors in human fingers, role of tactile sensing in perception, and importance of mechanical compliance matching of haptic technologies with human skin.

2.1. Physiology of the Human Touch and Spatiotemporal Resolution

Human touch encompasses spatiotemporal perception of different environmental stimuli through a scattered network of somatosensory receptors (mechanoreceptors—for pressure/vibration, thermoreceptors—for temperature, and nociceptors—for pain/damage) all over the body. Mechanoreceptors are encapsulated receptors, embedded within the human skin at different depths. The skin is multilayered and heterogeneous with viscoelastic properties, which vary between the different layers. The mechanical properties of the skin are important in tactile perception. The skin also acts as an interface for contact indentations and conveys information to the receptors. Mechanoreceptors are responsible for mediation of mechanical stimulus. Noticeceptors and thermoreceptors are attributed as free nerve endings because the unmyelinated terminal branches of these neurons branch out widely in the upper regions of the dermis and epidermis. Within the human hand, spatial distribution of the mechanoreceptors is high near the fingertips and low at the palm region. There are four primary types of mechanoreceptors: Meissner’s corpuscles (fast adapting [FA] I), Pacinian corpuscles (FA II), Merkel’s disks (slow adapting [SA] I), and Ruffini’s corpuscles (SA II) (Figure 1A). FA receptors (or rapidly adapting receptors) respond maximally but momentarily to the mechanical stimuli but the response decreases when the stimulus is preserved. SA receptors (or tonic receptors) keep responding as long as the stimulus is present. SA I and FA I receptors are distributed closer to the human skin and have higher densities compared with SA II and FA II receptors. Table 1 shows the properties and spatiotemporal thresholds for different mechanoreceptors in human skin. The interlinking of epidermis and dermis layers in the skin prevents their relative sliding and acts as a filtering mechanism for distributing stresses from contact point to the receptors. This plays a crucial role in determining the spatial resolution of receptors.
There are spatiotemporal limits and sensitivity ranges for somatosensory receptors in detecting various mechanical stimuli. This, in turn, directly affects our ability to perceive different objects and patterns and responsiveness to directional stimuli. Spatial resolution is the least possible separation the human hands can discriminate between two contact points.\(^\text{[27]}\) Spatial resolution for the similar receptors varies across different regions of the body, highest being in places like fingertips, where it can discriminate between two points as close as 1 mm.\(^\text{[28]}\) Pressure threshold and skin deformation are other accepted metrics of absolute tactile sensitivity.\(^\text{[29]}\) Pressure threshold differs with different body parts, and higher value of pressure threshold translates to lower tactile sensitivity.\(^\text{[29]}\) There is a difference of three times in the normal mean threshold values between palm and fingertips, with fingertips having much lower pressure threshold than rest of the body.\(^\text{[4]}\) Temporal resolution for receptors in humans allows for sorting of events 1.4 ms apart (discrimination of vibrations up to 700 Hz).\(^\text{[10]}\) Our receptors can successfully distinguish between two distinct contact events at distinct locations occurring at an interval of 30–50 ms, and this temporal separation allows us to feel the presence of multiple events.\(^\text{[13]}\) Spatial and temporal resolutions are inversely dependent on each other.\(^\text{[4]}\) The underlying reason for the inverse relationship between spatial and temporal resolutions is because of the respective contributions of SA1 and RA fibers to spatial perception.\(^\text{[32]}\) At high frequency, RA signal interferes with SA1 signal in a spatial acuity task, which affects the spatial resolution.\(^\text{[32]}\) Pressure threshold and skin deformation are other factors affecting the absolute tactile sensitivity.\(^\text{[4]}\)

### 2.2. Role of Tactile Sensing in Perception

Both cutaneous and kinesthetic sensing play an important role in discerning cues such as material properties and shape of the object in contact.\(^\text{[5]}\) Cutaneous sensing is better suited to discern material properties, especially when the object is larger compared with the scale of fingertip.\(^\text{[24]}\) Mehanoreceptors play an important role in shape detection of an object, dimensions lying within the size limit of contact area (7–12 mm) of the fingertip.\(^\text{[13,34]}\) The information about the object’s shape and orientation is signaled by the receptors with a major contribution from SA receptors. Object’s curvature and directionality of contact force can also be discerned from these signals.\(^\text{[35]}\) Location of mehanoreceptors within the human skin is near the intermediate ridges—the undulating epidermal tissues that descend into the epidermal–dermal junction.\(^\text{[4]}\) Intermediate ridges amplify the tactile signals from surface of the skin and concentrate the stress, in turn improving the ability of receptors to discriminate finer details of the object.\(^\text{[36]}\)

Ascertaining the surface roughness of an object is another crucial task for the receptors. Both spatial and temporal parameters play a role in perception of surface roughness.\(^\text{[37]}\) Fingerprints help us in discerning finer textures.\(^\text{[58]}\) Tangential motion between the object and skin, in either active or passive touch, also contributes to improving the ability of our receptors in perceiving the surface roughness.\(^\text{[39]}\) Surface roughness is closely related to friction, and this relation becomes stronger when tangential forces come into consideration.\(^\text{[40]}\) Receptors in our skin also play a crucial role in detection of slip, which is defined as the relative motion between an object and skin.\(^\text{[41]}\) Detection of slip is relevant for discerning surface roughness and adjusting the gripping force.\(^\text{[32,43]}\) Tactile feedback from an object affects the estimation of force required to support them. They provide information necessary for controllingprehensile forces for grasping objects.\(^\text{[44]}\) Discerning tactile information about shear helps in adjustment of the gripping force.\(^\text{[45]}\) Tactile feedback provides continuous information about the actual state of the system to our central nervous system.\(^\text{[46]}\) For instance, an object manipulation task can be visualized as a system constituted of several states like reaching, loading, lifting, holding, replacing, and unloading.\(^\text{[4,47]}\) Somatosensory receptors have different responses to each of these states, which helps our central processor to complete the manipulation task. This tactile information also helps in optimizing the temporal course of various stages of a manipulation task.\(^\text{[4]}\)

Discussion in the current section sheds light on the physiology of human touch and role of mechanoreceptors in deciphering different tactile information. It provides spatiotemporal thresholds for different mechanoreceptors. Somatosensory receptors distributed within the skin, along with the soft outer skin layer,
are significant for receiving the tactile information like shape, orientation, hardness, surface roughness, etc. The crucial role of human skin in gathering the tactile information and conveying to the central nervous system has been mentioned. This section establishes a range of performance parameters any suitable haptic technology must meet. In addition, it also emphasizes the mechanical compliance necessary between these technologies and human skin. Moving ahead, it is important to understand the role of haptics in different applications and the role of haptics in enriching user experience in physical and simulated environments.

3. Application Areas of Haptics

3.1. Virtual Reality/Augmented Reality

Virtual environments have broadened the dimensions of reality. They can provide users with an immersive experience, exposing the senses to an array of previously elusive experiences.\(^\text{3.14}\) There are broadly two main pathways to assimilate the virtual environment with the real world. The first approach is virtual reality (VR) (Figure 2A), where the user is immersed entirely in an artificially constructed environment and interacts with the virtual environment via haptic interfaces.\(^\text{4.48}\) The other approach is AR (Figure 2B), where the physical world is reinforced with virtual elements.\(^\text{4.49}\) The aim is to enrich the information flow provided to the user by augmenting the elements in the real world, rather than replacing it with a virtual environment.

Applications such as surgical training, rehabilitation, and physical therapy stand to benefit from developments in the field of VR/AR.\(^\text{3.14}\) For an immersive user experience in both VR and AR environment, it is necessary to conceive components of the virtual world utilizing elements of sensing, control, and haptic feedback, to recreate experiences of physical reality in a credible manner. This has remained a challenge till date, and haptics is expected to play a crucial role. Haptic feedback devices in these applications are expected to deliver feedback to the user persistent with the expectations of the objects and physical interactions in the real world. Furthermore, a highly immersive experience can be achieved for the users in VR/AR environments with haptic information complementing the audio-visual cues.

3.2. Microrobotics

Applications of microrobotics to conduct tasks at a microscale, such as precise manipulation and positioning, pick-up and distribution of objects such as cells, and telerobotic surgery, have gained widespread interest.\(^\text{3.50-52}\) Haptics is especially important in surgical microrobotics such as telesurgery (Figure 2C,D), endoscopic/laparoscopic surgery, single incision, or natural orifice surgery, where the robot applies forces to soft tissues.\(^\text{3.53,54}\) Human operators are expected to control these microrobots for conducting the tasks safely and responsibly. To this end, the human operator should receive consistent information about the interaction of the microrobot with the remote environment to maintain definitive control.\(^\text{113}\) Haptic feedback is one of the critical contributors to this information flow. Haptic feedback is effective in delivering the mechanical characteristics of the objects and features in the remote environment, while providing the information necessary for the navigation in an unstructured setting.\(^\text{113}\)

3.3. Touch Surfaces

In the past couple of decades, there has been an explosion of touch-sensitive devices such as smartphones, tablets, and display kiosks in home appliances and automobiles. Human–machine interaction comes to the forefront in these touch-intensive devices for an immersive experience.\(^\text{8}\) Haptic feedback devices, commonly known as surface haptics, are important for these interactive touch surfaces.\(^\text{12}\) Haptic feedback in these devices aims to provide the user a tactile feedback when interacting with the device (Figure 2E,F).\(^\text{55,56}\) Implementing haptic solutions to these touch-sensitive devices is an up-and-coming field of research and poses new kinds of challenges. Majority of these touch devices aim to communicate visuo–audio–haptic information to the user. This requires seamless integration of the haptic feedback devices, which necessitates a compact and sleek form factor. Also, energy efficiency is desired as these touch devices are portable. In addition, localized feedback is desired to convey the location of point of interaction to the user for efficient navigation.

3.4. Assistive Technologies for Visually Impaired

Individuals with visual impairment have extreme difficulty in navigating through daily activities.\(^\text{115}\) There are varying degrees of visual impairment, and sensory substitution systems are needed to assist these individuals. Visually impaired subjects rely heavily on their physical interactions with objects and beings to construct meaningful experiences. Haptic feedback becomes crucial for them for communication purposes as well. Alphabet systems such as Braille (tactile writing system) (Figure 2G,H) play a vital role in the lives of these visually challenged individuals to assimilate new information and rehabilitate them to some normalcy in their daily life.\(^\text{157,58}\)

It is evident from the physiology of the human touch system and the applications where haptic feedback plays a crucial role that compliance matching between the tactile feedback device and human skin, untethered actuation, and form factor of the devices are as critical as the static and dynamic physical response produced by these haptic feedback devices. Electrically driven actuation mechanisms for soft materials appear to be well placed to meet these challenges.

4. Mechanisms for Electrically Driven Actuation

Recently, there have been several demonstrations of such electrically driven soft haptic devices (Table 2). Our group demonstrated surface texture changes for haptic feedback using a transparent device in thickness-mode dielectric elastomer actuator (DEA) configuration (Figure 3A).\(^\text{3.8}\) Leroy et al. demonstrated complex haptic devices through hydraulically amplified electrostatic actuators (Figure 3B).\(^\text{53}\) Feng et al. fabricated a refreshable braille display using IPMC actuators (Figure 3C).\(^\text{56}\) Liu et al. produced small deformations on liquid crystal elastomer (LCE) surfaces by applying electric fields (Figure 3D).\(^\text{61}\) Besse et al.
utilized thin shape memory polymer (SMP) membranes with integrated and compliant joule heaters to produce pixelated haptic feedback devices (Figure 3E).\textsuperscript{[62]} Ji et al. demonstrated electrically driven vibrotactile haptic feedback device (Figure 3F).\textsuperscript{[63]} We shall review the various principles behind these electrically driven actuation mechanisms in the following sections.
Table 2. Summary of haptic demonstrations utilizing electrically driven soft actuation mechanisms detailing out the utilized material systems for active layer and compliant electrodes and their actuation performance.

| S. No. | Detail | Active layer material | Compliant electrode material | Soft actuator technology | Electric field \( [\text{V} \mu \text{m} / \text{C}_0^1] \) | Actuation performance | Reference |
|--------|--------|------------------------|-----------------------------|-------------------------|--------------------------------|------------------------|-----------|
| 1      | Untethered feel-through haptics | Polydimethylsiloxane (PDMS) | SWCNT | DEAs | 75 | Areal Strain (max.) 14\% @ 10 Hz Out-of-plane displacement (max.) 6 \( \mu \)m @ 10 Hz | Ji X. et al. [294] |
| 2      | Thickness-mode actuation | 3M VHB Acrylic 4910 | AgNWs and hydrogel | 21.4 | Out-of-plane displacement (max.) 155 \( \mu \)m | Ankit et al. [8] |
| 3      | Multiple active layers with rigid coupling | Silicone | Graphite particles | 60 | Out-of-plane displacement (max.) 350 \( \mu \)m | Phung, H. et al. [272] |
| 4      | Vibrotactile actuator platform | PDMS and poly(ethylene terephthalate) (PET) | Silver microwires (AgMWs) and indium tin oxide (ITO) | – | Out-of-plane displacement (max.) 36 \( \mu \)m @ 105 Hz Blocked force (max.) 150 mN | Kang J. et al. [295] |
| 5      | Transparent haptic interface | Blend of PDMS and Ecoflex | AgNWs and graphene | 13 | Out-of-plane displacement (max.) 100 \( \mu \)m | Jun K. et al. [56] |
| 6      | Multiple active layers with hydrostatically coupling fluid | 3M VHB Acrylic 4910 | Silicone/carbon black composite | 80 | Out-of-plane displacement (max.) 150 \( \mu \)m @ 10 Hz Blocked force (max.) 150 mN | Frediani G. et al. [296] |
| 7      | Tactile bump array of IPMC pixels | Nafion with PDMS bump | Platinum electrode | IPMCs | 8 | Maximum blocking force (4.1 mg load) 0.35 mN @ 0.5 Hz Bump height (4.1 mg load) 0.55 mm @ 0.5 Hz | Feng, G.-H. et al. [292] |
| 8      | Braille display with IPMC | Nafion with PDMS bump | Gold electrode | 2.5 | Maximum blocking force | 14.7 mN | Kato. Y. et al. [297] |
| 9      | Haptic bump | Silicone oil with silicone rubber-compliant layer | Conductive silver ink | Electrohydraulic actuators | 10 | Blocked force | 13 mN | Han, A. K. et al. [77] |
| 10     | HAXEL | Envirotemp FR3 dielectric oil with polyvinylidene fluoride (PVDF)-compliant layer | Mylar metalized with aluminum | 70 | Blocked force | 300 mN @ 80 Hz Out-of-plane displacement 0.1 mm @ 200 Hz | Leroy, E. et al. [192] |
| 11     | Large-area haptic display | Transformer oil encapsulated within BOPP pouch | JN0917-A commercial hydrogel | 4.8 | Blocked force | 0.6 N | Shao, Y. et al. [179] |
| 12     | Dielectric LCE actuator | 1,4-bis-[4-(6-acryloyloxyhexyloxy)benzoyloxy]-2-methylbenzene (RM 82) | Conductive carbon grease | LCEs | 31 | Out-of-plane displacement | 0.74 mm 22\% Areal strain 0.13 mm @ 0.1 Hz Frequency-strain response | Davidson, Z. S. et al. [298] |
| 13     | Protruding organic surfaces via in-plane electric fields | 1,4-Bis-[4-(3-acryloyloxypropoxy)benzoyloxy]-2-methylbenzene (RM 237) | ITO | 7.5 | Out-of-plane displacement 140 nm @ 900 kHz, 41\°C | Liu, D. et al. [82] |
| 14     | Oscillating LCE surface | RM 82 | RM2AE | ITO | 16.1 | Out-of-plane displacement | 125 nm @ 900 kHz | Feng, W. et al. [84] |
4.1. Dielectric Elastomer Actuators (DEAs)

The basic configuration of a DEA system consists of an electroactive polymer sandwiched between two compliant electrodes. The area on the polymer where the electrodes overlap is commonly referred to as the active area. When a sufficiently high voltage is applied, the resultant build-up of free charges on the surface of the polymer layer exerts an attractive Coulombic force, inducing compressive stress on the polymer. This causes the polymer layer to become compressed along the thickness and stretched along the plane of the polymer. The induced stresses on the polymer, more commonly known as Maxwell stresses, depend on the applied voltage, thickness of the polymer, and the relative permittivity of the polymer layer. This can be visualized mathematically using the following equation.
\[ p = \varepsilon \varepsilon_0 \left( \frac{V}{d} \right)^2 \]  

where \( p \) is the effective Maxwell stress on the polymer, \( \varepsilon \) is the relative permittivity of the polymer layer, \( \varepsilon_0 \) is the permittivity of free space \((8.85 \times 10^{-12} \text{ F m}^{-1})\), \( V \) is the voltage applied to the electrodes, and \( d \) is the thickness of the polymer layer.\(^{[65]}\)

DEAs offer advantages of facile fabrication and simplistic design for achieving complex device arrangements, typically desired in tactile feedback applications. Furthermore, thin device architecture makes them suitable for integration with different interfaces. However, achieving higher actuation strain with the use of high voltages and very thin polymer layers has limited practicability.\(^{[64,66–68]}\) Material design approaches have focused on increasing the relative permittivity of the polymer layer to achieve higher Maxwell stresses, which leads to greater actuation.\(^{[52,68,69]}\) Furthermore, as the elastic deformation of any material depends on its Young’s modulus, a measure of stiffness of the material, the choice of polymer layer determines the actuation strain as well.\(^{[65]}\) Thus, there is an ongoing effort in the search for soft materials with high relative permittivity for their potential applications as DEAs.
4.2. Electrohydraulic Soft Actuators (ESAs)

Electrohydraulic soft actuators (ESAs) are, in essence, a variation of DEAs. ESAs incorporate the working principle of DEAs with fluid-driven actuators, resulting in a different dynamic behavior driven by electrostatic forces. The encapsulation of a liquid dielectric within a compliant shell forms the basic configuration of these ESAs. Upon application of a voltage, Coulombic forces of attraction between the electrodes on the shell squeeze the liquid in the gap between the electrodes, displacing the liquid. The pressure exerted by the electrodes onto the liquid will be transferred to all points of the liquid with minimal loss. This resulting pressure exerts a force on the stretchable membrane, causing its deformation (Figure 4B). This actuation mechanism leverages on the amplification of hydraulic pressure, as described by Pascal’s law.

\[ F = p \times A \]  

where \( F \) is the force, \( p \) is pressure, and \( A \) is the effective surface area, where the liquid is in contact with the stretchable membrane. As Maxwell’s stress is effectively converted into fluid pressure with minimal losses, force amplification arises from the effective surface area of the stretchable membrane.

Like DEAs, ESAs have specific advantages, which make them suitable for haptic applications. Miniaturization and pixelation have already been demonstrated for ESAs and their force output characteristics make them a good alternative for wearable haptic feedback devices. However, while ESAs use the working principle of DEAs by electrostatic forces for activation, the dynamics of ESAs are not controlled solely by elastic effects. Instead, ESAs benefit from utilizing a liquid dielectric instead of an elastomeric polymer like in DEAs. In DEAs, electrostatic force exerted works against the elastic dielectric material, and some energy is lost as elastic potential energy, resulting in a lower output force of the DEA device. In contrast, the use of a liquid dielectric in ESAs prevents the storage of energy within the dielectric. As a result, electrostatic pressure exerted is converted completely to fluidic pressure, which in turn is converted into the output force with minimal loss of energy. However, the utilization of a liquid dielectric affects the actuation speeds of ESAs due to its viscosity. Furthermore, ESAs suffer from operation limits similar to DEAs such as high operation voltage, as the fluidic pressure is dependent on Maxwell’s stress, which is in turn dependent on voltage (Equation 1). In addition, careful design consideration and maintenance is required for ESA systems, as leakage of the liquid dielectric may lead to device failure.

4.3. Ionic Polymer–Metal Composites (IPMCs)

An ionic polymer–metal composite (IPMC) system consists of an ionic–polymer membrane sandwiched between two metal electrodes, in a cantilever-like configuration. The ionic polymer membrane, also known as a polyelectrolyte, is made up of polycations or polyanions, with soluble counterions. Before hydration of the membrane, the polymer network exists as a neutral species. Upon hydration, the dissolution of the polymer network occurs, causing the counterions to be free and mobile within the polymer membrane. Upon application of voltage across the electrodes of the IPMC system, redistribution of charges occurs due to the movement of counterions across the membrane. This leads to a regional change in volume of the polymer membrane which causes deformation of the material (Figure 4C). From a molecular point of view, factors such as ionic exchange capacity (IEC), water uptake (WU), and ionic conductivity (IC) directly affect the actuation performance of IPMC systems.

Higher IEC and IC allow for faster ionic movement within the ionic layer, leading to greater actuation speeds and forces. Higher WU improves the stability of the IPMCs, allowing for longer operation duration and enhanced actuation performances. However, commercially available Nafion and Flemion lack these qualities. As such, there are ongoing efforts to improve these qualities in commercially available membranes and synthesis of new ionic layers with these enhanced properties.

4.4. LCEs

Polymeric liquid crystals such as lightly crosslinked LCEs or densely crosslinked liquid crystal networks (LCNs) are polymers that display both the properties of solids and liquids. They are made up of liquid crystal molecules, also known as mesogens, which are either part of the polymer backbone or attached to the polymer network as side chains. These mesogens are typically rigid and largely planar. Anisotropic intermolecular interaction of mesogens allows for self-organization to crystalline phases. These interactions allow for orientational or directional order in these polymeric LC systems, similar to that of crystalline solids. However, these mesogens in LC systems lack positional order, and they can slide over each other. The unique dual characteristics of mesogens allow for a reversible phase change upon the application of external stimuli such as heat, light, or electricity. This reversible phase change drives the actuation mechanism of polymeric LCs, allowing for actuation without the need for reprogramming unlike traditional shape memory polymers.

LCNs are conventionally prepared via the polymerization of multifunctional mesogenic monomers to yield densely crosslinked glassy networks. Despite the limited strain (due to their high crosslink density) of LCNs, their potential utility for electrically stimulated actuation has been explored. LCs can efficiently couple to an external electric field due to the presence of polar functional groups. Under a high-frequency AC field, oscillatory motion of LCs ensues, subsequently causing molecular voids or geometric deformations, which increase free volume. LCEs are different from LCNs primarily based on their crosslinking density. They are prepared via hydrosilylation of siloxane monomers and vinyl or acrylate mesogens. The first step requires the synthesis of a lightly crosslinked polymer network, with mesogens existing in a polydomain state. Application of a uniaxial force realigns the mesogens into a monodomain state. The second and final step consists of a crosslinking reaction, yielding the desired LCE with mesogens locked in the monodomain state.

There are two main mechanisms via which LCEs respond to electrical stimuli: 1) direct response to applied electric field, electrically responsive LCEs, and 2) response induced by Joule...
heating via electrical current, electrothermally actuated LCEs. In electrically responsive LCEs, phase transitions between Smectic A and Smectic C* phases are observed upon application of electric field. Smectic A phase refers to the orientational and positional state of LCEs where mesogens are aligned parallel to the layer normal. Upon application of electrical stimuli, the mesogens undergo a molecular tilt to give the Smectic C* phase (Figure 4D). This molecular tilting between Smectic A and Smectic C* is also termed "electroclinic effect." This phase transition between Smectic A and Smectic C* typically requires high electric field strengths and result in small actuation strains.

In electrothermally actuated LCEs, phase transitions between nematic and isotropic phases are most common. Mesogens in the nematic phase possess long-range orientational order but short-range positional order. In the phase transition to the isotropic phase, orientational order is lost in these systems, resulting in actuation of the polymer. As LCEs are insulating in nature, electrodes must be patterned on these polymers to access electrothermal actuation of such systems via Joule heating. This indirect heating strategy allows for the localized and efficient temperature control of the polymer system, where the actuation can be precisely controlled in larger LCE systems.

4.5. Other Actuation Mechanisms

Other classes of materials have been widely studied for their promising applications as actuators. Shape memory alloys (SMAs) that actuate on the principle of a martensitic phase transition is one such example. Under thermal stimulus, phase change from monoclinic martensite phase to cubic austenite occurs and any deformation of the SMA in the martensite phase will be reversed. SMPs demonstrate shape changing properties similar to SMAs. When deformed under heat, SMPs can retain their deformation after the removal of thermal stimulus. The recovery of the original shape can be triggered upon reapplication of heat (Figure 4E). While the actuation of SMAs can be activated directly via resistive heating, SMPs are typically nonconductive, and further processing techniques must be utilized for electrothermal actuation to occur. Thermal bimorph actuators consist of two materials possessing different CTE. These actuators possess various possible configurations, with the most common being a bilayer configuration (Figure 4F). When the actuator is heated, the material with a higher CTE expands to a larger extent than the material with a lower CTE. This difference in expansion causes a deformation of the actuator toward the side with lower CTE. When the actuator is cooled to lower temperatures, deformation in the reverse direction can be observed. Beyond actuator materials for haptics, electrostatics have also been widely used. Such approaches involve supplying an AC voltage to the device, inducing an electrostatic force between the insulating surface and a contact point from the user. The magnitude of electrostatic force can be regulated by the AC voltage, which results in a change in the frictional force between the surface and the contact point from the user. Consequently, different sensations can be felt by the user.

5. Material Developments for the Active Layer

Having looked at the underlying mechanism for the electrically driven soft actuators, this section details out the material systems utilized in these soft actuators. The focus will be on mechanisms that provide direct response to the applied electric field: DEAs, ESAs, IPMCs, and LCEs. In the following section, we will look at the material approaches adopted for improving the electromechanical response of these soft actuators.

5.1. Material Systems for DEA Active Layer

Actuation performance of a DEA is primarily characterized by measuring the actuation strain produced under static and dynamic electrical inputs. Actuation strain produced in a DEA is given by

\[
S = \frac{1}{Y} \varepsilon \left( \frac{V}{d} \right)^2
\]

where all symbols have same meaning as Section 4.1 and \( Y \) stands for Young’s modulus. It is evident from the equation that actuation performance of a DEA (actuation strain produced) depends directly on relative permittivity of the material and depends inversely on the Young’s modulus of the material. Maximum applied voltage is directly dependent upon the electrical breakdown strength of the material. Moving forward, we take a look at the commercially available material systems for DEAs and different material approaches adopted by researchers to improve the actuation performance of DEAs.

5.1.1. Conventional/Classical Material Systems for Active Layer

Essentially, DEAs comprise two important components, 1) active layer, which responds to the applied electrical stimuli, and 2) compliant electrodes to apply the electrical stimuli. Conventionally, acrylics and silicones have been extensively used as active layers in DEAs due to the range of electrical and mechanical properties accessible within them. Pelrine et al., in their seminal paper, demonstrated the use of very high bond (VHB) strength acrylic films in a biaxially prestretched configuration to achieve high actuation strains. Proprietary VHB series from 3M corporation are commonly used acrylic films as active layers for DEA devices. These acrylic films are mixtures of aliphatic acrylates, based on the structure of a vinyl group and a carboxylic acid terminus. VHB series acrylic films possess low mechanical stiffness (≈50 kPa), high theoretical energy density (3.4 MJ m\(^{-3}\)), and a reasonable dielectric constant (4.5–4.8 @ 1 kHz). Approaches like interpenetrating networks (Figure 5A), minimum energy structures, combining stiff fibers, and laterally constrained structures in combination with these acrylic films have been utilized with VHB acrylics to make the DEAs versatile and improve on their actuation performance.

However, to achieve the high actuation strains and their expected level of electromechanical performance, it becomes absolutely necessary to prestretch these acrylic membranes. Prestretching leads to the necessity for rigid frames to support the membranes under constant tension and limits their applicability. Furthermore, they suffer from
significant viscoelastic losses affecting their actuation performance and lead to long-term relaxation. Long-term relaxation in turn leads to considerable losses in actuation performance over a period of time. In addition, they are very sensitive to ambient and their performance gets affected by moisture and temperature. Due to its proprietary nature, venues for major modifications to the underlying chemical structure of the material and tailoring of desired properties are limited.

Silicone polymers, or polysiloxanes, are blended inorganic–organic polymers based on silicon and oxygen backbone (Si–O).

The most commonly utilized silicone polymer is polydimethylsiloxane (PDMS) which has the methyl organic group. Silicons have an array of interesting material properties: high thermal stability, lower viscoelasticity, chemical and biological stability and inertness, transparency, and hydrophobicity. Silicone also provide the advantage of easy processability, and it can be tailored to suit the application (such as DEAs). However, polysiloxanes lag behind in terms of actuation performance due to their low dielectric constant (2.5–3.0 @ 1 kHz) and low energy density (0.75 MJ m⁻³). Other material
matrices such as styrene ethylene butylene styrene (SEBS) block copolymer, polyurethanes, and styrene rubber have also been utilized for DEAs but have provided limited success in terms of improving the mechanical performance.\textsuperscript{[107,117,118]}

5.1.2. Material Approaches to Improve the Electromechanical Properties of Active Layer

As seen from the governing equations for DEAs, the actuation performance depends on the characteristic material properties such as dielectric constant (also referred to as relative permittivity), mechanical stiffness (Young’s modulus or elastic modulus), and electrical breakdown strength.\textsuperscript{[119]} Hence, the efforts to improve the actuation performance of DEAs have focused on tailoring the abovementioned material properties of the matrices.

**Reducing the Mechanical Stiffness:** Addition of plasticizers is one of the traditional ways of lowering the Young’s modulus of pristine polymer networks and is effective for elastomer networks as well. Lowe et al. investigated the role of plasticizers (81-R, 81-F, and 81-VF) in tuning the mechanical stiffness of commercially available silicone elastomer (DC3481, Dow Corning).\textsuperscript{[120]} Plasticizers such as dioctyl phthalate and epoxidized soybean oil have also been used for acrylonitrile–butadiene rubbers (NBR).\textsuperscript{[121,122]} This approach leads to considerable improvement in the actuation performance of DEAs. However, the volatile and migrating nature of the plasticizers is the critical concern for this approach.\textsuperscript{[107,123]} This results in long-term stability and reliability issues for these actuators. It is also important to note that some of these plasticizers like dioctyl phthalate are extremely toxic in nature.\textsuperscript{[107]}

Direct chemical modification of the polymer backbone has also been pursued for altering the mechanical properties. One of the simpler strategies is to vary the stoichiometric ratio for commercially available polysiloxanes, which comes as a two-component system.\textsuperscript{[116]} Niu et al. and Duduta et al. have demonstrated tunable stress–strain behavior of DE films in a mixture of UV-curable urethane–acylate oligomers together with co-monomers and varying concentration of crosslinkers.\textsuperscript{[124,125]} However, changing the stoichiometric ratio from the recommended ratio leads to imbalanced reagents, which in turn leads to incomplete polymerization.\textsuperscript{[116]} The resulting disconnected polymer constituents and sol structures (fraction of polymer not covalently connected to the polymer active network) can lead to stability issues.\textsuperscript{[116]} Opris et al. investigated the role of the type of crosslinker (ethyliacteoylsilane, methylhydrosiloxane-dimethylsiloxane copolymer, and tetraethoxysilane) in tuning the mechanical properties of the silicone networks.\textsuperscript{[126]} Bejenaru et al. utilized a controlled addition reaction to lower the elastic modulus without affecting the viscoelasticity.\textsuperscript{[127]} Vatanakham–Varoosofaderani et al. demonstrated the concept of bottlebrush elastomers for DEAs.\textsuperscript{[128]} They managed to tailor the elastic modulus by introducing multiple covalently linked side chains along the polysiloxanes network strands (Figure 5B). However, the laborious and complicated chemical modification steps could often lead to detrimental effects on the viscoelastic properties and thus require careful execution.

**Chemical Alterations to the Polymeric Backbone:** A suitable strategy for increasing the dielectric constant of the polymer matrix is to alter the chemical structure of the existing polymer backbone (Figure 5C) or synthesize a novel polymer matrix.\textsuperscript{[129]} Stoyanov et al. reported a 470% increase in dielectric constant values by grafting π-conjugated conducting macromolecule, polyaniline (PANI), doped with dodecyl benzene sulfonic acid (DBSA) to the chemical backbone of polystyrene-co-ethylene-co-butylene-co-styrene-grafted maleic anhydride (SEBS-g-MA, thermoplastic copolymer).\textsuperscript{[130]} where a shift from dielectric to conductive behavior occurs at very low additive concentration (2%). Along similar lines, Kusmaul et al. showed an impressive increase of 200% in dielectric constant by addition of N-allyl-N-methyl-p-nitroaniline (push–pull dipole) to the PDMS backbone.\textsuperscript{[131,132]} This method of attaching dipoles to the polymer backbone was accompanied by a decrease in mechanical stiffness (Figure 5D).\textsuperscript{[133]} However, the large dipole content in the polymer backbone leads to moisture sensitivity. Racles et al. synthesized silicone networks with highly polarizable cyanopropyl groups via different synthesis routes.\textsuperscript{[134]} The resulting networks exhibited a threefold increase in dielectric constant. However, the brittle nature of the synthesized films rendered them unsuitable for DEA applications. Building upon this approach, Madsen et al. conducted the functionalization of siloxane copolymer by a pendant polar group (1-ethyl-4-nitrobenzene) using a catalyst-assisted cycloaddition reaction.\textsuperscript{[135]} This resulted in an increase of 70% in dielectric constant. Dünki et al. functionalized a poly(methylvinylsiloxane) chain with highly polarizable nitrile moieties, allowing for tuning of dielectric constant and mechanical stiffness.\textsuperscript{[136]}

**Elastomer Composites and Blends:** The approach of fabricating elastomeric composites by incorporating conductive and ceramic fillers offers much advantage in terms of processing simplicity and hence, is widely explored in literature for improving the dielectric constant. For conducting particles, addition to the polymer matrix below the percolation threshold keeps the particles far away from each other so that the electrical properties of the composite are directed by the medium itself.\textsuperscript{[137]} Behavior of composites fabricated from microparticles and nanoparticles has been observed to be different. The nature and concentration of nanofillers along with the interfacial interaction between the nanoparticle and polymer play a significant role in the resulting behavior of the composite.\textsuperscript{[138]} Various conductive fillers have been investigated for increasing the dielectric constant of elastomeric matrices: nickel nanoparticles in neoprene, silica-supported copper nanoparticles in poly styrene (co-ethylene-co-butylene-co-styrene) (SEBS); iron nanoparticles coated with silicon dioxide in silicone; and carbon black microparticles in SEBS and ethylene acrylic.\textsuperscript{[139,142]} Graphitic nanomaterials are also an attractive option for increasing the dielectric constant of polymers. Park et al. added multiaxial carbon nanotubes (MWCNTs) into PDMS matrix to achieve nearly two times increment in the dielectric constant.\textsuperscript{[143]} Liu et al. managed to align the MWCNTs dispersed in polysulphone (PSF) nanofibers by electrospinning to achieve very high relative permittivity values.\textsuperscript{[144]} Improved dispersion of the MWCNT within the PDMS matrix has been performed using an anionic surfactant, dodecylbenzene sulfonic acid DBSA.\textsuperscript{[145]} Romasanta et al. reported addition of functionalized graphene sheets (FGS) in PDMS, resulting in an increase of ten times in permittivity value.\textsuperscript{[146]} Similar results on increase in the dielectric constant were also reported via addition of reduced graphene oxide (RGO)/PDMS and expanded
graphite (EG)/polysobutylene-co-isoprene (IIR) composite. Tian et al. utilized hybrid nanoparticles (self-assembling graphene oxide on carbon nanospheres) in carboxylated nitrile rubber (XNBR) composite to achieve a significant increase in dielectric constant. To improve upon the interfacial interaction of nanofiller and polymer, an approach to microencapsulate the conductive particles with an insulating polymer layer has also been adopted. This in turn helps in increasing the concentration of conductive fillers without the risk of formation of percolative networks and particle agglomeration.

Apart from conductive particles, a large variety of high-permittivity inorganic, ceramic fillers has been added to the polymer matrices as well. Commonly used inorganic fillers for increasing the dielectric constant of polymer composites are titanium dioxide (TiO$_2$), boron nitride (BN), and barium titanate (BaTiO$_3$). Carpi et al. added rutile TiO$_2$ in the silicone matrix to achieve an increase in the composite’s dielectric constant, leading to improvement in actuation performance. Romasanta et al. fabricated silicone composites by addition of calcium copper titanate (CaCu$_3$Ti$_4$O$_{12}$), resulting in increase in dielectric constant.

Another strategy to improve the dielectric constant is to create elastomer blends, which involves mixing of two polymers (miscible or immiscible) (Figure 5E). Zhang et al. dispersed an organic polymer (copper–phthalocyanine [CuPc]) in a ferroelectric polymer matrix (poly(vinylidene fluoride-trifluoroethylene) (PVDF-TrFE)), electrostrictive elastomer to demonstrate significant improvement in the dielectric constant. Carpi et al. showed a large increase in dielectric constant by creating a highly polarizable conjugated polymer network by blending poly(3-hexylthiophene) (P3HT) to the PDMS network. Gallone et al. mixed PDMS and PU to create polymer blends with a high dielectric constant value by taking advantage of strong interfacial polarization (Maxwell–Wagner–Sillars relaxation).

Tian et al. incorporated polyethylene glycol in thermoplastic polyurethane (TPU) to achieve a coactive effect on electrical and mechanical properties of TPU, exploiting the strategy of improving interfacial polarization.

However, the approach of utilizing solid fillers could lead to degradation of mechanical softness and stretchability. It almost always becomes necessary to use additives and surfactants for improving the interfacial interaction between the fillers and the matrix to prevent issues like agglomeration and phase separation.

**Liquid-Filler Approach for Novel Polymer Composites:** Recently, an innovative approach of utilizing liquid fillers instead of solid fillers has been adopted by researchers to improve upon the electrical and mechanical properties of polymer matrices. Bartlett et al. mixed eutectic gallium indium (EGaIn), which is a low-melting-point liquid metal (LM) alloy, into silicone matrix. This resulted in microencapsulated EGaIn particles within the matrix and significant increase in the dielectric constant. However, the microparticles tend to settle down and phase separate during the crosslinking of silicone matrix. Stable EGaIn nanoparticles in different solvents have also been successfully fabricated. Utilizing EGaIn nanoparticles, Pan et al. formed silicone elastomer nanocomposites, demonstrating high dielectric constant values compared with the pristine elastomer matrix. Shi et al. utilized the approach of liquid incorporation to fabricate dielectric gels with low elastic moduli and high dielectric constant. Mazurek et al. utilized glycerol as liquid filler to demonstrate tuning of electrical and mechanical properties of silicone elastomers. Our group recently demonstrated utilization of an ionic liquid,
1-ethyl-3-methylimidazolium bis(trifluoromethylsulfonyl)imide, EMIMTFSI, as a self-enclosed liquid filler within the silicone matrix to produce synergistic improvements in electrical and mechanical properties (Figure 6B). This approach has great promise as it aims to mitigate the mismatch of interfacial interaction between solid filler inclusions and polymer matrices. Concurrently, Shi et al. also demonstrated improvements in the dielectric constant of polymer matrix by incorporation of ionic conductive liquid electrolyte inclusions.

5.2. Material Systems for ESAs

Typical approaches for elastomeric electrohydrodynamic systems involve a predefined area on the stretchable membrane that stretches upon displacement of the contained fluid. Dielectric constants and viscosity of the fluid are important considerations that affect the actuation performances of the electrohydrodynamic actuator. In most electrohydrodynamic systems, oil is utilized as the liquid dielectric. In “push–pull” electrohydrodynamic actuator systems, the liquid dielectric is enclosed between a passive and an active membrane (coated with compliant electrodes) forming a spherical shell. These membranes are typically prestretched to push and pull the dielectric in a pouch, where a portion of the pouch is then sandwiched between the electrodes. When voltage is applied to the electrodes, the resulting “zipping” motion causes the displacement of liquid dielectric, which exerts a force on the soft membrane. Recently, Acome et al. demonstrated such an actuator system, termed HASEL actuators (Figure 6C). These actuator systems have advantages over conventional DEA systems, as the use of liquid dielectrics allows for self-healing of the active layer, where functionality of the actuator systems is immediately recovered despite multiple dielectric breakdown events. Combining the advantages of Peano-fluidic actuators and HASEL actuators, Kellaris et al. presented a Peano-HASEL actuator. In this system, a noncompliant shell was used to enclose the liquid dielectric. Fast actuation speeds on the millisecond scale were reported, and the scalability of its configuration was demonstrated. However, actuation performances were diminished by an increase in the viscosity of the liquid dielectric at the same frequency. As such, liquid dielectrics possessing lower viscosity will be better suited for applications requiring high frequencies of actuation. Recently, electrohydrodynamic systems which rely on the acceleration of a liquid dielectric in the presence of a DC electric field are under exploration. When the electric field exceeds a certain threshold, the movement of mobile ions to the electrode of opposite polarity is induced. Consequently, neutral liquid molecules clustered to the ions are dragged along, causing fluidic flow. Cacucciolo et al. recently reported such an electrohydrodynamic system, demonstrated as a bidirectional soft pump. The encapsulation of a dielectric liquid, Fluorinert FC-40 (3M), within a compatible PDMS membrane allowed for reversible actuation strains of over 50%. Furthermore, the soft pump showed comparable performances to commercially available and previously reported pumps with significantly lower power consumption values.

5.3. Material Approaches for Ion Exchange Membranes in IPMCs

Conventional IPMCs use commercially available Nafion or Flemion thin films. Nafion and Flemion are perfluorinated polymers which are protic in nature, possessing sulfonate or carboxylate functional groups in their polymer chains, respectively. When hydrated, these functional groups are deprotonated and act as ionic channels that allow the passage of free hydrated protons. Under an applied electric field, the hydrated protons flow toward the cathode, causing a change in the osmotic pressure in the membrane. This results in the bending of the membrane toward the anode. Reversing the applied electric field inverts the flow of the hydrated protons, causing deformation of the membrane to occur in the opposite direction. The fast actuation speed, high flexibility, and low activation voltage (1–5 V) of Nafion or Flemion-based IPMCs have made them interesting materials for soft robotics. However, they suffer from hydration loss, poor actuation force, and relaxation under DC voltage.

5.3.1. Improvements to Commercially Available-Based Polymer Membranes

Several strategies have been pursued to improve the properties of conventional IPMC systems. Encapsulation of IPMC systems in other soft polymer materials such as Saran or polydimethylsiloxane have been used as direct strategies to effectively reduce water losses. However, the use of such materials results in an overall increase in stiffness of the system, which leads to poorer actuation performances. Another strategy focused on replacing water with nonvolatile solvents, such as ionic liquids. Ionic liquids possess negligible vapor pressure and larger electrochemical stability windows than aqueous electrolytes. Ionic liquid-based IPMCs have shown greater stability in air over conventional IPMCs and alleviate relaxation issues. However, they suffer from slower response time due to the higher viscosity of ionic liquids, which impedes the movement of mobile cations during operation.

In contrast, doping of polymer membranes shows more promising improvements in conventional IPMC systems. Landi et al. introduced single-walled carbon nanotubes (SWCNTs) to a Nafion membrane, improving its electrical conductivity and providing highly efficient conducting channels for proton migration, resulting in higher actuation performances. Considerably lower doping levels were required to achieve the same actuation threshold in SWCNT-doped Nafion compared with metal-doped Nafion. Along a similar line, Ru et al. utilized carboxylated MWCNTs as dopants in Nafion, eliminating back-relaxation at a low 2%wt loading (Figure 7A). The secondary carboxylate groups from the CNTs generate a secondary source of H⁺ ions, which migrate toward the cathode, counteracting the back-migration of water to the anode. However, IPMCs doped with CNTs have shown compromised performances due to the aggregation of CNTs. In an alternative approach, Bian et al. incorporated barium titanate (BaTiO₃) nanoparticles within the
Nafion membrane. BaTiO$_3$ is a ferroelectric with high dielectric constant, which enhances charge accumulation at the electrodes of the IPMC (Figure 7B), resulting in exceptional deformation strains (250%).$^{[176]}$ In addition, BaTiO$_3$ imbues the composite with greater tensile strength, with an increased blocking force (375%) as compared with pure Nafion.$^{[176]}$ Other dopants, such as 2D materials, possess extremely attractive electronic properties and their incorporation in IPMCs has only been recently studied.$^{[63,186,187]}$ Loelian et al. investigated the doping of Nafion with tungsten disulfide (WS$_2$). At a significantly low loading of 0.5 wt%, the proton-conducting channels of Nafion were extended, and improved proton conductivity by 160% was reported.$^{[63]}$ This is evidenced by improved actuation performance of almost two times (74.1%). Hydrophilicity of WS$_2$ also increased the WU of the composite (300%), resulting in softer actuators.$^{[63]}$

5.3.2. Novel Ionic Polymer Membranes

While doping strategies are highly attractive, inherent limitations of commercially available ionic membranes such as high cost and environmental unfriendliness are unavoidable.$^{[188]}$ Furthermore, commercially available membranes have limited thickness and are not melt-processable.$^{[183]}$ Thus, the synthesis of alternative ionic membranes has been pursued.$^{[189,190]}$ Jeong et al. first attempted the fabrication of such an IPMC actuator, utilizing a novel fluorinated acrylic copolymer membrane bearing a similar structure to Nafion.$^{[191]}$ The membrane was easily synthesized and processable, granting convenience in the fabrication of IPMC devices.$^{[191]}$ While no back-relaxation was observed in this IPMC, higher actuation voltages (3–8 V) were required.$^{[191]}$ Furthermore, environmental unfriendliness was not alleviated due to the use of fluorinated monomers. Since then, the synthesis of nonfluorinated polymers has been attempted, and subsequent sulfonation or carboxylic acidification (Figure 7C) was conducted to transform the polymers into polyelectrolytes for use in IPMCs.$^{[192]}$ For instance, Tang et al. developed a sulfonated polyphenylsulfone (SPPS) ionic membrane and studied the degree of sulfonation in the membrane.$^{[192]}$ The number of sulfonate pendant functional groups was directly correlated with WU and ion conductivity, which had a direct consequence on actuation performance. However, introduction of a rigid aromatic backbone in SPPS led to greater stiffness of the polymer membrane and slightly lower proton conductivity as compared with Nafion.$^{[192]}$ Nonetheless, the SPPS membrane showed a more rapid bending response with a maximum strain of approximately two times that of Nafion.$^{[192]}$ Recently, Khan et al. developed a sulfonated poly(1,4-phenylene ether-ether-sulfone) (SPEES) polymer membrane with functionalized SWCNT.$^{[193]}$ The aromatic linkages in PEES provide rigidity and strength to the polymer membrane, whereas ether linkages improve the processability of PEES.$^{[193]}$ The synergy of functionalized SWCNT and high degree of

![Figure 7. Materials for active layer in IPMCs. A) Functionalization of CNTs with carboxylate groups. Reproduced under the terms of Creative Commons Attribution 4.0 International License (CC BY 4.0).$^{[193]}$ Copyright 2018, the authors. Published by Springer Nature. B) Illustrative actuation of BaTiO$_3$ nanoparticles-doped Nafion membrane. Reproduced with permission.$^{[176]}$ Copyright 2016, American Chemical Society. C) Here, the representative example of sulfonation as a transformation step to convert polymers into polyelectrolytes for IPMCs is shown. Reproduced under the terms of Creative Commons Attribution 4.0 International License (CC BY 4.0).$^{[193]}$ Copyright 2018, the authors. Published by Springer Nature. Poly(1,4-phenylene ether-ether-sulfone) is used as an example here. D) IPMC consisting of block copolymer of polystyrene and poly(1-ethyl-3-methylimidazolium-4-styrenesulfonate) in an ionic liquid electrolyte as the active layer. Reproduced under the terms of Creative Commons Attribution 4.0 International License (CC BY 4.0).$^{[301]}$ Copyright 2018, the authors. Published by John Wiley and Sons.](image-url)
sulfonation in SPEES resulted in enhanced IEC and proton conductivity, improving ion transport, which leads to greater actuation performances and speeds.\(^{[93]}\) Nguyen and coworkers presented a block copolymer of polystyrene and poly(1-ethyl-3-methylimidazolium-4-styrenesulfonate) in an ionic liquid electrolyte (Figure 7D), which was shown to possess fast and large actuation displacements at ultralow voltage with high cyclic performance (14000 cycles).\(^{[95]}\) Incorporation of the ionic liquid, 1-ethyl-3-methylimidazolium tetrafluoroborate (EMImBF\(_4\)), selectively plasticized the polystyrene–EMIm block and improved ion transport of EMIm\(^+\).\(^{[95]}\) Furthermore, the larger difference in the size of cations and anions resulted in a larger volume change, leading to greater bending of the IPMC. Lu et al. and Li et al. have also demonstrated the use of biocompatible polymers such as chitosan as the active layer in IPMCs. These chitosan/CNT composites with ionic liquid electrolyte showed superfast response speeds accompanied with high force output.\(^{[194,195]}\)

5.4. Material System in LCEs

Current device architectures utilizing LCNs to achieve topographical deformations on a smooth surface, which can be utilized for tactile feedback, mainly focus on the patterning of electrodes. However, LCNs have high crosslinking density, which gives them very high mechanical stiffness. In contrast, LCEs are much softer and have demonstrated higher actuation strains. In this section, we will be reviewing developments in LCE material systems, categorized under electrothermally actuated LCEs and electrically responsive LCEs, which have the potential to offer better actuation performances.

5.4.1. Electrically Responsive LCEs

Liquid crystal displays utilize the orientation of free mesogens (molecular tilting) in the presence of electric fields to modulate the transmission of light. As these mesogens are free-flowing and lack positional order, they are unable to exert any mechanical stress.\(^{[93]}\) The introduction of a soft elastomeric network stabilizes the positional order of mesogens, overcoming this limitation.\(^{[93]}\) In the presence of an electric field, the electroclinic effect causes a phase change from smectic A to smectic C. (Figure 4D) This induces an in-plane shear of the polymer, which is translated via the soft elastomeric network, resulting in a constriction of the smectic layers. Hence, a macroscopic constriction of the material is observed.

One key attribute mesogens must possess to access smectic C phase is chirality.\(^{[93,196]}\) Chiral mesogens lack the mirror symmetry operation about the tilt plane and therefore can develop spontaneous electrical polarization perpendicular to the tilt plane.\(^{[196]}\) The tilting of mesogens for phase transition from smectic A to smectic C can hence be controlled in the presence of an electric field, resulting in electrostriction. Lehmann et al. were the first to report exceptional electrostriction of LCEs, where 4\% strain was achieved. The electric field strength required for electrostriction in this LCE was 100 times lower than poly(vinylidene fluoride-trifluoroethylene).\(^{[93]}\) The electroclinic effect was observed to be most pronounced at a critical temperature, where the spontaneous breaking of symmetry occurs.\(^{[93]}\)

Since then, efforts have focused on achieving higher electrostriction strains in such materials.\(^{[95]}\) Guin et al. investigated the effects of incorporating SWCNTs in LCE systems. The addition of SWCNT was found to complement the molecular tilting of mesogens in the applied electric field (5 MV m\(^{-1}\)), where rapid electrostriction (<1 s) of 18\% at 100 °C was achieved.\(^{[197]}\) SWCNTs were believed to play a key role in interfacial charge accumulation with the dipoles of the mesogens, which resulted in a synergistic strong electrostriction.\(^{[197,198]}\) Spillman et al. presented a chiral smectic A elastomer shown to exhibit unusual twisting strains (Figure 8A) under the presence of a low electric field (16 V).\(^{[92]}\) This twisting actuation deviated from the expected parallelogram distortion, arising from a synergy of the electroclinic effect and asymmetry between the surfaces of the elastomer. Furthermore, well-studied cholesteric liquid crystals, known for their fast reorientation properties, were used to fabricate LCEs as well. Fuchigami et al. demonstrated such a system, where strains of up to 32\% with rapid reversible actuation were observed.\(^{[199]}\) However, advances in the field of smectic LCE actuators are limited, as the application of the electroclinic effect in optically switching materials holds greater potential.\(^{[79]}\)

Beyond the scope of smectic LCEs, molecular reorientation was also observed in nematic LCEs\(^{[200–202]}\). For example, Hashimoto et al. presented main-chain LCEs swollen with 4-n-pentyl-4-cyanobiphenyl (SCB) in a polydomain state, with multiple chain director orientations arising from each domain.\(^{[203]}\) Here, the first occurrence of electric field-induced transition from polydomain to monodomain phase was observed, caused by concurrent rotation of chain directors parallel to the electric field direction. An accompanying contraction of 19\% of the LCE occurred within 10 min.\(^{[203]}\) Okamoto et al. also demonstrated this polydomain–monodomain transition in a different LCE system.\(^{[204]}\) The LCE was first polymerized in the isotropic state, before being cooled down to form the nematic state.\(^{[204]}\) The transition was evidenced by the large increase in transmittance values of the LCE (Figure 8B). Contraction of the LCE was observed in the direction of the electric field, concomitant with uniform expansion in the normal plane to the electric field of 35\%.\(^{[204]}\) These polydomain nematic LCEs offer the advantage of convenient synthesis as no additional alignment techniques are required in their fabrication.

5.4.2. Electrothermally Actuated LCEs

Electrothermally actuated LCEs depend on resistive heating of conductive pathways incorporated in the LCE. These electrothermally actuated LCEs are advantageous as they provide higher heating concentrations and controlled energy input.\(^{[205]}\) As LCEs are insulating in nature, conductive dopants or patterned electrodes are typical strategies used to access electrothermal actuation.\(^{[87]}\) For example, Agrawal et al. fabricated a biocompatible side-chain LCE with siloxane backbone, doped with carbon black via a hydrosilylation process.\(^{[206]}\) The flexible Si–O–Si linkages within the siloxane based LCEs imparted desirable properties (high failure strain, low Young’s modulus, and glass transition temperature) for soft actuators.\(^{[207]}\) While the addition of carbon black nanoparticles imparted electrical conductivity to the LCE, it decreased the maximum strain exhibited by the
polymer. Nonetheless, sample contractions of over 5000 cycles were observed upon application of electrical current with rapid response (0.6 s). Main-chain LCE/CNT composites were also explored. Interestingly, Kim et al. investigated the aligning properties of CNT sheets, utilizing it as a tool to access a twisted nematic state of the mesogens, which led to different actuation behaviors. The toughness and Young’s modulus of these LCE/CNT composites were higher than that of neat LCEs and could be heated up to 150 °C in 5 s via resistive heating. The LCE/CNT composites were twisted into fibers and demonstrated a work capacity 2.5 times that of mammalian muscles, showing the potential of these LCEs as actuators. In a separate study, Liu et al. demonstrated the use of extrusion printing coupled with cellulose nanocrystals (CNCs) to align mesogens in LCE/CNT composites. The shear force during extrusion induces mesogen order along the printing path, which was further facilitated by the CNCs due to its ability to form LC phases as well. Enhanced mechanical robustness and actuation speed were further achieved with the loading of rigid and thermally conductive CNTs, at a maximum work capacity of 38 J kg⁻¹. In addition to carbon-based additives, LM–LCE composites have also been explored, taking advantage of the compliancy of LM to achieve a conductive, mechanically unconstrained LCE. Ford et al. demonstrated that even with a significantly high LM wt loading of 50% in an LCE, its actuation capabilities were still retained (Figure 8C). These composites possessed damage detection capabilities and were able to actuate despite sustaining significant punctures. However, the higher loading of LM within the LCE decreases the work content of the active LCE. In addition, high power consumption is required for the heating of these LM–LCE composites. Besides doping of LCEs with conductive materials, most strategies focus on processing and fabrication techniques to attach soft and flexible conductive pathways onto LCE layers (Figure 8D). These strategies utilize commonly synthesized LCE layers, to achieve fast actuation speeds with low voltage requirements. RM 257 is a commonly used mesogen for synthesis of such layers (Figure 8E).

Table 3 provides a summary of different material approaches adopted for the aforementioned electrically driven soft actuation mechanisms. A few examples for these approaches have been presented with their characteristics material properties and potential haptic device application.

### 6. Material Development for Compliant Electrodes

Stretchable electrodes are an integral part of electrically driven haptics, where, in addition to electrical conductivity, high
Table 3. Summary of different material approaches adopted to improve actuation performance with material properties and potential applications mentioned for notable examples (RP: relative permittivity; EM: elastic modulus; EBS: electrical breakdown strength; IEC: ionic exchange capacity; \( \lambda \): ionic conductivity; SU: solvent uptake).

| S. No. | Soft actuation mechanism (material approach) | Remarks | Active material matrix | Material properties | Actuation performance | Potential haptic application | Reference |
|--------|---------------------------------------------|---------|------------------------|---------------------|----------------------|----------------------------|-----------|
| 1      | DEAs (commercially available material systems) | 300 % Biaxial prestretch | Acrylic (3 M VHB 4910) | RP 4.8 \( @ 1 \text{ kHz} \), EM 3 MPa | Area strain: 158% | Cutaneous haptics | Pelrine R. et al. [64] |
|        |                                             | 10% uniaxial prestretch | Dow Corning (Sylgard 184) | RP 2.8 \( @ 1 \text{ kHz} \), EM 2.49 MPa | Thickness strain: 0.5% | Kinesthetic haptics | Risse S. et al. [112] |
|        |                                             | 300 % biaxial prestretch | SEBS 217 (GLS Corp) | RP 1.8--2.2 \( @ 1 \text{ kHz} \), EM 0.002--0.133 MPa | Area strain: 245--47% | Cutaneous haptics | Shankar R. et al. [590] |
| 2      | DEAs (reducing mechanical stiffness)        | PDMS Bottlebrush elastomers | Monomethacryloxypropyl-terminated poly(dimethylsiloxane) | RP 2.94 \( @ 1 \text{ kHz} \), EM 1--10 kPa | Area strain: 300 ±50% | Cutaneous haptics | Vatankhah--Varnoofsfaderani M. et al. [728] |
|        |                                             | UV-curable acrylic elastomer | CN 9021 oligomer (sartomer) with 5% HDDA (1,6-hexanediol diacrylate) | RP \( \approx 5 \) \( @ 1 \text{ kHz} \), EM 4.38 MPa | Area strain: \( \approx 90\% \) | -- | Niu X. et al. [724] |
| 3      | DEAs (chemical modifications to the polymer backbone) | Compatibilized-high-permittivity filler | Vinyl-terminated poly(dimethylsiloxane) with silica particles | RP \( \approx 5 \) \( @ 1 \text{ kHz} \), EM 250 kPa @ 39% filler loading | Transverse strain > 8% (filler loading 19.4%) | Kinesthetic haptics | Risse S. et al. [113] |
|        |                                             |                                     | Moieties with high dipole moment grafted to PDMS network | EM 20 V \( \mu \text{m} \)^{-1} @ 39% filler loading | Transverse strain < 4% | Kinesthetic haptics | Risse S. et al. [112] |
| 4      | DEAs (elastomer composites and blends)      | Conductive fillers | PDMS (Rhodorsil MF620U) with 2% FGS | EM \( \approx 15 \) \( @ 1 \text{ kHz} \), EM 0.99 Mpa | -- | -- | Romasanta L.J. et al. [146] |
|        |                                             | Elastomer blend | Poly(3-hexylthiophene) (6% loading) blended with PDMS (BJB enterprises) | EM \( \approx 13 \) \( @ 1 \text{ Hz} \), EM 46 kPa | Transverse strain: 2.3% @ 8 V \( \mu \text{m} \)^{-1} | Kinesthetic haptics | Carpi F. et al. [157] |
| 5      | DEAs (liquid fillers)                       | LM nanoparticles (100 nm) | PDMS (Sylgard 184) with 5--20% of eutectic gallium indium (EGaIn) | EM 4.13--7.62 \( @ 1 \text{ kHz} \), EM 1.54--1.98 Mpa | \( \approx 22.5 \) mN blocked force @ 53 V \( \mu \text{m} \)^{-1} | Kinesthetic haptics | Pan C. et al. [786] |
|        |                                             | Self-enclosed ionic liquid fillers | PDMS (Sylgard 184) with 20% loading of [1-ethyl-3-methyllimidazolium bis(trifluoromethylsulfonyl) imide, EMIMTFSI)] | EM \( \approx 7 \) \( @ 1 \text{ kHz} \), EM 0.01 Mpa | Transverse strain: \( \approx 6\% \) @ 3.55 V \( \mu \text{m} \)^{-1} | Cutaneous haptics | Ankit et al. [22] |
| 6      | IPMCs (commercially available)               | Sulfonylic \( \text{SiO}_2 \) doped | Nafion (DuPont) | IEC 1.66 meq \( g^{-1} \), EM 83 Mpa | Maximum deflection: 16.43 mm | Kinesthetic haptics | Han Y. et al. [300] |
|        |                                             | BaTiO\(_3\) doped (3 wt%) | Nafion (DuPont) | EM 103 Mpa | Maximum deflection: 4 mm | Kinesthetic haptics | Bian K. et al. [776] |
mechanical compliance is a critical requirement, to ensure minimal addition to stiffness, thus allowing exploitation of the full actuation potential of the underlying material. While flexible electronics material systems are usually subjected to modest strain levels between 6% and 10%, haptics and adaptable surfaces material systems are consistently subjected to high biaxial strain (100–300% strain) and innumerable deformation cycles and hence, the electrodes used should be mechanically robust and compliant, while maintaining acceptable conductivity across deformations. Here, stretchable electrodes are reviewed based on their type of conduction vis-a-vis electronic or ionic. Composite/hybrid electrodes are also reviewed.

6.1. Electronic Conductors

Stretchable electrodes such as carbon based, metal based, and LMs that rely on electronic conduction for their high conductivity are discussed in this section.

6.1.1. Carbonaceous Materials

Carbon-based electrodes such as carbon grease, carbon black, and ink formulations based on carbon black have been staple for the formation of compliant electrodes in reports of electrically driven actuators due to their low cost, ease of pattern ability, and acceptable conductivity (Figure 9A). Loose carbon powders used in initial demonstrations were soon replaced by carbon grease to overcome the detachment issues exacerbated during actuation. However, carbon grease electrodes are prone to diffusion in the underlying actuating membrane affecting reliability. In spite of the high compliance and acceptable conductivity, the reliability and robustness of such carbon-based electrodes are under scrutiny. An aging study was conducted on compliant carbon electrodes of various compositions and among them were commercial and solvent-diluted carbon grease, loose carbon black powder, carbon black powder suspension fabricated via manual application, and stamping on silicone-based DEAs. The DEAs were subjected to a 5% biaxial strain at 50 Hz for up to 40 million cycles and it was observed that manually applied carbon grease electrodes were the least reliable, with the shortest lifetime (<10⁵ cycles).

Single-layer carbon material, graphene, and its derivative graphene oxide have also been used to fabricate transparent and flexible electrodes. However, highly flexible (bending radius of up to 0.8 mm), single-layer films show degraded conductance even under moderate strains. To accommodate higher strains on actuators, Zhao et al. developed a “crumpled” graphene electrode (Figure 9B). CVD-grown graphene was transferred to a prestretched elastomer and by sequential biaxial relaxation which

| S. No. | Soft actuation mechanism (material approach) | Remarks | Active material matrix | Material properties | Actuation performance | Potential haptic application | Reference |
|--------|-------------------------------------------|---------|------------------------|---------------------|----------------------|-----------------------------|----------|
| 7      | IPMCs (novel polyelectrolyte membranes)    | Ionic-liquid electrolyte | poly[(r-butylstyrene)-b-(ethylene-r-propylene)-b-(styrene-r-styrene sulfonate)-b-(ethylene-r-propylene)-b-(r-butyl-styrene)] (SSPB)/BMIM TFSI composite | IEC  2.0 meq g⁻¹  λ 1.01 × 10⁻⁴ S cm⁻¹  SU 98%  EM 5 MPa | Maximum deflection: 12 mm | Cutaneous haptics | Lee J. W. et al. [202] |
|        | SWCNT doped                              |         | poly[1,4-phenyleneetherether-sulfone] | IEC  2.5 meq g⁻¹  λ 0.02321 S cm⁻¹  SU 80%  EM | Maximum deflection: 16 mm | Kinesthetic haptics | Khan A. et al. [193] |
| 8      | Electrothermally actuated LCEs            | LCE–Liquid metal composite | RM 257–ECaIN composite | Tubular actuator | EM 0.2–1.2 MPa | Linear Strain: 62% | Kinesthetic haptics | Ford M. J. et al. [210] |
| 9      | Electrically stimulated LCEs (electrostriction) | Nematic-isotropic genesis | 6-(4-cyano-biphenyl-4'-oxy)hexyl acrylate (AGOCB) | SWCNT-doped @ 100 °C | RM 82 and RM 2AE | EM 36 MPa | Contractive strain: 18% | Guin T. et al. [197] |
| 10     | ESAs                                     | Elastomeric shell filled with dielectric fluid | Envirotex FR3 (Cargill) within PDMS (Sylgard 184) | Elastomeric shell filled with dielectric fluid | EBS 33 kV mm⁻¹ | Area Strain: 46% | Cutaneous and kinesthetic haptics | Acome E. et al. [71] |
|        |                                          |         | Fluorinert FC-40 (3M) within PDMS (Sylgard 184) | EBS 18 kV mm⁻¹ | Linear strain > 50% | | Cacucciolo V. et al. [72] |
induced partial delamination of the graphene layer, a crumpled graphene electrode was obtained. Utilizing the crumpled graphene electrode in a DEA actuator configuration which unfolded during actuation, an area strain of 100% was achieved, with minimal change in conductivity. However, the maximum transparency during actuation was limited to 60%. In another report, by incorporating graphene scrolls formed in situ (during etching and transfer process) into multilayer graphene films, Bao et al. demonstrated highly stretchable (up to 120%) and transparent (>90%) graphene electrodes. The inclusion of such nano-scrolls ensured that the integrity of the conductive pathway was maintained on application of strain and resulted in only a 50% increase in resistance at 100% strain when compared with the 300% increase in resistance observed for multilayer graphene without scrolls. Graphene oxide can also be used to form compliant electrodes by improving the conductivity via reduction. Faul and coworkers demonstrated the use of trained conductive graphene oxide, reduced via in situ laser scribing, as the compliant electrode with no decrease in breakdown strength, as was observed in the trained control carbon grease electrode actuator which suffered terminal breakdown at 9 kV.

To capitalize on the conductivity of carbon-based electrodes, without additional processing steps required to accommodate high strains during actuation, CNT can be used. CNT networks are attractive candidates for the formation of compliant electrodes with high transparency achievable via thickness control. The alignment of CNTs within the network enables conduction under actuation-induced strain. Pei et al. demonstrated the self-clearing ability of SWCNT network, which limited the breakdown locally, thus providing improved fault tolerance compared with carbon grease electrodes, which suffered a terminal breakdown, in elastomer-based actuators. This self-isolation of faults provided by SWCNTs allowed for continued actuation even at high voltages albeit at a reduced strain. Utilizing nanometer-thick (<60 nm) functionalized SWCNT networks formed via Langmuir–Schaefer’s method, which reduces the stiffening effect on thin elastomers (6 μm), Shea et al. demonstrated 450 V-operated low-voltage-stacked DEAs (LVSDEAs). However, surface resistance was in the order of 0.3 Ωm²/μm at 0% strain. In the case of complaint CNT electrodes, high conductivity can only be achieved via increased thickness, which adversely affects the transparency of the electrode and adds to the stiffness of the actuator.

6.1.2. Metallic Materials

Metallic and transparent metal oxide thin films are widely used as electrodes primarily due to their high conductivity. However, they are unattractive as compliant electrodes due to their high Young’s modulus and low elasticity, which hamper actuation, resulting in microcracks when subjected to stress- or strain-induced deformation, which consequently drastically affects their conductivities. To overcome this hurdle, stretchable metallic electrodes (e.g., serpentine, nanomesh, corrugated electrodes) were fabricated by microfabrication techniques. The designs ensured that the electrodes were not directly subjected to the strain during the actuator operation, with the in-built patterned
structure taking the brunt of the strain induced. However, this is a process-intensive approach and results in reduced area coverage. Buckled electrodes, which allow for biaxial stretching with full area coverage, are another widely adopted technique for utilizing metallic contacts. Crumpled metal electrodes formed by electroless deposition of silver on the biaxially stretched elastomer exerted minimal stiffening effect and on actuation exhibited an area strain of 128% at 1.8 kV. Although the initial resistance was higher for crumpled silver (90 Ω) electrodes compared with the flat silver film (10 Ω), such electrodes could endure a radial strain of 110%, whereas the flat silver film lost conductivity at a mere 10% radial strain. Transfer of wrinkled silver electrodes due to their inherent compliance. The thin oxide skin can be achieved via imprint and stencil lithography and they can accommodate the deformation without interrupting the conduction path.

Conductive metallic 1D nanomaterials such as silver nanowires (AgNWs) have been widely researched and utilized as conductive, transparent electrodes for flexible electronics. AgNWs (Figure 9C) have now also emerged as promising candidates for the formation of transparent conductive electrodes for electrically driven haptics due to their impressive mechanical and electrical properties. Such materials are resistant to deformation-induced cracks, which plague metallic thin-film electrodes when subjected to high biaxial strains and cyclic bending (Figure 9D). With sufficient overlap ensuring a percolation network, 1D metallic electrodes can maintain their conductivity over a large range of strain and repeated bending cycles. A comparative study of stretchability of AgNWs and thin-film silver electrodes has been conducted by Oh and coworkers.

Although the thin-film silver electrodes demonstrated lower initial resistance ($R_\text{0} = 105 \, \Omega \, \text{sq}^{-1}$) compared with silver nanowire electrodes ($R_\text{0} = 0.001 \, \Omega \, \text{sq}^{-1}$), under deformation (bending radius of 1.2 and 4 mm) and cyclic bending tests, thin silver electrodes showed significant increase in resistance due to structural rupture right from the initial bending, whereas the resistance of the silver nanowire electrode remained approximately unaffected up to 20 bending cycles due to the nanowire network structure which accommodates the deformation without interrupting the conduction path.

Another class of materials, LMs, such as gallium and its alloys with their high conductivity ($10^6 \, \text{S cm}^{-1}$), conformability, and high deformability are attractive candidates for stretchable electrodes due to their inherent compliance. The thin oxide skin formed in ambient conditions allows for adherence to surfaces without hampering the conduction. Patterning can be achieved via imprint and stencil lithography and they are compatible with printing with resolution up to 100 μm. The ablative laser micromachining subtractive process has been used by Majidi et al. to form imperceptible LM circuitry with line widths of up to 4.5 μm on PDMS, where a biphasic deposition of LM (on thin Cr/Cu) ensured sufficient adherence. The printed LM lines maintained the low sheet resistance (2.95 Ω sq$^{-1}$) and transparency (85%) at 100% strain. Another important property, not demonstrated in solid metallic electrode, self-healing capability, has been demonstrated in LMs by Liu et al., where the in-plane self-healing capability of gallium-indium–tin alloy was unlocked via DEA actuation. The negligible stiffness imposed by LM electrodes enabled high actuation strains of up to 250%. On DEA actuation on the ruptured LM film, the attractive force acting on the opposing charges on either side of the rupture reduced the rupture distance and facilitated the breaking of oxide shell, which enabled the flow of LM to form a merging site that led to the healing of the electrode.

### 6.2. Ionic Conductors

Ionic conductors consist of polymer networks swollen with electrolyte or ionic liquids which enable ionic conduction. Their inherent stretchability and limited change in conductivity under high biaxial strains make them ideal candidates for compliant electrodes, in spite of the lower conductivity exhibited by them when compared with their electronic counterparts, which is attributed to slower migration of heavy ionic species. Their primary advantage over electronic conductors is the incomparable high transparency offered by such ionic conductors (Figure 10A).

#### 6.2.1. Hydrogels

Conductive hydrogels are generally formed by polymerization in aqueous electrolyte solutions, with the conduction provided by the ionic species in the electrolyte (e.g., Li, Na, K). Keplinger et al. demonstrated ionic gel conductors based on NaCl-loaded hydrogels, which were stretchable (up to 167%) and capable of operation at frequencies beyond 10 kHz and voltages above 10 kV. These hydrogel electrodes were highly transparent to all colors in both their unactuated and actuated states. The measured resistance–stretch curve of ionic hydrogel closely followed the $R/R_\text{0} = \lambda^2$ trend expected of an ideal stretchable electrode, where $R$ and $R_\text{0}$ is the resistance before and after the conductor is stretched $\lambda$ times its initial length. They also demonstrated the use of NaCl-loaded hydrogels in the fabrication of highly stretchable, biocompatible, and transparent sensory sheets, “ionic skin,” capable of measuring strains of up to 500%. The hydrogel fabricated using lithium chloride for ionic conduction and encapsulated between two dielectric elastomers was used by Yang et al. as compliant electrodes to demonstrate a DEA-powered electronic fish, where a DEA bending of around 26.9° was translated to complex undulatory motion for propulsion. Tough hydrogels with fracture energies of $\approx$9000 J m$^{-2}$ and high stretchability have been demonstrated by Suo et al., utilizing polyacrylamide: alginate composite.

Crosslinking in alginate hydrogel is ionic in nature, where the guluronic acid blocks the alginate crosslink through divalent cations, whereas the crosslinking in polyacrylamide hydrogel is covalent in nature. In the composite hydrogel, the covalent crosslinking provided high fracture resistance, whereas the unzipping of ionic crosslinking aided in high stretchability and mechanical energy dissipation. Kong et al. demonstrated continuous printing of tough hydrogel microfibers and 2D and 3D structures using the polyacrylamide:alginate composite hydrogel. When swollen in 0.01 or 0.1 KCl solution, conductive tough hydrogel microfibers (resistivity: $\approx$1.7 Ω m), which exhibit negligible change in resistance for high stretches of $\lambda = 11$, are obtained.
In spite of such attractive properties, the hydrophilic polymer chains of hydrogels can be adversely affected by harsh operation conditions such as high humidity, temperature, etc.

### 6.2.2. Ionogels

Ionic liquids, high ionic-strength salts, can be used in the polymer network to form ionogels, which retain compliancy and transparency offered by hydrogels, while providing environment stability. Volatility studies in the thermostatic chamber maintained at 100°C were conducted on ionogels and hydrogels. Hydrogels suffered a 93.69% decrease in weight in an hour, drastically affecting its reliability, whereas only a 12.43% decrease in weight (stabilized in 12 h) was observed for ionogels. Ionogels formed by acrylic acid polymerized in the ionic liquid 1-ethyl-3-methylimidazolium ethylsulfate ([C₂mim][EtSO₄]), which exhibited a transparency of 96.95%, conductivity of 0.23 S m⁻¹, and negligible change during stretches of λ = 4, were used as complaint electrode for dielectric elastomer transducers to achieve a 140% areal strain. The target ionogels were fabricated by a two-step method of photoinduced polymerization and solvent exchange. Utilizing Pluronic F127 (F127DA) with high molecular weight to crosslink poly(acrylic) acid in aqueous medium resulted in a highly porous structure with high swelling ability. This paved the way for a highly efficient solvent exchange when immersed in 1-ethyl-3-methylimidazolium dicyanamide ([EMIm][DCA]) room-temperature ionic liquid, and this resulted in tough, fatigue-resistant ionogels with high conductivity (1.9 S m⁻¹) and stretchability of ≈850%. Self-healing capability of such ionic conductors has been reported by Cao et al., through harnessing the reversible ion–dipole interaction observed in a poly(vinylidene fluoride-co-hexafluoropropylene) (PVDF-co-HFP) polymer network crosslinked with 1-ethyl-3-methylimidazolium trifluoromethanesulfonate (EMIOTf) ionic liquid (Figure 10B). The use of copolymer with higher than typical HPF content (45%) which exhibited high dipole moment (5.507 Debye) and inclusion of high ionic strength ionic liquid, the polymer chains were crosslinked by reversible ion-dipole interactions and the ionic liquid plasticized the polymer chains to a glass transition temperature lower than room temperature, which enabled self-healing at room temperature. The resultant ionic conductor was capable of enduring strains exceeding 5000%, while maintaining high conductivity (10⁻⁴ S cm⁻¹) and transparency of 92%. Recently, Jiang and coworkers reported a hydrogen bonding-enabled (PEA)-based elastomer:1-ethyl-3-methylimidazolium bis-(trifluoromethylsulfonyl)imide

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**Figure 10.** Ionic conductors and composite/hybrid systems. A) Performance of hydrogels in comparison with various other conductive electrodes. Hydrogels provide the highest transmittance at 550 nm with acceptable conductivity and unparalleled compliance. Reproduced with permission.[246] Copyright 2013, The American Association for the Advancement of Science B) Ionic conductor with self-healing properties via ion–dipole interactions, capable of self-healing in 24 h at room temperature. Reproduced with permission.[253] Copyright 2017, John Wiley and Sons. C) Micropatterns formed by laser ablation of carbon–silicone composite bonded to thick and thin elastomers. Reproduced with permission.[256] Copyright 2015, American Chemical Society. D) Cross-sectional SEM images of MXene/PEDOT:PSS composite electrodes with 1:2 and 1:4 ratios. Schematic of origami-inspired narcissus flower robot operation, which “blooms” on application of 3 V DC input. Reproduced with permission.[267] Copyright 2019, The American Association for the Advancement of Science.
Composite/hybrid configurations exploiting the features of various material systems to enhance the compliance and stretchability of electrodes have also been widely reported. As a replacement of pure carbon-based electrodes, conductive rubbers, carbon–polymer composites utilizing silicone, PDMS, etc. have been demonstrated, where the polymer encapsulation overcame the diffusion and displacement issues and resulted in improved adhesion and reliability. These polymer–carbon composites, inherently thicker than pure carbon-based electrodes, are however more suitable for thicker actuation layers as their contribution to stiffness during actuation is non-negligible. Such composite electrodes are also compatible with high-resolution (sub-200 um) patternability via laser ablation, as was demonstrated on a PDMS–carbon-compliant electrode (Figure 10C). This scalable technique utilized optimized plasma treatment to improve adhesion to the silicone elastomer layer without drastic increase in stiffness. Polymer matrices have also been successfully utilized for improving the stretchability of CNT electrodes. Pei et al. reported the use of the interpenetrating SWCNT–polyurethane bilayer electrode, where the water-based polyurethane acted as a protective overlayer to CNT against terminal failure due to corona discharge in air, with negligible effect on self-clearing, conductivity, and actuation, enabling stable, continuous actuation (5.5 h) at constant voltage and 150% area strain for 1000 cycles. Nanowire–polymer composite systems are also widely used as compliant electrodes. One of the earlier demonstrations for actuators was utilizing a silver nanowire–polymer composite, consisting of an ultrathin layer of silver nanowire in a poly(tert-butylacrylate-co-acrylic acid) poly(TBA-co-AA) layer, where the electrode retained electrical conductivity at 140% strain albeit with a resistance increase of 10–100 times. Carbon-based electrodes can also be used to improve the conductivity of the AgNW network, as reported by Lee et al., where an addition of a mere 5 ng/mm² of CNT on the AgNW network leads to a decrease in resistance from KΩ sq⁻¹ to 3 Q sq⁻¹ with only 11% decrease in transparency. In addition, an increase in strain (from 35% to 146%) and breakdown field (from 70 to 128 V μm⁻¹) with the addition of CNT was also observed, which was attributed to the effective increase in electrode area, improving the electronic flux density, and more uniform distribution of electrical and thermal stresses, decreasing electromigration-induced stress. Capillary force-induced cold welding was also utilized to fabricate dense AgNW/SWCNTs, where the capillary force exerted during removal of water, during the moisture exposure-drying step, induced a tight welding of the AgNW/CNT stacked network. The resultant composite electrode exhibited 42% increase in resistance under high uniaxial strain of 580%, with a 100% increase in resistance under 200% biaxial strain.

Recently, Cao et al. demonstrated a MXene/rGO composite electrode capitalizing on the high conductivity of MXene, while utilizing large flake size-compliant rGO to maintain the integrity of the composite electrode under large strain. Such electrodes (with 50% rGO) fabricated in a wrinkled/ridge configuration, on a 100%, 200%, and 300% pretrained elastomer substrate, allowed tolerance to uniaxial strain of up to 250% and maintained the resistance (150 Ω) when subjected to 1000 stretching cycles, whereas pure MXene films displayed microcracks right during the wrinkled/ridge fabrication stage. Polymer matrix can also be used for metallic conductors to improve the compliance of electrodes. The stretchable polymer–metal composite conductor was demonstrated by Kotov et al., utilizing a polymer: Au composite. Layer-by-layer fabrication approach exploiting the positively charged polyurethane and negatively charged citrate-coated Au nanoparticles enabled uniform and high nanoparticle loading. Reconfigurable nanoparticle organization during stretching ensured good performance under stress, with conductivity decreasing from 11000 to 2400 S cm⁻¹ at 110% strain. A facile polymer matrix-independent route for forming a conductive polymer–silver composite electrode was reported by Wu et al., iodizing silver flakes with KI followed by photolysis via sunlight converted the silver iodide and surface lubricant to silver nanoparticles, primarily located on individual flakes. The improved percolation pathway provided by these nanoparticles leads to a resistivity of 9.43 × 10⁻⁴ Ω cm for 75 wt% silver flake/silver nanoparticle filler loading in PDMS. The matrix-independent behavior was demonstrated by successfully incorporating such silver flake/silver nanoparticle fillers in silicone matrices (Ecoflex 00-30, Sylgard 184, Silbione LSR 4305), isobutylene–isoprene rubber (IIR), styrene butadiene rubber (SBR), nitrile rubber (NBR), and butadiene rubber (BR), thus showing the suitability of its incorporation in any substrate-compatible polymer matrix which guarantees good adherence. Even at 119% elongation, such systems limited the increase in resistivity to three times the prestretched values. Composite/hybrid electrodes can also be used to provide a superior performance in comparison with individual components by virtue of material interaction. MXene (Ti₃C₂) crosslinked with PEDOT: PSS was utilized as electrode to form highly robust, artificial muscles, where the PEDOT:PSS molecules intercalated between the MXene sheets, increasing the electrode surface area, while simultaneously preventing restacking of MXene sheets (Figure 10D). High conductivity was obtained via change in the electronic structure of MXene upon ionic crosslinking and by the linearly aligned PEDOT quinoid phase. When used as electrodes for actuation, maximum strain of ~1.3% at 1 V input and long-term cyclic stability (97% upto 18 000 cycles) were achieved.

By using such hybrid/composite structures, highly compliant transparent electrodes with robust performance can be achieved.

7. Perspectives and Future Directions

Applications such as VR/AR, microrobotics, touch surfaces, and Assistive technologies for visually impaired are set to benefit
most from technological advancements in the field of tactile feedback. However, they set out certain requirements for the next generation of haptic technology: 1) ability to convey an array of tactile information like vibrational feedback, texture changes, and force feedback using the same device for an immersive user experience; 2) untethered operation to allow mobility of the user; 3) compact and sleek form factor to enable seamless integration with touch-intensive devices like display kiosks; 4) energy efficiency to enable long-term portable usage; 5) ability to provide localized feedback to enable discerning location; and 6) ability to be manufactured as large-area devices with possibility of pixelation. In addition, static and dynamic physical response produced by these haptic feedback devices is crucial, together with achieving compliance matching between them and the skin. Future haptic devices have to meet all or few of these requirements.

Electrically driven actuation mechanisms for soft materials appear to be well placed to meet these challenges. However, going beyond the improvement in material properties of active layer and compliant electrode material, there are new opportunities to improve upon the current generation of haptic feedback devices. These opportunities lie in material innovations, utilization of advanced fabrication techniques, and imbining control strategies to optimize performance.

7.1. Material Innovation for Improved Function

Instilling self-healing properties in soft actuators is extremely desirable to improve lifetime and robustness. Ideally, self-healing of soft actuator devices would require both compliant electrodes and active layers to possess the self-healing property. However, much of the focus has been on self-healing of the mechanical and electrical properties of compliant electrodes. There is a need to mimic the ability of self-healing in the active layer as well. To this end, few recent works have tried to achieve self-healing of the active layer. Li et al. reported that a PDMS-based elastomer that was able to self-heal in 72 h by incorporation of coordination complexes within the PDMS matrix (Figure 11A). Wang et al. utilized reversible disulfide bonds as a tool to induce self-healing within LCEs (Figure 11B).

In HASEL actuators, the use of a liquid dielectric instead of conventional elastomeric layers incutlade self-healing abilities (Figure 11C). The system shows resilience against electrical damage and actuates even after several counts of electrical breakdown attributed to the natural healing ability of the liquid layer. Our group has reported a self-healable transparent electrode based on a combination of AgNWs and a polyurethane-based elastomer with the ability to self-heal via the reversible Diels–Alder reaction (Figure 11D). In these examples, the incorporation of self-healing abilities has shown no deterioration of mechanical properties and has even enhanced the actuation performance in some cases. Even with these few demonstrations, there are still a number of avenues to explore. Rapid, low-temperature healing without any external aid is a distant milestone. In addition, tailoring self-healing materials into the form factor required for haptic devices is challenging.

Another interesting material innovation is to imbibe a material with the ability to modulate its mechanical property on demand. The modulation of rigidity in soft actuators is extremely interesting as it allows them to mimic nature more closely. Being able to produce fluidic actuation and concurrently modulate their mechanical properties would imbibe soft actuators with greater operational dexterity and versatility. Tonazia et al. demonstrated a variable-stiffness fiber (Figure 11E) by incorporating a low-melting-point alloy within silicone tubes. The fiber was able to exhibit stiffness modulation, bearing 200 g of load at temperatures below the phase-change temperature. Piskarev et al. also incorporated phase-change LMs for modulation between flexible and rigid states (Figure 11F). However, most of these systems are augmentative in nature. The reversible rigidity mechanism works independent of the actuation mechanism and does not contribute directly to actuation performance. In addition, they can make the systems bulky. Future efforts would involve exploring opportunities for synergistic combination of both systems and the replacement of high-cost rigidity modulation materials.

7.2. Development of Additive Fabrication Techniques

To match the spatial density and complexity of human tactile sensing physiology, an arrangement of an array of high-performance haptic devices is required. While the performance part can be addressed by innovations in material systems, the arrangement of a dense array of devices can be tackled by innovations in fabrication techniques. Flexibility provided by recent progress in digital printing techniques allows for combining the deposition of active material and compliant electrodes in an additive manner. This kind of additive fabrication technique also allows for a free-standing design with a high-resolution, dense array of actuators distributed over a large area. This would enable futuristic haptic feedback devices for wearables and prosthetics.

Techniques such as screen printing, gravure printing, and spray coating have been utilized for printing of electrodes. However, these techniques are not versatile and do not provide the capability of making changes on the go. There are a variety of other mainstream printing techniques that could prove useful for the development of complete haptic feedback devices. Different materials can be deposited using these techniques, depending on the viscosity of the material to be printed, composition of ink formulations, mechanical properties, etc. Lower-viscosity material can be printed by techniques like inkjet printing, stereolithography, and electrohydrodynamic printing, whereas the higher-viscosity material can be printed by extrusion techniques (material formulation could be in the form of filament, gel, or paste) and laser sintering. Apart from the material properties, the desired feature size that would be used for deposition of active and electrode material will also help in defining the suitable printing technique to be used.

Drop-on-demand techniques (Figure 12A) are noncontact in nature and can enable material deposition on rough surfaces. Feature size is dependent on the volume of droplet and interaction of droplet with the substrate. In extrusion techniques, material is directly in contact with the surface while being deposited. This provides better quality of the film with higher-viscosity material, but the feature size is limited by the inner diameter of the nozzle. Photopatterning techniques use light to crosslink...
polymers. The technique limits the type of material used (as it needs photoinduced crosslinking) and does not allow for printing multiple materials in the same layer. Ladd et al. demonstrated direct writing of LM microstructures at room temperature for applications such as electrical interconnects (Figure 12B).[277] Skylar–Scott et al. recently demonstrated multimaterial multinozzle printing of soft materials (Figure 12C), controlling the composition and structure of materials at a microscale.[278]

Although the onset of these printing and material deposition techniques for multimaterial actuators has huge potential, there are still challenges that remain to be solved. There are critical concerns associated with multimaterial printing like delamination between layers due to mismatch in material properties, fidelity in replication of design, and autoadjustable surface printing on variable surfaces.[275] In addition, tuning of ink formulations also needs to be done so that different material systems could be adapted for printing on a single setup.

7.3. Electrical Control Strategies

Although there has been much progress in new materials for soft actuators, there needs to be concurrent development in electrical control strategies as well. There are three major concerns with electrical control. 1) Much of the reports involve the actuation process running in open loop, that is, without any sensors. To move toward a closed-loop mechanism, actuators need to be combined with sensors to provide feedback and adjust for the errors. 2) Applications such as Braille and surface haptic displays require a large number of independent actuators. These applications pose additional concerns for control such as slow response time, “bulkiness” of circuit, and addressing individual devices in an array of actuators. 3) To move the haptic feedback devices closer to the complexity and attributes of biological tactile sensing, efficient neuroinspired control strategies for these soft actuators are required.
Self-sensing is a mechanism that involves sensing the current state of the actuator, without the need for additional sensing elements. Typically, self-sensing can be achieved through two approaches: 1) an integrated device with actuating and sensing parts separated by patterned electrodes and 2) estimation of the change in physical parameters of the actuating electrodes/device, that is, electrode resistance or impedance–capacitance of the device which is affected by deformation.[279] For a closed-loop system, the actuators and sensors should function in tandem. Gisby et al. developed an algorithm for self-sensing, which could detect capacitance of the actuated DEA along with the series resistance of the electrodes and the leakage current through the active material.[280] Acome et al. also demonstrated the self-sensing of HASEL actuators using a similar principle (Figure 13A).[71] The capacitive measurement was enabled by pulse width modulation (PWM), which simultaneously generated a high voltage for actuation superimposed with a high-frequency, low-amplitude AC voltage signal for sensing. A custom program and digital acquisition system were used to output combined actuation and sensing signals and receive signals from a high-voltage amplifier. Rosset et al. demonstrated a closed-loop actuation control of DEAs using the self-sensing strategy.[281] IPMCs also exhibit self-sensing behavior as an electric current is generated in IPMCs in response to the bending moment (Figure 13B).[282] This generated voltage (tens of millivolts) for large bending displacements could enable self-sensing.

Local control via an array of independent actuators was demonstrated by Marett et al. by use of flexible high-voltage thin-film transistors (HVTFTs) to control a 4 × 4 array of DEAs (Figure 13C). The system used a single high-voltage power supply and zinc–tin oxide (ZTO)-based HVTFTs. Using a gate voltage of 30 V, switching between pixels consisting of DEAs operating at excess of 1 kV was demonstrated. Smith et al. also demonstrated a high-voltage organic thin-film transistor using pentacene as the active layer.[284] However, these are proof-of-concept experiments and most of the current generation of DEAs still require high voltages. Further scaling down for micro-DEAs will lead to higher electric fields with smaller transistor dimensions.

Recently, a neuromorphic approach for controlling electrically driven soft actuators has also been explored. Lee et al. have shown an electronic synapse functionalized with an artificial sensory receptor and an artificial motor unit as bioinspired soft electronics (Figure 13D).[285] The voltage pulses from self-powered photodetector triggered by optical signals drive a stretchable organic nanowire synaptic transistor; the resultant synaptic outputs are used for optical wireless communication and light-dependant actuation of artificial muscle, similar to biological muscle movements. Other synaptic studies have also been
conducted in recent years for control of robotic components. With very small power consumption and capability to integrate on the device itself, these sensorimotor synapses could be promising for further development of electrically driven soft actuators.\cite{286}

Furthermore, neuromorphic approach has been explored the other way around as well, by mimicking the tactile signals similar to the response produced by mechanoreceptors and using them to drive artificial synapses. Contact electrification-activated artificial afferents have been demonstrated as triggers for manipulating synaptic plasticity in sensory neuromorphic.\cite{287,288} Strain pulses acting as perceptual inputs have been utilized to modulate the synaptic weights in an artificial sensory synapse utilizing piezoelectric nanogenerators.\cite{289} The strain-triggered piezopotential is capacitively coupled to a graphene synaptic transistor via an ion gel (Figure 13E).\cite{287} By replacing the conventional voltage-driven control with haptics, a substantial reduction in power is also achievable, where energy dissipation is limited to 11.9 fJ per spike for contact electrification (triboelectric)-activated artificial afferent system consisting of an ion-gel-gated MoS\textsubscript{2} transistor.\cite{290} In addition to providing the external mechanical stimuli’s spatiotemporal information, these haptic components also provide self-activation of the afferent system by acting as the power source for the synaptic transistor.

8. Summary

In summary, haptics has a crucial role in a variety of applications such as VR/AR and touch-sensitive devices. These applications necessitate untethered operation and compact design for the haptic feedback devices. In addition, the physiology of the human tactile sensing mechanism requires compliance matching between the tactile devices and human skin. Electrically driven actuation mechanisms utilizing soft and compliant materials are well equipped to mitigate these challenges and enable the next generation of haptic feedback devices. In this Review, we have covered the electrically driven actuation mechanisms of soft
materials. In these electrically driven soft actuators, there exists an active layer, which responds to electrical stimuli supplied by compliant electrodes. The material composition of both the active layer and compliant electrodes plays a crucial role in the actuation behavior and performance of these soft actuators. There have been significant research efforts in enhancing the electromechanical properties of these soft actuators via modifications in active layer and compliant electrodes.

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Conflict of Interest

The authors declare no conflict of interest.

Keywords

electroactive polymers, haptics, smart materials, soft robotics, tactile feedback

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[1] B. Hannaford, A. M. Okamura, Springer Handbook of Robotics (Eds: B. Siciliano, O. Khatib), Springer International Publishing, Cham 2016, pp. 1063–1084.
[2] S. Biswas, Y. Visell, Adv. Mater. Technol. 2019, 4, 1900042.
[3] L. Skedung, M. Arvidsson, J. Y. Chung, C. M. Stafford, B. Berglund, M. W. Rutland, Sci. Rep. 2013, 3, 2617.
[4] R.S. Dahiya, G. Metta, M. Valle, G. Sandini, IEEE Trans. Robot. 2010, 26, 1.
[5] J. Yin, R. Hinchet, H. Shea, C. Majidi, Adv. Funct. Mater. n.a., 20074128.
[6] R. Dahiya, N. Yogeswaran, F. Liu, L. Manjakkal, E. Burdet, V. Hayward, H. Jortnerl, Proc. IEEE 2019, 107, 2016.
[7] X. Yu, Z. Xie, Y. Yu, J. Lee, A. Vazquez-Guardado, H. Luan, J. Ruban, X. Ning, A. Akhtar, D. Li, B. Ji, Y. Liu, R. Sun, J. Cao, Q. Huo, Y. Zhong, C. Lee, S. Kim, P. Gutfre, C. Zhang, Y. Xue, Q. Guo, A. Chempakasseril, P. Tian, W. Lu, J. Jeong, Y. Yu, J. Cormann, C. Tan, B. Kim, K. Lee, X. Feng, Y. Huang, J. A. Rogers, Nature 2019, 575, 473.
[8] Ankit, N. Tiwari, M. Rajput, N. A. Chien, N. Mathews, Small 2018, 14, 1702312.
[9] S. Okamoto, H. Nagano, Y. Yamada, IEEE Trans. Haptics 2013, 6, 81.
[10] A. Billard, D. Kragic, Science 2019, 364, 6041.
[11] I. Poupyrev, S. Maruyama, J. Rekimoto, Secondary, Association for Computing Machinery, Paris, France 2002, pp. 51–60.
[12] C. Basdogan, F. Giraud, V. Levesque, S. Choi, IEEE Trans. Haptics 2020, 13, 450.
[13] C. Pacchierotti, S. Scheggi, D. Prattichizzo, S. Mishra, Front. Robot. AI 2016, 3.
[14] P. B. Shull, D. D. Darmian, J. NeuroEng. Rehabil. 2015, 12, 59.
[15] F. Sorgini, R. Caliò, M. C. Carrozza, C. M. Oddo, Disabil. Rehabil. Assist. Technol. 2018, 13, 394.
[16] Ankit Ankit, J. Y. Chan, L. L. Nguyen, F. Krisnadi, N. Mathews, Feby Krisnadi, Nirapan Mathews, Proc. SPIE 10966, Electroactive Polymer Actuators and Devices (EAPAD) XXI, 2019, 10966.
[17] Ankit Ankit, A. C. Nguyen, N. Mathews, Nirapan Mathews, Proc. SPIE, Electroactive Polymer Actuators and Devices (EAPAD), 2017, 10163.
[18] P. Delmas, J. Hao, L. Rodat-Despoix, Nat. Rev. Neurosci. 2011, 12, 139.
[19] D. Rues, M. T. Tolley, Nature 2015, 521, 467.
[20] C. Majidi, Adv. Mater. Technol. 2019, 4, 1800477.
[21] G. M. Whitesides, Angew. Chem., Int. Ed. 2018, 57, 4258.
[22] Ankit, Ankit, N. Tiwari, F. Ho, F. Krisnadi, M. R. Kulkarni, L. L. Nguyen, S. J. A. Koh, N. Mathews, ACS Appl. Mater. Interfaces 2020, 12, 37562.
[23] A. Delalleau, G. Josse, J.-M. Lagarde, H. Zahouani, J.-M. Bergheau, J. Biomech. 2006, 39, 1603.
[24] G. J. A. D. Purves, F. David, L. C. Katz, A-S. LaMantia, J. O. McNamara, S. M. Williams, Sinanor Associates, Sunderland, MA 2001.
[25] H. Phung, C. T. Nguyen, T. D. Nguyen, C. Lee, U. Kim, D. Lee, J.-D. Nam, H. Moon, J. C. Koo, H. R. Choi, Meccanica 2015, 50, 2825.
[26] L. Crampfnorn, B. Ward-Cherrier, N. F. Lepora, IEEE Robot. Automat. Lett. 2017, 2, 1336.
[27] J. C. Craig, K. B. Lyle, Percept. Psychophys. 2001, 63, 337.
[28] J. C. Craig, J. M. Krisner, Somatosens. Motor Res. 1998, 15, 29.
[29] R. S. Dahiya, M. Valle, Robotic Tactile Sensing, Springer Netherlands, Springer, Science+Business Media Dordrecht, 2012, 19–41.
[30] R. L. Klatzky, S. J. Lederman, Handbook of Psychology, I.B. Weiner (Ed.), pp. 147–176, https://doi.org/10.1002/0471264385.wei0406.
[31] J. C. Craig, X. Biaiha, Percept. Psychophys. 1990, 47, 22.
[32] S. J. Bensmaia, J. Craig, K. O. Johnson, J. Neurophysiol. 2006, 95, 1783.
[33] RH. LaMotte, MA. Srinivasan, J. Neurosci. 1987, 7, 1672.
[34] P. S. Khalsa, R. M. Friedman, M. A. Srinivasan, R. H. Lamotte, J. Neurophysiol. 1998, 79, 3238.
[35] P. Jenmalm, I. Birznieks, A. W. Goodwin, R. S. Johansson, Eur. J. Neurosci. 2003, 18, 164.
[36] S. J. Lederman, D. T. Pawluk, Advanced Tactile Sensing for Robotics, World Scientific 1992, pp. 151–192.
[37] C. J. Cascio, K. Sathian, J. Neurosci. 2001, 21, 5289, https://doi.org/10.1523/JNEUROSCI.21-14-05289.2001.
[38] J. Scheibert, S. Leurent, A. Prevost, G. Debreges, Science 2009, 323, 1503.
[39] S. J. Lederman, Bull. Psychon. Soc. 1981, 18, 253.
[40] A. M. Smith, C. E. Chapman, Mélanie, Deslandes, J.-S. Langlais, M.-P. Thibodeau, Exp. Brain Res. 2002, 144, 211.
[41] I. B. O. Bodegård, A. Ledberg, S. Geyer, E. Naito, K. Zilles, P. E. Roland, J. Neurosci. 2000, 20, RC51.
[42] D. Accotto, F. Damiani, R. Sahai, D. Campolo, E. Guglielmelli, P. Dario, presented at Proc. 2007 IEEE Int. Conf. on Robotics and Automation, April 2007.
[43] D. Accotto, R. Sahai, F. Damiani, D. Campolo, E. Guglielmelli, P. Dario, Sensors Actuat. A: Phys. 2012, 187, 201.
[44] G. Cadoret, A. M. Smith, J. Neurophysiol. 1996, 75, 1963.
[45] R.S. Johansson, G. Westling, Exp. Brain Res. 1984, 56, 550.
[46] R. S. Johansson, J. R. Flanagan, Nat. Rev. Neurosci. 2009, 10, 345.
[47] E. Olivier, M. Davare, M. Andres, L. Fadiga, Curr. Opin. Neurobiol. 2007, 17, 644.
[48] J. Sutherland, J. Belec, A. Sheikh, L. Chepelev, W. Althoibaby, B. J. W. Chow, D. Mitsouras, A. Christensen, F. J. Rybicki, D.J. La Russa, J. Digital Imag. 2019, 32, 38.
[49] A. Blaga, C. Militaru, A.-D. Mezii, L. Tarnas, Robot. Comput. Integr. Manufact. 2021, 68, 102057.
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