Low-Voltage Soft Actuators for Interactive Human–Machine Interfaces

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Soft electrical actuators driven by low voltages are promising for interactive human–machine interfaces (iHMI) applications including executing orders to complete various tasks and communicating with humans. The attractive features of low-voltage soft electrical actuators include their good safety, low power consumption, small system size, and nonrigid or deformable characteristics. This review covers three typical classes of electrical actuators, namely, electro-chemical, electrothermal, and other electrical (dielectric, electrostatic, ferro-electric, and plasticized gel) actuators according to their mechanism and working potential range. For each kind of actuators, the advantages, working principle, device configuration/design, materials selection, recent progress, and potential applications for iHMI are summarized, with the strategies for enhancing the actuation performance under low voltages being highlighted. Finally, the challenges for those soft actuators and possible solutions are discussed.

1. Introduction

Interactive human–machine interface (iHMI) is an emerging bidirectional interface that enables direct communication between humans and machines to perform specific operations efficiently and complete complex tasks.[1,2] At this interface, electronic systems sense human interactions generally through physical or electrophysiological parameters and return an interactive feedback that stimulates various human sensations. Conventionally, iHMI has taken various forms from rehabilitation robots to touch panels, at which rigid sensors and actuators are typically utilized.[3–5] However, stemming from the mechanical mismatch with the human body, these rigid components are often uncomfortable, hinder the mobility of users, display signal instabilities from motion, and reduce mechanical transmissions to users.[6] To overcome these limitations, next-generation iHMI has adopted soft, flexible, and stretchable devices that can conform to curved and dynamic surfaces. At the same time, by matching the compliance of the skin, safe and realistic interactions between human and machines are realized, thereby widening the range of applications that iHMI can achieve.[6]

Soft actuators have played a critical role within iHMI systems, allowing machines to engage humans through physical interaction. Owing to their development, significant efforts have been realized toward designing next-generation interactive electronic skins, epidermal electronics, haptic systems, soft robots, and medical rehabilitation devices.[3,7–12] For example, combination of stretchable epidermal electronics (e.g., sensors and circuits)[13,14] with soft actuators could lead to interactive systems applied on the skin, which allow sensing physiological information from skin[15,16] and generating tactile/stretching feedback[11] or executing an order (e.g., delivering a drug into the skin).[16] Utilizing soft materials such as hydrogels, elastomers, fluids, films, yarns, textiles, and paper, soft actuators can be activated by various stimuli, including thermal, magnetic, light, hydraulic, pneumatic, and electric fields.[8,17–19] Among those, soft electrical actuators allow simple and convenient control of their actuation behavior just by varying the applied electrical field over a range of voltage/current amplitudes and frequencies. Moreover, these electrical actuators can be integrated with well-established power sources and driver circuits to achieve untethered portable devices.[8,17,20] This simple integration can also lead to a much smaller volume/lighter weight of the actuation system as compared with the counterparts with traditional soft hydraulic, pneumatic, light, and magnetic actuators, which require bulky sources (e.g., liquid/gas tanks, bulbs, magnets) and control units (e.g., valves).

Toward designing these electrically driven soft actuators for iHMI, it is critical to ensure that users are not exposed to electrical discharges upon interaction where burns and even cardiovascular injuries may occur.[21,22] Generally, the mode and severity of these electrical injuries depend on the voltage of the electrical current and time of exposure. As such, high-voltage (>1000 V) injuries have often been related to a large amount of morbidity and mortality where amputations, cardiac arrest, severe burns, and long-term psychological issues have been observed.[21–24] In comparison, low-voltage injuries (<1000 V) have seen lower severities, but specific cases of lung and muscle injuries have been reported and thus should not be underestimated.[25–28] Therefore, it is imperative to utilize and...
continuously design low-voltage soft actuators which may provide significant protection to users and allow easier commercial adoption. Concurrently, when determining the efficacy of soft actuators for iHMI with human subjects, additional safety protocols should be considered such as insulating layers and current limits.\textsuperscript{[29,30]} Safe limits of dielectric elastomer actuators (DEAs) were investigated by Pourazadi et al. who proposed that electrical currents of devices should be limited to below 20 mA where no potential harm toward ventricular fibrillation occurs.\textsuperscript{[29]} Also, for safe electrical discharge, critical capacitance of the device should be calculated based on the minimum resistance load.

In this article, we review the progress of various electrical soft actuators and the strategies in lowering their voltages for iHMI applications. The scope of this review is therefore different to reviews and reports that focus on deformable conductors,\textsuperscript{[31,32]} electronic devices and sensors for human–machine interfaces,\textsuperscript{[31,32]} and soft actuators for soft robotics and small-scale robots.\textsuperscript{[8,17]} The article provides a panoramic overview on three categories of soft electrical actuators in three sections: electrochemical actuators (ECAs), electrothermal actuators (ETAs), and other electrical actuators (dielectric, electrostatic, ferroelectric, and plasticized gel actuators) according to the actuation mechanism and working voltage range. Each category will introduce the working principles, highlight the progress towards enhancing the actuation performance under low voltages, and show their potential applications towards iHMI. The challenges and outlook for each group of electrical actuators will also be provided in the final section.

## 2. Soft ECAs

ECAs are usually assembled from two electrodes and an electrolyte. They can generate various deformation and/or force under a low electrical voltage stimulation due to electrochemical processes occurred in the electrodes and electrolyte. As compared with other actuators, ECAs are advantageous in many aspects. They are generally assembled from soft materials and can provide more conformal interface with the targets/substrates than bulk mechanical actuators. ECAs possess a simple configuration and can be driven by small batteries, leading to a smaller system size and a lower weight than mechanical/pneumatic/hydraulic/magnetic actuators. As a low voltage (usually <10 V for a single device) is required for ECAs, they are much safer than high-voltage-driven dielectric actuators. In contrast to thermal actuators that dissipate a lot of heat during actuation, ECAs produce little heat and have low energy consumption because much energy can be stored in them during actuation and released to loads during relaxation. Consequently, ECAs have promising applications in soft robotics, tactile/brail displays, portable electronics, and biomimetic plants/animals.

### 2.1. Working Principle

The working mechanism of ECAs is based on charge injection induced electrochemical processes (e.g., formation of electrochemical double layers or redox reactions), which involves the electric-field induced movement of electrolyte ions between the electrode and (gel) electrolyte and possible physical change (size, morphology/conformation) in the active layer and electrolyte. The exact mechanism is dependent on the device configuration and the composition of the electrodes and electrolytes, which can be tailored by the prescribed actuation behavior and even structural evolution of a single electrode and the gel electrolyte. The actuation behavior of a single electrode is important and can be studied in a three-electrode cell\textsuperscript{[33]} with an active electrode, a counter electrode, a reference electrode, and a liquid electrolyte that guide the design of practical ECAs for generating bending/torsion/extension/contraction deformation and force. The actuation performance (strain, speed, stress, and cycling stability) of ECAs is greatly affected by the electrodes, electrolyte, and their interface bonding.

Take a bendable ECA as example, it usually consists of two active electrode strips/films sandwiching a gel electrolyte membrane (Figure 1). Assuming the active electrodes and the polymer in the gel electrolyte are all nonionic, the applied voltage will drive the cations and anions (either bare or solvated) in the gel electrolyte to adsorb onto/insert into the cathode and anode, respectively, and generate different lengthwise expansion strains determined by the size of the ions and the amount of injected charges. If the cation is larger (or smaller) than the anion, the lengthwise expansion of the cathode will be larger (or smaller) than that of the anode, leading to a bending of the device toward the anode (or cathode). If the active material contains dissociable ions such as doped conductive polymers, the movement of those ions is much more complicated, but the bending actuation is still caused by the strain difference between the cathode and the anode. If the gel electrolyte films are ionic polymer gels, they may bend themselves under a voltage due to the gradient distribution of free ions across the films.\textsuperscript{[34]} Apart from the ion movement induced strains, the injection of electrons/holes into the electrode may change the bond length of active materials (such as graphite and carbon nanotubes) and induce an expansion/contraction strain on them.\textsuperscript{[33]} Considering all the factors, the actual bending of an ECA is a result of the ion redistribution and the structural change in the active material and the electrolyte. The performance of ECAs based on some typical active materials and electrolyte membranes is summarized in Table 1 at the end of Section 2.3.

![Figure 1. Schematic illustration of a typical bendable ECA. In this case, the gel electrolyte is composed of a nonionic polymer matrix filled with electrolyte, where the cation is larger than the anion. The ion insertion into the electrodes causes a strain difference between them, which leads to the bending of the ECA toward the anode side.](image-url)
2.2. Electrode Design

The actuation performance of ECAs is greatly affected by the actuation behavior of the active electrodes, which can be determined by their mechanical strength/modulus, electrical conductivity, ion storage capacity, ion transport ability, and the orientation of active materials. Generally, the electrodes should have a high electrical conductivity for electron/hole transport, a porous structure for fast ion diffusion and ion storage, and a low (high) modulus for generating high deformation (stress). These properties are determined by the active material and the structure of the electrode. Therefore, rational selection of active materials and design of the electrode structure are critical to construct high-performance ECAs. Active materials should have a high specific surface area for ion storage through surface adsorption or redox reactions and preferably high electrical conductivity. Typical active materials for ECAs include metals, conductive polymers, carbons, metal oxides/sulfides, and their hybrids. Metals and carbons store ions mainly through forming electrochemical double layers on their outer and inner surfaces (non-Faradic process), while conductive polymers and metal oxides/sulfides through redox reactions (Faradic process). As metal oxides/sulfides generally have a low electrical conductivity, they are usually mixed with conductive materials to form active electrodes, which will be discussed in the subsection of hybrid electrodes.

2.2.1. Metal-Based Electrodes

Among various ECAs, ionic polymer–metal composite (IPMC)-based actuators have been widely investigated because of their fast ion migration induced bending mechanism and precise electrical control of actuation performance.[35] IPMC actuators are normally composed of polymer electrolyte membrane coated with stable nanostructured metals (Pt, Au, etc.) as electrodes via electroless plating/electroplating, hot pressing, or chemical/physical vapor deposition processes (Figure 2).[36,37]

However, IPMC actuators suffer from cycling degradation and short lifetime due to different Young’s modulus between polymer electrolyte membrane and metal electrodes, thus resulting in metal cracks during repetitive bending/actuation.[38] These irregular metal cracks would also be generated when the bending angles exceed their yield strain. These metal cracks would then peel off from polymer electrolyte membrane and inevitably lead to electrical disconnection, uneven electrical distribution, and device distortion of IPMC actuators under pulsed electrical field. Electromechanical force and response time of IPMC actuators would also be severely affected. Thus, extensive studies have been conducted to develop novel metal electrodes with enhanced stability and conductivity. Generally, it is effective to incorporate carbon materials and conductive polymers into metal electrodes to improve electrode flexibility and suppress/avoid electrode cracking during repetitive bending. For example, by electrografting the conductive polymer poly(ethylene oxide) (PEDOT) onto Pt nanoparticles, the prepared IPMC actuators could be operated with prolonged lifetime and enhanced electromechanical properties.[39] Engineering the polymer electrolyte membrane is another direction to improve the lifetime and electromechanical properties of IPMC actuators, which would be discussed in detail in Section 2.3.

2.2.2. Conductive Polymer Electrodes

Conductive polymer is another type of electromechanical active materials for ECAs, such as polyaniline (PANI), polypyrrole, and poly(3,4-ethylenedioxythiophene):poly(styrene sulfonate) (PEDOT:PSS).[40–43] It should be noted that volume change and actuation performance in the conductive polymer-based ECAs are dependent on the size/mobility of dopants and whether the conducting polymers are n- or p-doped.[44] For example, when p-doped conductive polymer is reduced and the dopant anions are mobile and small, the anions and solvent would move out of polymer chain to neutralize the charges, thus generating contraction deformation. If p-doped conductive polymer with large and immobile anions is reduced, small and mobile cations together with solvent would go into the polymer network to balance the charges, thus causing swelling deformation. For n-doped conductive polymer with small/mobile cation dopants, the small and mobile cations would move out of the polymer networks when it is oxidized, thus the conductive polymer would shrink. The solvent movement and ion migration would lead to mass transfer, volume change, expand/shrink deformations, and actuation performance in conductive polymers during redox reactions and polymerizations under pulse electric field. Owing to such rate-limiting ion diffusion/migration mechanism, the conductive polymer-based actuators exhibit slow response/actuation speed.[45] Another drawback needing to be resolved in the conductive polymer-based actuators is their inferior cycle life due to the structure/framework change/collapse during redox reactions of conductive polymers under pulse electric field.[46] Among the conductive polymers, PEDOT:PSS shows a good stability and processability and is frequently used as the flexible active electrodes,[43,46–48] which can be prepared by simply coating onto the electrolyte membrane and shows a good frequency response (Figure 3).[43] Synthesis of hybrid electrodes[49] is one effective approach to enhance the actuation speed and improve cycling life/stability in conductive polymer-based actuators, which will be presented in Section 2.2.4.

2.2.3. Carbon-Based Electrodes

Carbon materials especially nanocarbons are very promising active materials for ECAs due to their high electrical conductivity, good electrochemical stability, large specific surface area, high mechanical strength, and low cost, as compared with conductive polymers and noble metals. Nanocarbons can include 0D (e.g., fullerene, carbon dot, and carbon black), 1D (e.g., carbon nanotubes [CNTs], and carbon nanofibers [CNFs]), 2D (e.g., graphene and graphyne), and 3D porous nanocarbons (e.g. activated carbon and carbon aerogel) according to their morphology, all of which have been investigated as ECA electrodes. This section will mainly focus on 1D and 2D nanocarbons because they are mostly studied for ECAs due to their higher conductivity and strength/modulus and better processability than 0D nanocarbons and better flexibility than 3D nanocarbons.
1D Nanocarbons: CNTs\(^{33,50–56}\) and CNFs\(^{57–59}\) are typical 1D nanocarbons that have been investigated as active electrodes for ECAs. This section will focus on CNT-based electrodes due to their generally higher electrical conductivity and specific surface.

Figure 2. Metal electrode based ECAs (IPMCs). a) Preparation procedure of IPMC actuators and scanning electron microscope (SEM) image of the cross section of the electrolyte–electrode interphase and actuation mechanism in IPMC actuators. Reproduced with permission.\(^{[37]}\) Copyright 2017, American Chemical Society. b) Synthesis procedure of gold nanoparticles and structure design of gold-based IPMC actuators. Reproduced with permission.\(^{[36]}\) Copyright 2017, Wiley-VCH.
SWCNTs were widely studied as the active electrodes,\(^33,52,55\) and ECAs based on CNT electrodes can be enhanced. Therefore, conductivity and mechanical performance. By enlarging the specific surface area of the electrodes, the actuation performance of ECAs based on CNT electrodes can be enhanced. Therefore, SWCNTs were widely studied as the active electrodes,\(^13,32,52,55\) and SWCNT-based electrodes were shown to provide better actuation performance.\(^54\) Baughman et al. demonstrated the first SWCNT-based ECA by adhering two SWCNT paper strips on opposite sides of a polyvinyl chloride (PVC) strip, which bent reversely under a triangular voltage of \(\pm 1\) V in a 1 M NaCl solution due to the expansion difference of the two electrodes.\(^33\) In a three-electrode cell, a SWCNT strip electrode generated a lateral strain up to 0.2% at a low frequency and produced a higher stress (0.75 MPa) than the human muscle (0.3 MPa) under a 0.1 Hz square wave potential between \(-0.5\) and 1.5 V. They attributed the actuation to the dimension changes in the covalent bonded directions of SWCNT due to the quantum chemical and the electrostatic double-layer effects.\(^13\) Xie group synthesized strong, conductive, hierarchal SWCNT films with a strongly bonded network directly by chemical vapor deposition and used them to assemble a bendable ECA with a half-dried EMIBF\(_4\)/glycerol/chitosan membrane.\(^52\) The ETA showed a high tensile strength (50 MPa) and Young’s modulus (1–2 GPa) due to the strong SWCNT films. Interestingly, the actuation displacement showed a sharp peak at \(\approx 30\) Hz under \(\pm 4\) V, which was assigned to the resonance frequency, resulting a superfast response and a high strain (stress) generation rate of 0.9% s\(^{-1}\) (1080 MPa s\(^{-1}\)) due to the hierarchical structure, and high electrical and mechanical properties of the SWCNT film.\(^52\) However, the aggregation-induced low specific capacitance and high cost of SWCNTs limit their actuation performance and large-scale application.

The surface chemistry of CNTs can affect their affinity to the electrolyte and intertube interactions and therefore the actuation performance. Pristine CNTs tend to aggregate due to the strong \(\pi-\pi\) interaction and reduce the accessible surface for ion adsorption. Surface modification of CNTs by covalent or noncovalent functionalization was proven effective to enhance the actuation performance of CNT-based electrodes. Covalent functionalization includes the surface oxidation treatment and a possible further grafting/cross-linking with other functional groups.\(^56-63\) Carboxylated multiwalled carbon nanotube (MWCNT)/polyvinylidene fluoride (PVDF)/EMIBF\(_4\) gel electrodes showed significantly larger actuation strain differences and blocking stresses than the counterparts based on pristine MWCNT/PVDF/EMIBF\(_4\) gel electrodes due to the enhanced specific capacitances and Young’s modulus of the modified electrodes.\(^56,61\) Cross-linking of oxidized SWCNTs with phenylenediamine significantly increased the specific capacitance of a SWCNT/PVDF/EMIBF\(_4\) gel electrode, and the resulting ECA generated much higher strain differences under all frequencies lower than 100 Hz and showed much faster response.\(^61\) Noncovalent functionalization includes using organic/polymer dispersant/binder and solvation with ionic liquid, which is useful for producing free-standing flexible electrode with a good interface binding with the electrolyte membrane. Using Triton X-100 as surfactant, CNTs suspended well in water and the resulting CNT film electrode fused well with an ionic liquid/Nafion electrolyte membrane to produce a bendable ECA with a high modulus.\(^64\) Disperse MWCNTs with chitosan to obtain homogeneous, porous film electrodes attained good compatibility with the EMIBF\(_4\)/chitosan electrolyte membrane.\(^65\) The assembled ECA showed an actuation speed of 2 mm s\(^{-1}\) under a square wave voltage of \(\pm 3\) V of 0.5 Hz. However, the use of insulating polymer binder may decrease the electrical conductivity and high-frequency response.

Solvating CNTs by the liquid electrolyte to produce a bucky gel electrode is a very successful strategy to improve both the actuation strain and speed of CNT electrodes. Ionic liquids are good...
choices to produce bucky gel electrodes for ECAs because they have a strong interaction with CNTs apart from a high ionic conductivity and low volatility.\[53–56,66\] SWCNT/EMIBF$_4$/PVDFHFP (13:54:33 in weight) gel electrodes with exfoliated, finer bundles of SWCNT led to a doubled specific capacitance as compared with the pristine ones without ionic liquid.\[56\] The resulting bendable ECAs generated a strain difference of 0.9% and a stress of 0.1 MPa under a 0.01 Hz square wave voltage of ±3.5 V but showed a fast decrease in the displacement under 1–10 Hz due to the low content of SWCNTs.\[56\] With millimeter-long SWCNTs and increased loading ratio of 50 wt\%$, a freestanding EMITFSI/SWCNT gel electrode without any polymer showed significantly higher tensile strength, Young’s modulus, and electrical conductivity than the counterpart based on short SWCNTs (Figure 4a–d).\[54\] A bendable ECA based on those electrodes showed sustainable displacement when the frequency was increased from 1 to 10 Hz due to the highly conductive porous network structure.\[54\] However, the gelation of CNTs with ionic liquid would obviously decrease the Young’s modulus and electrical conductivity and lead to a reduced actuation force/stress and increased inner resistance.

The orientation of CNTs in the CNT film/fiber electrodes has a critical effect on the actuation performance, which can be controlled by the processing method because the ion insertion into a CNT bundle can induce a much higher expansion perpendicular to the bundle than along it. A much higher lateral expansion (12.1\%) of the vertically aligned CNTs (VACNT)/Nafion film than the neat Nafion film (8.3\%) was attained after absorption of an ionic liquid.\[67\] A bendable ECA based on the VACNT/Nafion electrode produced an ultrahigh strain difference of 8.2\% under a square wave voltage of ±4 V and 0.05 Hz. Over 80\% of the maximum strain can be achieved at 2 s and a high blocking stress of 4.7 MPa was generated under 4 V. The ultrafast response and high actuation strain were attributed to the vertically aligned structure, which provided very short pathways for ion and electron conduction and enhanced the lateral expansion induced by ion insertion.\[67\] However, the loading of CNT is relatively low as compared with traditional electrodes and the fabrication process is hard to be scaled up. The maximum strain and stress are expected to be improved by increasing the volume fraction of CNTs to over 30\%, which is challenging. A MWCNT yarn twist-spun from MWCNT forests can rotate −590 reversions min$^{-1}$ in a TBAPF$_6$/acetonitrile solution under a 5 V pulse voltage and showed a high contraction strain of ≈1% under a voltage of −2 V due to the large volume expansion induced by electrochemical double-layer charge injection into the bias-aligned CNTs.\[51\]

2D Nanocarbons: Typical 2D nanocarbons useful for ECAs include graphene,\[68–73\] graphyne (graphdyene),\[74\] and biomass/polymer-derived carbon nanosheets.\[75,76\] This part will focus on graphene-based active electrodes because graphene has a large specific surface area, high electrical conductivity, excellent mechanical strength and flexibility, good chemical stability, and potentially low cost. Macroscopic hydrogels/foams\[68–71,77\] and films\[68–71,78\] assembled from graphene sheets have been studied as electrodes for either extensible or bendable ECAs. Similar to CNT-based electrodes, the actuation performance of graphene-based electrodes can be affected by the size,
number of layers, defects, alignment, and surface chemistry of the graphene sheets, the porous structure, modulus, conductivity, and surface symmetry of the electrode, as well as the composition of the electrolyte. The easy stacking of graphene sheets in the electrodes is an obvious issue limiting the actuation performance because it will lead to reduced surface area for ion insertion/adsorption and lengthened pathways for ion diffusion. Therefore, the actuation performance can be enhanced by tuning the surface chemistry and defects of graphene sheets, solvating graphene with a solvent or electrolyte, controlling the in-plane porosity and alignment direction of graphene sheets, as well as incorporation of guest fillers (will be discussed in Section 2.2.4).

The surface chemistry and defects of graphene sheets can be tuned by controlling the reduction degree (for reduced graphene oxide),[76] heteroatom doping,[79,80] chemical activation,[81] as well as plasma treatment and laser scribing.[68–70] Graphene films/foams are generally prepared by chemical/thermal reduction of graphene oxide (GO) films/solutions, and the residual O-containing groups can affect their electrical conductivity, specific capacitance, Young’s modulus, and affinity to the electrolytes. O content in reduced GO (RGO) hydrogel films can be tuned by varying the weight ratio of hydrazine reductant to GO (RGO) to improve the hydrophilicity and achieve the highest specific capacitance with lowest stiffness, which resulted in a highest strain of 0.2% under −1 V and 0.01 Hz in 1 M NaCl solution.[78] Doping graphene with other atoms such as N and S could create more electrochemically active sites, enlarge the interlayer spacing, and improve the affinity to the electrolyte; therefore, the specific capacitance and actuation performance can be improved. A bendable ECA based on N-doped RGO electrodes showed much higher specific capacitance and generated larger displacements and quicker responses under a sinusoidal ±2 V voltage of 0.01–5 Hz, as compared with the counterpart based on pristine RGO electrodes because of the high doping level (8.72 at%) and the wrinkled morphology of the doped RGO.[79] Generally, RGO films showed a poor hydrophilicity, low electrochemical activity in aqueous electrolytes, and poor bonding with polymer electrolyte membrane, which can be improved by O2-plasma or laser scribing treatment.[68–70] O2-plasma-treated RGO films exhibited higher hydrophilicity and affinity to 1 M NaClO4 electrolyte and lower charge transfer resistance,[70] thus generating significantly higher strains (≥0.85%/0.35%/0.2% under a square wave potential of ±1.2 V of 0.005/0.05/0.1 Hz, respectively) as compared with the original one. An asymmetric RGO film with one surface treated by laser scribing was used to assemble bendable ECAs with improved interface adhesion between electrode/electrolyte membrane and slightly increased specific capacitance due to the generated rough surface with a higher specific surface area (Figure 4e–f).[68] Meanwhile, the pristine surface was hydrophobic and impermeable to water and ionic liquid. As a result, the assembled ECA with an EMIBF4/Nafion membrane generated a large displacement under a 0.1 Hz sinusoidal voltage of ±5 V and showed a much better stability for 6 h than conventional Pt-based IMPC ECAs. However, the severe stacking of RGO sheets limits the charge storage capacity and thus the actuation strain and stress, which could be solved by solvating graphene or hybridizing with nanofillers.

Solvating graphene sheets with a solvent or electrolyte (e.g., water or ionic liquid) can effectively prevent their stacking and provide many ion diffusion channels and charge storage sites, thus increasing their actuation strain and response speed. RGO hydrogel paper (hydrated RGO) can be directly used after wet filtration without drying, showing much higher specific capacitance and generated a much higher lateral strain than the dried RGO paper (0.1% vs 0.037% at −1 V and 0.01 Hz in 1 M NaCl aqueous solution) because of the much larger accessible surface in the former.[78] An obviously enlarged average distance between the solvated graphene sheets can be achieved in BMImBF4/RGO (1:1 in weight) films by casting their mixture in NMP and subsequent drying, as confirmed by X-ray diffraction (XRD) and transmission electron microscope (TEM) results.[73] As a result, the solvated film generated a peak-to-peak out-of-plane strain of 38% under a −0.005 Hz square wave potential of ±2 V, in contrast to no observable strain for the neat dried RGO film due to the tight stacking between RGO sheets. When the content of BMImBF4 was increased to 66.7%, a higher strain up to 98% was generated but showed a low cycling stability due to the reduced interaction between RGO sheets.[73] However, the high content of ionic liquid (over 50%) clearly decreases the electrical conductivity, and the usable actuation displacement and forces are limited to the small thickness of the RGO film.

Considering the anisotropic properties (e.g., different electron conduction, ion diffusion, and actuation strain along different direction) of stacked graphene sheets, the actuation performance of graphene foams/films can be greatly enhanced by aligning the graphene sheets perpendicular to the target direction, increasing the packing density of the electrodes, as well as reducing the geometric size of the electrodes. Compressed graphene hydrogel pillar with a height reduction of 85% generated a significantly higher out-of-plane strain (≥1.3%) than the pristine one (≥0.05%) under a 0.02 Hz square wave potential of ±0.8 V in a 1 M NaClO4 solution[72] because the packing density and the alignment of graphene sheets along the in-plane direction were greatly increased after deep compression. However, the actuation strain is still low because of the porous structure. Chen’s group claimed that the actuation performance of a graphene film electrode could be enhanced by applying the electrical field along the in-plane direction, which could guide the ion diffusion along the channels between graphene sheets and was realized by coating the silver paste on its edge rather than on the surface for applying the voltage.[73] Through parallel microchannels made by filtration of a graphene/PTEF suspension onto a patterned PMMA template,[81] the patterned graphene/PTEF cathode electrode showed much higher out-of-plane expansion strains of 77.5% and 19.1% than the nonchannelled counterpart (46.3% and 16.6%) under a triangle voltage of 0–4 V at 50 and 500 mV s−1, respectively, due to the instant ion diffusion into the electrodes from the electrolyte in the microchannels.

2.2.4. Hybrid Electrodes

All the aforementioned active materials have their advantages and shortcomings. Metal-based electrodes have a high electrical conductivity, but suffer from ease of cracking during cyclic bending, leading to a decreased conductivity, electrolyte leakage, and
therefore a low durability. Conductive polymers show a high redox activity coupled with ion doping/dedoping, but the electrical conductivity generally decreases when they are reduced (dedoped) and the cycling stability is usually poor due to degradation. Metal oxides and sulfides also have a very high pseudo-capacitance but show a low conductivity and mechanical strength. Nanocarbons (especially 1D and 2D type) have a high electrical conductivity, specific surface area, and mechanical strength but tend to aggregate/stack together, which decreases the charge storage capacity and actuation performance. Combining two or three kinds of those active materials together into a hybrid electrode is proven a successful strategy to improve the actuation strain, stress, and speed of the electrodes and ECAs due to the synergistic effect. Among these, 1D/2D nanocarbons are mostly used to hybrid with other active materials due to their superior electrical and mechanical properties.

Hybridizing metal particles with 1D/2D nanocarbons (e.g., CNTs and graphene sheets) can reduce the cracking of metal electrodes and also reduce the stacking of nanocarbons, leading to an increased strength and charge storage capacity of the electrode and thus enhanced actuation performance of the actuators. Tamagawa et al. fabricated an ECA based on silver/MWCNT hybrid coated Naion (IPMC) membrane, which showed a higher displacement and better controllability (less drifting of the displacement) than the counterpart based on silver-coated Naion under a rectangular voltage of 2 V. The reason is that MWCNTs deposited on the Naion promoted the deposition of more silver particles and enhanced the stiffness of the actuator, which reduced the unrecoverable deformation. MWCNT coating on the Pd–Pt electrode can fill the cracks of the metal layer, prevent the evaporation/leakage of water in the electrolyte, and reduce possible oxidation of Pt nanoparticles. As a result, the ECA based on MWCNT/(Pd–Pt) hybrid electrodes generated a more stable displacement and blocking force and much longer air operation time than the one based on Pd–Pt electrode under 2 or 3 V. Chen’s group obtained a homogenous silver nanoparticle/reduced graphene oxide (Ag/RGO) hybrid film, which showed a significantly higher electrical conductivity than the RGO film. The ECA based on the Ag/RGO electrodes and an ionic liquid/polymer electrolyte exhibited much higher strains and strain rates than the counterpart based on RGO film electrodes due to reduced stacking of RGO sheets and higher conductivity. Interestingly, it also showed obviously higher displacement and cycling stability than the ECA based on Ag nanoparticle electrodes because graphene provided a large surface for ion storage and effectively prevented the oxidation of Ag nanoparticles. However, the actuation strain difference is still low (<0.036% at ±0.5 V of 0.1 Hz) as compared with neat graphene-based actuators (>0.12% at ±2.5 V of 0.1 Hz) due to the low applied potential limited by Ag.

Conductive polymer/nanocarbon (1D/2D) hybrid electrodes can make use of the high specific capacitance and binding ability (e.g., PEDOT:PSS) of the conductive polymers as well as the high conductivity and modulus of the nanocarbons and could also reduce the stacking of nanocarbons if the polymer was synthesized/coated on their surfaces. Therefore, those hybrid electrodes generally showed enhanced actuation performance as compared with the electrodes based on only nanocarbon or conductive polymer. PEDOT:PSS/SWCNT/EMIBF4 hybrid gel electrode showed much higher Young’s modulus, electrical conductivity, and specific capacitance than the PEDOT:PSS/EMIBF4 gel electrode, due to the high conductivity and strength of SWCNTs and a possible synergistic effect between SWCNT and redox active PEDOT:PSS (Figure 5a–c). Consequently, the ECA based on hybrid gel electrodes generated significantly higher strain differences (up to 0.57%) and stresses (up to 1.45 MPa) than the counterpart based on PEDOT:PSS/EMIF4 gel electrodes at 0.005–5 Hz under a triangular voltage of ±2 V. A PANi/RGO (6/4 in weight) hybrid film, which was formed by growing PANI nanoparticles on RGO homogeneously, showed an obviously higher specific surface area and capacitance than neat RGO film due to the synergistic effect. As a result, a bendable ECA based on the hybrid electrodes produced a significantly higher strain difference (0.327% under a ±0.5 V square wave voltage of 0.01 Hz) than the ECAs based on either RGO or PANI electrodes. The displacement retention was also improved a lot as compared with the one based on PANI electrodes after 200 actuation cycles. As the PANI/RGO hybrid electrode showed a lower modulus and strength than the neat RGO film, this may have affected the actuation stress. A bendable ECA based on PEDOT:PSS/doped RGO hybrid gel electrodes also showed much higher strain differences (0.36% under a 0.1 Hz sinusoidal voltage of ±1 V) as compared with the counterpart based on neat PEDOT:PSS electrodes due to the enhanced conductivity and specific capacitance by the introduction of doped RGO sheets, which had a strong interface interaction with PEDOT:PSS.

Metal oxide (sulfide)/nanocarbon hybrid electrodes can increase the charge storage capacity of metal oxides (sulfides) by instant electron/hole injection from the conductive nanocarbon network and simultaneous ion storage on their surfaces. It is critical to achieve a homogeneous distribution of metal oxide (sulfide) within the carbon network. Terasawa et al. demonstrated that a RuO2/MWCNT hybrid gel electrode (with EMIBF4 and PVDFHFP) showed higher specific capacitance than neat gel electrode based on only RuO2 or MWCNT because the conductive MWCNTs network can transport electrons/holes effectively to the pseudocapacitive RuO2 particles for storing much more ions. Therefore, the bendable ECA based on RuO2/MWCNT hybrid gel electrodes generated much higher strain differences at all frequencies below 10 Hz (up to ≈0.4% at 0.05 Hz under ±2 V) and higher stress (≈0.4 MPa) than the counterparts based on neat gel electrodes. The high cost of RuO2 may limit their large-scale application. They also observed similar results for a MnO2/MWCNT hybrid gel based ECAs, which showed superior strain differences under all frequencies below 5 Hz (<0.6% at 0.1 Hz under ±2 V) to the counterparts based on only MnO2 or MWCNT gel electrodes. The addition of semiconducting MoS2 or WS2 microparticles in the SWCNT gel electrode (with BMIBF4 and PVDF) significantly increases the charge storage capacity of the assembled bendable ECAs, therefore obviously improved strain differences (up to 0.73% and 0.76% at 0.025 Hz and ±2 V for MoS2 and WS2, respectively) and stresses as compared with the counterpart based on only SWCNT gel electrodes. However, the low modulus and strength of those hybrid gel electrodes with ionic liquid would lead to low actuation stresses/forces. Huang et al. showed that Fe3O4/RGO hybrid films showed a more porous structure and
higher gravimetric capacitance and thus a larger in-plane actuation strain than neat RGO films due to reduced stacking of RGO sheets by Fe3O4 nanoparticles.[90] A bendable ECA based on the ternary hybrid electrodes of flower-like porous MoS2/RGO hybrid and PEDOT:PSS (Figure 5d–f) generated much higher strain differences and larger forces as compared with the counterpart based on the binary RGO/PEDOT:PSS hybrid electrodes because the active MoS2 and porous RGO can provide more ion storage capacity.[89]

2.3. Electrolyte Design

The electrolyte membrane plays a significant role in the actuation performance of ECAs. Their potential window, ion size, ionic conductivity, and modulus can affect the ion diffusion kinetics and charge storage capacity of the electrodes, therefore the actuation strain, stress, and speed of ECAs. The electrolyte membrane can be divided into two classes according to the polymers used: ionic polymer-based electrolytes and nonionic polymer-based electrolytes. Ionic polymers contain functional groups (e.g., —SO3H, —SO3Li, —COOH, —COONa, —NH3Cl), which can dissociate into mobile ions and fixed counter ions bound to the backbone when contact with water or polar organic liquids. When a voltage is applied across the ionic polymer electrolyte in the actuator, the mobile cations/anions would migrate and accumulate onto/inside the negative/positive electrode, thus inducing a volume difference and bending actuation of the device. Typical ionic polymers used in ECAs are fluorinated polymers, e.g., Nafion[92] and Flemion.[34,92] Hydrated Nafion has a hydrophobic backbone and an interconnected network of hydrophilic ionic phase that can serve as nanochannels for cation migration,[93] and was directly used as the electrolyte membrane in the first IPMC actuator.[94] The bending displacement can be enhanced by exchanging protons in Nafion with large sized cations such as hydrated Li+, TMA+, and TBA+. However, Nafion membrane has a narrow potential window, a poor ionic conductivity in dry air, back relaxation under DC voltage, and a high cost, which limit their actuation performance and large-scale application. In addition, the small diameters (<4 nm) of the inner channels restrict ion migration, also limiting the actuation performance.[95,96]

Those issues of Nafion can be avoided by addition of liquids of low volatility and nano fillers and using alternative ionic polymers. To enable the dry operation of Nafion-based IPMC in air, ethylene glycol, glycerol, or ionic liquid is generally added for absorbing water from air.[38,97–100] Addition of ionic liquid can also increase the ion conductivity, provide more mobile cations, enlarge the electrochemical potential window, and thus enhance the actuation displacement.[97] Various nanofillers such as SiO2,[101,102] silicates,[96,103] GO,[104,105] RGO,[106,107] and CNTs[108–110] were also added into Nafion at appropriate loadings, which can create more inner surfaces/interfaces for ion diffusion and generally improve the ionic conductivity and modulus of Nafion membrane and therefore the actuation strain and stress of the corresponding ECAs. Note that too much conductive RGO or CNT may increase the electrical conductivity of the electrolyte membrane and lead to a large leakage current and short retention period of the actuated state. Other ionic polymers, such as poly(2-acrylamido-2-methyl-1-propanesulfonic acid) (PAMPS),[111] fluorinated polymer grafted with acrylic acid or

Figure 5. Hybrid electrode based ECAs. a–c) PEDOT-PSS/SWCNT/IL gel electrode based ECAs: a) schematic of the configuration and fabrication process; b) structure of PEDOT-PSS and the IL; c) frequency dependence of the actuation strain of ECAs based on different electrodes. Reproduced with permission.[49] Copyright 2016, American Chemical Society. d–f) MoS2/doped RGO hybrid electrode based ECA: d) SEM image of the MoS2/doped RGO hybrid; e) frequency dependence of the actuation strain of the ECA based on the hybrid electrode; f) blocking force of the ECAs based on hybrid electrodes and neat RGO electrodes under different voltages. Reproduced with permission.[89] Copyright 2019, Wiley-VCH.
organic ammonium salt\textsuperscript{[112]} and carboxylated cellulose\textsuperscript{[47,113]} and chitosan-acid\textsuperscript{[52,114]} are potential low-cost alternates to Nafion, but the creation of highly ionic conducting network in those polymer membranes is necessary for providing high actuation response. For example, Dai et al. showed that PAMPS/PVA membrane can be cross-linked by heating at 100 °C and the resulting film showed a high ionic conductivity and water uptake.\textsuperscript{[111]} Park et al. obtained a polyacrylic acid grafted PVDFFFHFP film as the anionic electrolyte membrane by radiation-assisted grafting and showed that the IPMC based on it produced comparable displacements with the Na\textsuperscript+ radiation-assisted grafting and showed that the IPMC based on it produced comparable displacements with the Na\textsuperscript+ radiations of the polymers and electrolytes, there are many choices for the cathode and anode, respectively. Because of the various combinations of the polymers and electrolytes, there are many choices with different ionic size, ionic conductivity, Young's modulus, and binding capability. Hydrophilic polymer/aqueous electrolyte membranes, such as PVA/H\textsubscript{2}SO\textsubscript{4} gel electrolyte,\textsuperscript{[115]} can have a high ionic conductivity and enable a high charge storage capacity for the active electrodes, but have a low potential window and a small size difference between anions and cations. Therefore, nonvolatile ionic liquids are preferred to combine with nonionic polymers to work as gel electrolyte membranes, which can provide a larger potential window without a drying issue. The viscosity and ionic strength of the liquid and the Young's modulus and the ionic diffusivity network of the polymer membrane have a strong influence on the actuation performance. Generally used nonionic polymers for ionic liquids include fluorinated polymers (e.g., PVDF\textsuperscript{[74]} PVDFFFHFP\textsuperscript{[56]}, PEO\textsuperscript{[116]} nitrile butadiene rubber (NBR),\textsuperscript{[46]} and cellulose.\textsuperscript{[117,118]} It is critical to construct a continuous ion conducting network in those nonionic polymers, which can be achieved by strategies such as inducing a phase inversion of the polymer solution and constructing an interpenetrating polymer network (IPN). Ionic liquid/PVDF membrane prepared by solution casting is widely used because the weak miscibility between them can induce a

| Table 1. Summary of the structures and performances of some typical soft ECAs. |
|-----------------------------------------------|
| Active electrodes | Electrolyte membrane | Device size [mm\textsuperscript{2}] | Device modulus [MPa] | Applied voltage [V] | Peak strain difference [%] | Actuation force/stress | Energy density [kJ m\textsuperscript{3}] | Energy efficiency [%] | Ref. |
|-------------------|----------------------|-----------------|---------------------|------------------|------------------------|-----------------|---------------------|-------------------|-------|
| Deposited Pt layer | Stacked Na\textsuperscript{+} | 50 \times 5 \times 0.95 | – | 0-3 (step) | \approx 4.6\textsuperscript{a} (0.1 Hz) | 144 mN (DC 4 V) | – | – | [37] |
| Deposited Au layer | EMIBF\textsubscript{4}/HNC/ PAA-co-PAN\textsuperscript{a} | 25 \times 2.5 \times 0.35 | 0.35 | ±5 | 1.04 (0.02 Hz) | 0.01 mN (0.02 Hz) | – | – | [36] |
| PEDOT/PSS coating | BMIBF\textsubscript{4}/graphene/ cellulose acetate | 40 \times 10 \times 0.15 | 20.34 | ±3 (sinusoidal) | 0.15 (0.1 Hz) | 0.441 mN (DC 3 V) | – | – | [43] |
| PEDOT | EMITFSI/PEO/NBR | 18 \times 11 \times 0.25 | 43 | ±2 (square)\textsuperscript{a} | 2.4 (0.05 Hz) | \approx 33 mN | – | – | [121] |
| Chitosan/MWCNT | EMIBF\textsubscript{4}/glycerol/ chitosan | 30 \times 5 \times 0.17 | – | ±3 (square) | \approx 0.15 (0.5 Hz) | – | – | [65] |
| SWCNT/BMIBF\textsubscript{4}/ PVDFHFP | BMIBF\textsubscript{4}/PVDFHFP | 15 \times 1 \times 0.28 | – | ±3.5 (square) | 0.9 (0.01 Hz) | 0.1 MPa (0.01 Hz) | – | – | [56] |
| SWCNT/EMITFSI/ PVDFHFP | EMITFSI/PEO/NBR | 15 \times 1 \times \approx 0.06 | – | ±2.5 (square) | \approx 0.228 (10 Hz) | \approx 0.362 MPa (10 Hz) | – | – | [54] |
| Au-coated VACNT/ Nafion/ | Nafion | 5 \times 0.8 \times 0.049 | – | ±4 (square) | 8.2 (0.05 Hz) | 0.3 mN/4.7 MPa (DC 4 V) | – | – | [67] |
| MWNT/PEDOT: PSS | EMIBF\textsubscript{4}/TPU | 20 \times 5 \times 0.11 | 82.2 | ±2.5 (harmonic) | 0.64/0.47 (0.1/1 Hz) | 1.43 mN (DC 2.5 V) | – | \approx 0.08 (0.1 Hz) | [355] |
| SWCNT/EMIBF\textsubscript{4}/ PEDOT-PSS | EMIBF\textsubscript{4}/PVDFHFP | 10 \times 1 \times 0.15– 0.175 | – | ±2 (triangular) | ±0.2 (0.1 Hz) | 1.45 MPa (5 mHz) | – | – | [49] |
| MWNT/MnO\textsubscript{2}/ EMIBF\textsubscript{4}/PVDFhFP | EMIBF\textsubscript{4}/PVDFhFP | 10 \times 1 \times 0.15– 0.175 | – | ±2 (triangular) | ±0.6% (0.1 Hz) | ±1.05 MPa (5 mHz) | – | – | [83] |
| Graphene/PVDF | EMIBF\textsubscript{4}/PVDF | 25 \times 2.5 \times 0.08 | – | ±2.5 | 0.12% (0.1 Hz) | 1.92 mN/2.41 MPa | \approx 0.62 (0.1 Hz) | ±0.1 | [74] |
| Graphidyne/PVDF | EMIBF\textsubscript{4}/PVDF | 25 \times 2.5 \times 0.08 | 420 | ±2.5 | 0.78% (0.1 Hz) | 3.37 mN/3.11 MPa (0.1 Hz) | 11.5 (0.1 Hz) | 6.03 (0.1 Hz) | [74] |
| PANi/RGO | H\textsubscript{2}SO\textsubscript{4}/PVA | 25 \times 2 \times 0.25 | – | ±0.5 (square) | 0.327/0.032 (0.01/0.1 Hz) | 30 MPa (0.01 Hz) | – | – | [75] |
| N,S-doped RGO/ PEDOT-PSS | EMIBF\textsubscript{4}/SPB\textsuperscript{a} | – | – | ±1 (sinusoidal) | 0.36/0.037 (0.1/1 Hz) | 1.25 mN (DC 2 V) | – | – | [80] |

\textsuperscript{a}Estimated by us; \textsuperscript{b}HNC, halloysite nanoclay; PAA-co-PAN, poly(acrylic acid) and polyacrylonitrile copolymer; \textsuperscript{c}Square wave potential; \textsuperscript{d}Sulfonated polybenzimidazole.
phase inversion of the PVDF solution into a porous network during drying of the solvent. Hierarchical porous PVP/PVDF membrane with a 3D interconnected channels is found to be beneficial to increased loading of both aqueous and ionic liquid electrolytes and enhanced actuation performance of the corresponding ECAs (Figure 6a). An ionic liquid/cellulose electrolyte membrane can be obtained by a water-induced phase inversion method, and generated a highly porous structure for loading a high amount of ionic liquid (Figure 6b). However, the content of cellulose is limited and may lead to a low modulus of the electrolyte membrane and a small actuation stress of the final actuator. Festin et al. showed that a PEO/SBR IPN membrane obtained by cross-linking had improved mechanical properties and a comparable ionic conductivity to the PEO membrane after swollen by a large amount of ionic liquid. The resulting bendable ECA based on PEDOT-coated ionic liquid/IPN membrane showed a very high strain difference of 2.4% and a blocking force of ≈33 mN under a 0.05 Hz square voltage of 2V.

2.4. Potential Application for iHMI

As soft ECAs can generate deformations and forces under a low voltage, they have potential applications in soft robotic fingers/gippers/hands for executing various actions on force-sensitive targets, as well as tactile/haptic displays (e.g., braille devices) and morphing devices (e.g., humanoid robot face) for information communication. For example, Shahinpoor showed that IPMC actuators can be used to grasp and dexterously manipulate bodily organs and tissues. ECA based on MoS₂/graphene electrodes can work as robotic fingers to interact safely with fragile surfaces of smartphone and tablets (Figure 7a). Another application of ECAs is in braille displays for vision-impaired people. A lightweight thin braille display prototype was developed using bucky gel-based ECAs to drive the pop-up of braille dots over 300 μm at an operation voltage of 4 V (Figure 7b). The output force is expected to be improved for a better user experience.

Interestingly, ECAs especially IPMC actuators are promising for flexible tactile sensors for human–robot/machine interaction. The external force applied on the IPMC can induce a deformation (e.g., bending) that forces the ions to move across the thickness direction and generate a small voltage signal. The output voltage is sensitive to the external mechanical stimuli and can be calibrated and correlated to the applied forces/stresses. The unique characteristics of softness and passive sensing render ECAs as promising haptic feedback/tactile sensors for robotic diagnosis/surgery and wearable monitoring systems for human activities. A tactile sensor with three degrees of freedom was developed using four IPMC sensors, which can detect both the direction and velocity of the motion of the center tip. By combining two IPMC strips (one as sensor and the other as actuator) into a multifunctional tactile sensor, detection of the contact of a target and measurement of its stiffness showed promises in biomedical applications such as catheterism and laparoscopy. A vibrating probe based on IPMC strips was further developed to recognize different materials it contacted that change the resonance frequency of the probe. IMPC strips/loops placed on surgical robotic end-effectors can be used to interact with bodily tissues/organs and provide voltage signals due to the deformation of IPMC, which can be correlated with the kinesthetic organ/tissue forces and used to provide surgeons with haptic/tactile feedback during surgery (Figure 7c). IPMC sensors fabricated by uniformly depositing gold on a Nafion membrane showed a high sensitivity and high cyclic bending stability, and were integrated with a circuit into a smart glove for detecting the subtle pulse of a human hand, reading precise braille, and controlling a robot hand timely by the wearer. However, the sensing signals of IPMC sensors are generally low (of several millivolts) and need to be improved for more accurate and sensitive applications.
3. Soft ETAs

Soft ETAs can produce deformations (strains) and/or forces (stresses) under an input electrical signal due to the temperature increment (heat generation) caused by the electrothermal effect. As compared with DEAs, ETAs can work under much lower voltages, generally ranging from several to tens of volts, which simplifies the power source (e.g., one or several batteries) and provides a safer environment for various applications. In contrast to low-voltage soft ECAs that require electrochemically active materials and liquid/gel electrolytes and work within a narrow temperature range, soft ETAs can be fabricated from ordinary solid materials, work in air under a much wider temperature range (e.g., \(-200\) to \(200\) °C, depending on the materials used), and produce relatively higher strains/stresses for longer cycles. In addition, the structure and fabrication process of soft ETAs are also relatively simpler. Therefore, soft ECAs are promising for many applications that require a low driving voltage, small footprint, and high reliability under various conditions.

3.1. Working Principle and Device Configuration

The actuation principle of ETAs is based on the thermal expansion/shrinkage of their components due to the electrical heating (Joule heating) of the conductive component under an input voltage/current (either DC or AC). Detailed mechanisms vary in device configurations and choices of materials. ETAs have many configuration designs for generating bending, linear, torsional, and even 3D actuators for a variety of applications. Commonly adopted basic configurations for ETAs (Figure 8) include cantilever beams (e.g., U-shaped hot-and-cold-arm actuators\(^{134-136}\) and bimorph actuators\(^{137-139}\), doubly clamped beams (e.g., straight\(^{140,141}\) and V-shaped [chevron] beams\(^{134,142,143}\)), and drum/balloon-like structures (e.g., inflation actuators\(^{144-146}\)). The beam is usually a strip, fiber/yarn, spring, or a large film/textile, and the skin of the drum/balloon is an elastic film. ETAs use either a homogenous material or a set of different materials for the components, which can be denoted as homogeneous ETAs or heterogeneous ETAs, respectively. Some designs, shown in Figure 8c-e, are usually based on the thermal expansion/shrinkage of a homogenous material or the inflation by the heated sealed gas/liquid\(^{140,141,146}\), while others, shown in Figure 8a,b, are based on the thermal expansion difference between different parts/materials connected/adhered together\(^{138,139}\). More complicated configurations can be designed by combining two or more basic ones, such as cascade V-shaped structures\(^{144,145}\). Among those, using a tortuous beam with an increased total length such as a spring (coil)/snake/Z-shaped structure can greatly amplify the nominal actuation strain as compared with a straight beam\(^{148,149}\).

3.2. Materials Selection

Apart from the configuration design, the actuation performance (strain, stress, speed, work capacity, power density, durability, and actuation direction) of soft ETAs is greatly reliant on the choice of materials with different electrical, mechanical, and thermal properties. According to the working principle, enhancing the expansivity of a homogenous material or the expansivity difference between different parts/layers is the key to enhance their actuation performance. The expansivity (\(\Delta L/L\) or strain \(\epsilon\), along the length direction) of a component in an ETA is generally the product of the temperature (\(\Delta T\) increment and the
thermal expansion coefficient (TEC, $\alpha$) along the length direction) of the material used according to Equation (1)

$$\varepsilon_1 = \Delta L / L = \alpha_1 \Delta T$$

Therefore, a large/fast temperature increment is critical for all types of ETAs to achieve a large/fast actuation. As the temperature increment results from Joule heating of the electrical conductive component (heating layer/circuit), choosing an efficient heating layer is very important. The consideration for the TEC of different components is also critical and depends on the configuration of ETAs. Furthermore, the actuation strain/displacement will be affected by the Young’s modulus of the components. Generally, a material with a lower modulus will have a higher TEC and favor the generation of a higher strain/displacement and faster response. As the Young’s modulus is usually inversely related to the square of the materials bends out of plane due to the expansion difference between the layers. Reproduced under the terms and conditions of the CC-BY license.© 2020, Mary Ann Liebert.

d) A V-shaped (chevron) beam, in which the shuttle moves linearly due to the thermal expansion/shrinkage of the arms. Reproduced under the terms and conditions of the CC-BY license.© 2019, The Authors, published by MDPI. e) A drum-like structure inflates due to the thermal inflation of the air inside heated by the skin. Reproduced with permission.© 2014, Royal Society of Chemistry.

$$\Delta Q = V^2 \sigma A_L t / L = mC \Delta T = \rho A_L L C \Delta T$$

$$\Delta T / t = (V / L)^2 \sigma / (\rho C)$$

$$\varepsilon_1 / t = (V / L)^2 \alpha_1 \sigma / (\rho C)$$

where $\Delta Q$, $\Delta T$, $V$, and $t$ are the electrical generated heat, temperature increment, applied voltage, and time; $A_L$, $L$, $m$, $\sigma$, $C$, and $\rho$ are the cross-sectional area, length, mass, electrical conductivity, specific heat capacity, and density of the beam, respectively. It can be seen that the heating and strain rates are determined by a material quality factor, $\sigma / (\rho C)$, and $\alpha_1 \sigma / (\rho C)$, respectively, for a fixed voltage and length of the heating layer. Therefore, using a beam with a high specific conductivity, a low specific capacity, and a very positive/negative TEC can increase the heating rate and reduce the required voltage. Considering the heat loss through thermal irradiation and convection in air, reducing the surface emissivity and surface area of the beam is helpful to reduce the heat loss and therefore the required voltage. However, this may decrease the recovery speed and limit the operation under high frequency.

Highly conductive metals such as gold, silver, and copper ($\sigma > 1 \times 10^5 \text{ S cm}^{-1}$) with a low emissivity are generally utilized in homogeneous ETAs for microelectromechanical systems. However, the low TEC ($1-2 \times 10^{-3} \text{ K}^{-1}$) and flexibility of those metals limit the actuation strain/displacement.
under low voltages and durability, and their high density and cost and weak thermal stability (for copper and silver) also limit their wide applications. Conductive carbon (e.g., CNT and graphene) films and fibers with smaller density, higher flexibility and thermal stability, and potentially lower cost are investigated for macroscale homogeneous ETAs.\textsuperscript{144,154} However, both CNT and graphene have a smaller TEC ($0.75 \times 10^{-3}$ K$^{-1}$ for the lattice constant of SWCNT and $-0.8 \times 10^{-3}$ K$^{-1}$ for a single layer graphene\textsuperscript{155,156}) and much lower conductivities in the form of macroscale films/fibers (usually $<1 \times 10^{3}$ S cm$^{-1}$), limiting their actuation strain/displacement. Therefore, semicrystalline/elastomer polymer or paraffin with a significantly higher TEC (usually $>5 \times 10^{-3}$ K$^{-1}$) was incorporated into carbon films/fibers to fabricate composite-based homogeneous ETAs,\textsuperscript{140,141,157–159} which showed improved strains due to the high expansion/shrinkage of the fillers, although the electrical conductivity may be decreased. In addition, conductive polymers are also investigated in soft homogenous ETAs due to their higher TECs and lower moduli ($\approx8.1 \times 10^{-3}$ K$^{-1}$ and 0.28 GPa for a camphorsulfonic acid-doped polyaniline) as compared with metals and carbons.\textsuperscript{160,161} Coating a conductive beam (film or yarn) with thermally active polymers also improved strain/displacements significantly, which lead to heterogeneous ETAs.\textsuperscript{162}

For heterogeneous ETAs, they are usually configured from an electrically conductive component and an electrically insulating but thermally active component, in a form of bilayer strip/film or core–sheath fiber/yarn for achieving various actuation functions. It is more complicated to choose the conductive and insulating materials and combine them into high-performance heterogeneous ETAs. The temperature increment contributed from Joule heating of the conductive component is affected not only by the properties of the conductive component but also by the relative thickness, thermal conductivity, and specific heat capacity of the insulating component. Using highly efficient heating component is more critical for fast heat generation, and increasing the thermal conductivity of the insulating component is important for fast increment of its temperature. Apart from the temperature increment, the total actuation strain or strain difference of heterogeneous ETAs is also dependent on the TEC, thickness ratio, and modulus ratio of both components and the configuration used. If the conductive component is poorly stretchable (i.e., a thin film of a metal\textsuperscript{163} and graphene\textsuperscript{164}), the actuation displacement/curvature is determined by the expansion difference between the conductive and insulating components. This implies that increasing the difference of TEC between the conductive and insulating components will lead to a large actuation strain difference. Bending and torsional bimorph ETAs usually belong to this case. However, if the conductive component is highly stretchable (e.g., a CNT layer wrapped around a coiled polymer fiber with a bias angle),\textsuperscript{18,161} the total actuation strain is largely determined by the thermal expansion of the insulating component because the former can adapt itself to the deformation of the latter. Torsional and contractile core–sheath fiber/yarn ETAs generally fall in this case. In both cases, it is favorable to use an insulating component with a high TEC for improving the actuation performance.

### 3.3. Device Fabrication and Actuation Performance

The fabrication process of soft ETAs involves the preparation and assembly of soft conductive and insulating components which exert a strong influence on their actuation performance. Using commercially available conductive materials and polymers (e.g., paper and tape) is possible to construct scalable soft ETAs with moderate actuation performance from rational designs. Preparation of components from new materials or old materials with new structures allows the exploration of high-performance ETAs. The electrical, thermal, and mechanical properties of the components are decided by the basic properties of the materials used as well as their microstructures (e.g., crystallinity, porosity, and alignment), which can be controlled by their fabrication process. How the components are assembled into an ETA also affects its actuation performance such as durability, actuation direction, and total actuation strain. This section reviews the recent progress on the fabrication and performance of mostly investigated film/fiber-based soft ETAs, with strategies for enhancing the actuation performance being highlighted. The performances of some typical ETAs are shown in Table 2.

#### 3.3.1. Film-Based Soft ETAs

Heterogeneous film-based ETAs are usually assembled in the form of a two/three-layered structure from an electrically conductive film and one/two insulating films by deposition, infiltration, lamination, or adhering with an adhesive layer. Film-based ETAs are generally referred as bimorph ETAs, which usually generate bending motions due to the expansion difference between two layers. Homogeneous film-based ETAs using only one conductive film were also investigated,\textsuperscript{140,141} but usually showed a small actuation displacement. Commonly utilized electrical conductive layers are made of metals (e.g., Au,\textsuperscript{166,167} Ag nanowires [AgNWs],\textsuperscript{158} Ni–Cr wires\textsuperscript{169}), carbons (e.g., CNTs,\textsuperscript{138} graphene,\textsuperscript{170} and carbon fibers\textsuperscript{171}), MXenes (e.g., Ti$_3$C$_2$T$_x$\textsuperscript{172,173}), conductive polymers (e.g., PEDOT:PSS\textsuperscript{174}), or conductive composites (e.g., AgNW/polymethylsiloxane [PDMS]\textsuperscript{175}, graphene/silicon rubber\textsuperscript{145} and Cu/ Ni-coated fabric\textsuperscript{176} CNT/chitosan\textsuperscript{140} MXene/cellulose nanofiber\textsuperscript{173} PEDOT:PSS/paper\textsuperscript{179} AgNW/PEDOT:PSS/paper\textsuperscript{177}). Insulating films are generally made of semicrystalline and elastomer polymers with either a low TEC (as the passive layer) or a high TEC (as the active layer), e.g., polyimide (PI),\textsuperscript{167,169,174} polyethylene terephthalate (PET),\textsuperscript{178} SU-8\textsuperscript{166} cellulose\textsuperscript{177} polycarbonate (PC),\textsuperscript{173} polyethylene (PE),\textsuperscript{168} polypropylene (PP),\textsuperscript{176,177} PVC,\textsuperscript{168} PDMS,\textsuperscript{138,159,179} and liquid crystal elastomer (LCE)\textsuperscript{167,169,180}. As compared with semicrystalline polymers, elastomers generally show higher TECs and may generate higher strains/displacements but possibly lower stresses/forces. Theoretically, there are many combinations for assembling soft ETAs, but considering how to improve the actuation performance is essential. The recent progress on those ETAs will be discussed according to the conductive layers used.

**Soft ETAs based on Metal Films/Layers**: Metal-based conductive layers have a very high electrical conductivity for efficient heating under low voltages, but their high modulus will limit the bending displacement of the insulating layer and therefore the whole ETA. This issue can be avoided by using an ultrathin metal
### Table 2. Summary of the structures and performances of some typical soft ETAs.

| Device structure | Device size [mm²] | Δαb [10⁻⁵ K⁻¹] | Eröb [MPa] | Applied voltage /power | Max. temp. [°C] | Actuation displacement/strain/angle/curvature | Actuation force/stress | Energy density [kJ kg⁻¹] | Te [s] | η [%] | Ref. |
|------------------|-------------------|----------------|------------|-------------------------|---------------|----------------------------------------|-----------------------|------------------------|------|-----|-----|
| (PI/Au/PI)/LCE bilayer | ≈0.503 mm thick | – | 0.5 | 8.2 V | – | 24% | – | – | – | – | [167] |
| Kapton/(Ni–Cr wire)/LCE trilayer | 35 × 3 × 0.22 | – | – | 8.2 V for 5 s | 78 | – | 0.38 MPa | – | 15 | – | [169] |
| PDMS/(AgNW/PDMS)/PI trilayer | ≈0.213 mm thick | 290 | – | 4.5 V for 40 s | ≈155 | 720/2.6 cm⁻¹ | – | – | 60 | – | [175] |
| PVC/AgNW/LDPE trilayer | 30 × 20 | 434 | 100 | 142 mW for 30 s | 42 | 2.5 cm⁻¹ | 1.33 mN | – | 10 | – | [168] |
| (SACNT/PDMS)/PDMS bilayer | 30 × 6 × 0.77 | 304 | – | 40 V for 5 s | 98 | 9.5 mm | – | – | 60 | – | [182] |
| SACNT/BOPP bilayer | 70 × 18 × 0.047 | 121 | – | 5 V for 10 s | 47.6 | 389/1.03 cm⁻¹ | 7 × its gravity | – | 15 | – | [138] |
| PDMS/graphene/PI trilayer | ≈0.25 mm thick | – | 2.5 | 12 V for 3 s | – | 180° | 4 × its gravity | – | 4 | – | [139] |
| Spongy RGO/PI bilayer | 30 × 9 × 0.18 | 128 | – | 16 V for 10 s | ≈100 | 0.55 cm⁻¹ | 37 × its gravity | – | 8 | – | [164] |
| GO/RGO/BOPP trilayer | 25 × 5 × 0.063 | 136 | – | 60 V for 10 s | ≈81 | 72° | – | 15 | – | [170] |
| (MXene/cellulose)/PC bilayer | 40 × 10 × 0.005 | – | – | 5.1 V | – | 147° | – | – | – | – | [173] |
| (AgNW/PEDOT:PSS/paper)/PP bilayer | 45 × 9 × 0.15 | ≈121 | 1500 | 7 V for ≈4 s | 35 | 360°/1.07 cm⁻¹ | 18 × its gravity | – | 18 | 0.028 | [177] |
| Coiled wax/CNT hybrid yarn | d = 0.15 mm⁴ | – | – | 15 V cm⁻¹ of 0.4 Hz (0.05 s on) | – | 10% @ 5.5 MPa | >5.5 MPa | 1 @ 84 MPa | 2.45 | Up to 0.55 | [154] |
| Coiled TPU/CNT hybrid yarn | l = 35 mm | d = 0.21 mm | – | – | 0.05 A cm⁻¹ of 0.1 Hz (square) | ≈230 | –13.8% @ ≈3 MPa | >33 MPa | 0.547 @ 33 MPa | 5 | – | [158] |
| Coiled methanol/silicone/CNT hybrid yarn | d = 0.22 mm | – | – | 5 V for 3 s | ≈75 | –30.5% @ 3.1 MPa | >23 MPa | 0.85 @ 23.4 MPa | 2 | – | [189] |
| Coiled PU@CNT yarn | d = 0.065 mm | – | – | 0.25 W cm⁻¹ for ≈6 s | 85 | –12.6% @ 42 MPa | >56 MPa | 1.28 @ 42 MPa | ≈6 | – | [50] |
| Coiled nylon yarn wrapped by CNT | d = 0.076 mm | – | – | 30 cm⁻¹ of 1 Hz (0.2 s on) | ≈100 | –10.5% @ 22 MPa | >85 MPa | – | 0.8 | Up to 1.08 | [18] |
| CNT-coated spandex yarn | – | – | – | 20 V of 0.2 Hz | 70 | –33% (no load) | >17 MPa | 0.64 | 2.5 | – | [165] |
| CNT-coated rubber | l = 7 cm d = 2.5 mm | 1.1 | 0.7 W cm⁻¹ (150 V) | 125 | –23.2% @ 0.8 Ma | >2 MPa | 1.06 @ 1.6 MPa | – | – | Up to 0.5 | [191] |

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²Difference of thermal expansion coefficient between the bending layers (conducting and insulating layers for bilayered ETA, two insulating layers for the trilayered ETA); ³Modulus of the softer/softest layer; ⁴Recovery time; ⁵Electrical to mechanical energy transduction efficiency; ⁶Diameter.

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film/mesh,[166,181] a serpentine metal wire/pattern,[167,169] a metal-nanowire-based thin pure/composite film,[168,175,177] or a liquid metal film,[160] all of which can adapt to the deformation of the thermal active layer in the ETAs. For instance, Zhu et al. fabricated a micro-ETA based on an Au/SU-8 film with a very thin (200 nm) Au layer deposited by e-beam evaporation, which can produce reversible, rapid, and large folding below 3 V.[166] Nevertheless, the cracking of the Au layer during repeated folding might be an issue. A thin, deformable, serpentine PI/Au/PI (thickness: ≈1.2/0.2/1.2 μm) heater was used to prepare a contractile ETA by attaching it onto a thick LCE film, which showed a large contracting strain of ≈24% under a voltage of 8.2 V due to the thermal shrinkage of the LCE at ≈80 °C.[167] Meanwhile, a serpentine Ni–Cr wire was sandwiched between a Kapton film and a liquid crystal network of elastomer (LCN) film to give a programmable ETA (Figure 9a), which can rise to 78 °C and curl up to 760° under 8.2 V for 5 s because of the thermal expansion of the Kapton and ultralarge thermal shrinkage of the LCE.[169]
this design, quality of the interface between the conducting wire and the polymer will be very important. Using stretchable AgNW/PDMS film as the heating layer, a PDMS/(AgNW/PDMS)/PI trilayer bimorph ETA showed a bending of 72° with
a curvature of 2.6 cm\(^{-1}\) under a voltage of 4.5 V (0.2 V sq\(^{-1}\)) for \(\approx 40\) s owing to the very large TEC difference between PDMS and PI films (\(310 \times 10^{-6}\) vs \(20 \times 10^{-6}\) K\(^{-1}\)).\(^{[176]}\) A transparent, percolated AgNW network was deposited onto a low-density PE (LDPE) film and covered with a PVC film to assemble a soft ETA, which can bend to a curvature of \(2.5\) cm\(^{-1}\) under an input power of \(142\) mW for \(30\) s due to the large TEC difference between two polymers (\(\alpha_{LDPE} = 502.7 \times 10^{-6}\) K\(^{-1}\) along the longitudinal direction, \(\alpha_{PVC} = 68.7 \times 10^{-6}\) K\(^{-1}\)).\(^{[184]}\) However, the high cost of Au and Ag and thermal oxidation of Ag will limit their practical application.

**Soft ETAs based on CNT Films:** Conductive 1D CNT films are widely studied as the heating layers for soft ETAs due to their higher flexibility, durability, and surface area (favorable for bonding with the insulating layer) than traditional metal films. Li et al. fabricated a (CNT/PDMS)/PDMS bilayer ETA simply by pouring excess PDMS precursors onto \(50\) layers of super-aligned CNT (SACNT) sheets followed by thermal curing, which showed an increased temperature of \(98\) °C and a bending displacement of \(9.5\) mm (initial length: \(30\) mm) under \(40\) V for \(5\) s, resulting from the large TEC difference between the conductive SACNT/PDMS layer (with \(5\) wt% CNT, \(\alpha = 6 \times 10^{-6}\) K\(^{-1}\)) and insulating PDMS layer (\(\alpha = 3.1 \times 10^{-4}\) K\(^{-1}\)).\(^{[182]}\) The significantly reduced TEC of the composite was close to that of the SACNT (<\(3 \times 10^{-6}\) K\(^{-1}\)) because the strong interaction between PDMS and SACNT restricts the expansion of PDMS along the alignment direction. To simplify the fabrication, a commercial biaxially oriented PP (BOPP) film (\(40\) μm) was adhered onto a compact SACNT film (\(7\) μm) to give a bilayer ETA, which achieved a bending angle of \(389\)° and a curvature of \(1.03\) cm\(^{-1}\) under only \(5\) V for \(10\) s due to the low thickness and large TEC (\(124 \times 10^{-6}\) K\(^{-1}\)) of the BOPP film.\(^{[139]}\) The good transparency of thin SACNT film also makes it possible to fabricate a transparent ECA. To sandwich an ultrathin SACNT film as the heating layer between a PET and a PDMS film, the resulting transparent ETA can bend over \(220\)° under \(100\) V within \(8\) s and was used to control the magnification effect of a solid-state lens.\(^{[178]}\) However, the required high voltage and high temperature during operation may limit their application in wearable fields.

The alignment of CNT in the conductive film affects its thermal expansion and electrical conductivity along the target actuating direction and therefore the actuation performance of the ETAs. For example, T-shaped (SACNT/PDMS)/PDMS ETAs (Figure 9b) with different bending directions were fabricated by directly cutting the (SACNT/PDMS)/PDMS film into a T shape at a desired angle relative to the SACNT alignment direction.\(^{[179]}\) The arm with perpendicularly aligned CNT showed much larger bending due to its higher resistance and temperature increment during Joule heating. ETAs with complex actuation behavior were also achieved by directly cutting different patterns from the composite.\(^{[179,184]}\)

**Soft ETAs based on Graphene and MXene Films:** Conductive 2D Graphene and MXene films are also potential conductive heaters for soft ETAs due to their good flexibility, low density, and high electrical conductivity. Graphene films have excellent thermal stability and potentially low cost as compared with noble metal films. The conductivity, porosity, flexibility, and thermal expansion/shrinkage of graphene films can be controlled by the preparation method, generally by reduction of GO films, filtration from RGO dispersion, or laser scribing of polymer or GO films. A PDMS/graphene/Pt trilayer ETA using a laser-induced porous graphene pattern on a PI film (Figure 9c) attained a bending of \(1080\)° (curvature = \(3.3\) cm\(^{-1}\)) under 30 V (8 V sq\(^{-1}\)) and can lift a load of \(\approx 110\) times of their weight, resulting from the low sheet resistance of graphene layer and the optimized thickness and modulus of the PDMS layer.\(^{[139]}\) This method allows the fabrication of various shaped ETAs with complex actuation behavior due to the facile laser patterning and cutting. Yang et al. produced a spongy RGO (SRGO) film with microgasbags by electrical heating of a RGO film, which showed an abnormal large negative TEC of \(-1.16 \times 10^{-4}\) K\(^{-1}\).\(^{[164]}\) The assembled SRGO/Pt bilayer ETA can bend to a curvature of \(0.55\) cm\(^{-1}\) under 16 V for 10 s and generated a blocking force of \(37\) times its weight due to the large shrinkage of SRGO and expansion of Pt. In addition, hygroscopic GO films can also be used as a thermal shrinkage layer due to dehydration under heating. A Janus RGO/GO film made by laser scribing of a GO film was used to assemble a BOPP/RGO/GO trilayer ETA, which can bend from \(-10\)° to \(-82\)° under \(60\) V (42 mW mm\(^{-1}\)) for \(10\) s due to the thermal expansion of the BOPP tape and thermal shrinkage of the GO film during electrical heating by the RGO layer.\(^{[150]}\) Due to the low conductivity of the laser scribing induced RGO, a high applied voltage for the actuation is required.

MXene-based films can also be used as heating layers due to their high electrical conductivity. Interestingly, hygroscopic MXene film can shrink during electrical heating due to dehydration. Cai et al. fabricated a layer-inspired bilayer ETA (Figure 9d) by simple filtration of a MXene/cellulose nanofiber composite film onto a porous PC film, which can achieve a bending angle change of \(147\)° under only \(5.1\) V (106 mW cm\(^{-1}\)) due to the thermal expansion of the PC film and thermal shrinkage of the hygroscopic MXene/cellulose layer.\(^{[173]}\) The porous PC film also favored the fast evaporation/absorption of water during heating/cooling and thus a quick response. In addition, a MXene/graphene (1.25 in mass) bilayer film was used as a heating and mechanical sensing layer simultaneously for a multifunctional soft ETA due to their ultralow temperature coefficient of resistance.\(^{[184]}\) However, the weak stability of MXene in air might limit their durability.

**Soft ETAs based on Conductive Polymer Films:** The primary conductive polymer films used for soft ETAs are PEDOT:PSS-based ones due to their high electrical conductivity, excellent flexibility, and good processability. The high hygroscopicity of PEDOT:PSS can also be used to enhance the thermal shrinkage. Hamedi et al. produced a flexible PEDOT:PSS-coated cellulose paper as the conductive heating layer, on which a Scotch tape was attached to give a soft bendable bilayer ETA (Figure 9e).\(^{[19]}\) It can bend toward the paper side and generate a blocking force of up to \(20\) times its gravity under \(20–100\) V (0.3 W cm\(^{-2}\)) due to the thermal expansion of the tape layer and shrinkage of the hygroscopic PEDOT:PSS/paper layer.\(^{[19]}\) A creased-sawtooth ETA was also fabricated simply by attaching the tape onto alternative sides of a folded PEDOT:PSS/paper, which can generate accordion-like motion. However, due to the large resistance of PEDOT: PSS/cellulose paper (70–350 Ω sq\(^{-1}\)), a relatively high voltage to drive the actuator is required. Amjadi and Sitti fabricated a similar multilayer ETA but from a highly conductive AgNW/PEDOT:PSS-coated copy paper (\(R_s = 1.03\) Ω sq\(^{-1}\)) and a
self-adhesive hydrophobic PP film, which can bend to a curvature of 1.07 cm$^{-1}$ (360°) at a low-temperature increment (≤35°C) under 7 V.[177] The excellent actuation performance is ascribed to a similar mechanism enhanced by the efficient heating by the highly conductive layer. The anisotropic structure of the paper also rendered a tailor actuation by changing the angle between the fiber direction of the paper and the longitudinal direction of the ETA.[177] But the actuation performance is affected by the humidity and may be difficult to control.

In summary, using highly conductive heating layers and enlarging the thermal expansion difference between the layers can effectively enhance the actuation performance of bilayer/trilayer ETAs under low heating voltages. One way to enlarge the expansion difference using two layers with opposite thermal expansion, i.e., one layer expands while the other shrinks under the electrical heating. Liquid crystal elastomer with ultrahigh thermal contraction along the drawing direction is preferred as the insulating layer.[167, 169, 180] Hygroscopic layers, either conductive or insulating, also show a high thermal shrinkage due to the thermal dehydration of the hydrophilic material incorporated (e.g., cellulose paper,[167, 184] cellulose nanofiber,[173] PEDOT:PSS,[19] polyvinyl alcohol,[185] graphene oxide,[170] and MXene[173]). A second way is to use a layer with giant thermal expansion, which can be realized by filling the layer with a phase change material (e.g., a low melting point wax and volatile liquid such as ethanol)[146, 186] that shows a large volume expansion during the phase change from solid to liquid or liquid to gas. In addition, it is worth noting that the actuation direction of a film-based ETA is affected by the anisotropic characters (electrical conductivity, thermal expansion, and Young’s modulus) of both the conductive and insulating layers and can be controlled by aligning their main orientation directions to a desired angle during the assembly/fabrication process.[168, 176, 179] Improving the thermal conductivity of insulating polymer layers could also enhance the actuation response, which can be achieved by addition of thermal conductive nanofillers such as CNTs and carbon black.[167, 187]

3.3.2. Fiber-Based Soft ETAs

Fiber-based soft ETAs are assembled from one or two kinds of fibers in the form of a yarn of textile and are promising for applications such as smart clothing and lightweight exoskeletons due to their high mechanical strength, flexibility, permeability, and weavability. The main fibers investigated are carbon-based fibers and polymer-based fibers due to their low density. Fiber-based ETAs can generate bending, torsional, and tensile actuation strain and stress depending on their configurations.

Carbon-based fibers include traditional carbon fibers, nanocarbon-based fibers (e.g., CNT or graphene-based fibers), with the latter mostly used for yarn/textile ETAs because of their better flexibility and higher breaking strain than traditional rigid carbon fibers. Straight carbon fibers generate low actuation strain under a low voltage due to their small TEC. Effective strategies for improving the actuation performance include incorporation of fillers with a high TEC inside or on the surface, twisting and coiling treatment of the fiber, as well as combing both.[149, 154, 158, 159, 188–190] A SWCNT/GO/PVA hybrid fiber clamped at both ends (distance of 2 cm) generated a maximum bending displacement ≈5 μm under a 0.2 Hz sinusoidal potential of 5 V due to the thermal expansion.[157] However, the displacement is very small and may be due to the low electrical conductivity, relatively small TEC of the fiber, and the configuration used. Lima et al. showed that twisting and coiling of a CNT yarn can obviously amplify the tensile stroke and work capacity.[154] Further infiltration of wax into the coiled CNT yarn (Figure 10a) dramatically increased the thermal tensile contraction due to the large volume expansion of the wax (≈20%/between 30 and 90°C). A wax-filled coiled CNT yarn achieved a 3% tensile contraction for 1.4 million cycles while lifting a load of 17 700 times its own weight (14.3 MPa) under an 18.3 V cm$^{-1}$ square wave voltage of 20 Hz.[154] It is likely that the leakage and flowing of wax may occur during high-temperature operation.

Modification of a coiled CNT yarn by infiltrating polymer inside or coating on the surface was proven effective to improve the actuation performance.[50, 158, 188, 189] To this end, elastomers are preferred because of their high TEC. A coiled, homogeneous thermoplastic polyurethane (TPU)/CNT hybrid yarn can be prepared by solution infiltration of TPU into a twisted CNT yarn before cooling treatment (Figure 10b).[158] The hybrid yarn ETA lifting 1905 times its weight generated a contraction stroke of ≈13.8%/within 5 s under a square wave power of 0.175 A (3.5 cm) and 0.1 Hz and showed excellent retention of tensile contraction over 600 cycles, which was ascribed to the reversible, high thermal expansion of the shape-memory TPU.[158] Further work in this direction, a mixture of elastomer and volatile methanol was infiltrated into a coiled CNT yarn to give a hybrid yarn ETA, which can produce a tensile contraction of 30.5%/within ≈3 s under a 3.1 MPa stress and a 5 V DC voltage due to the large volume expansion induced by the confined evaporation of methanol during electrical heating.[189] But the gradual loss of methanol through evaporation out of the elastomer may be an issue which may limit its service life. Compared with infiltration of polymer inside a coiled CNT yarn, surface coating may allow higher actuation performance due to less confinement of the polymer by the CNT. Mu et al. produced a set of coiled polymer@CNT (sheath@core) yarns as shear-run artificial muscles (SRAM) by coating a polymer (PU or PEO-SO$_3$), a blend of polyethylene oxide and fluorinated copolymer) layer only on the surface of a twist-spun CNT yarn before coating treatment (Figure 10c), which showed more than 100% higher work capacity and power density than the coiled polymer/CNT hybrid yarn muscle (HYAM) under an input electrical power of 0.25 W cm$^{-1}$ due to the larger volume and modulus changes in the sheath layer.[50] The PU@CNT yarn generated a contraction stroke of ≈12.6%/and work capacity of 1.28 kJ kg$^{-1}$ under an input power of 0.25 W cm$^{-1}$ for ≈6 s. And the PEO-SO$_3$@CNT yarn operated in air produced a contraction stroke of 3.2%/and a fast input power of 2.6 W g$^{-1}$ and 9 Hz.[50] However, the low thermal conductivity of the polymer sheath may limit their actuation response and recovery rates.

Polymer-based fibers are promising in soft ETAs due to their significantly higher TEC and lower cost as compared with carbon-based fibers. To allow electrothermal heating, intrinsically conductive polymer fibers or insulating polymer fibers coated with electrically conductive layers are used for ETAs. The electrothermal actuation performance of conductive polymer fibers is affected by their electrical conductivity and mechanical
An ethylene glycol (EG)-doped PEDOT:PSS fiber was fabricated by wet-spinning and hot-drawing, and showed a high electrical conductivity (3132 S cm⁻¹) and therefore a large heating rate (63 °C/s) comparable with metal wires. It also exhibited a fast cooling rate due to the absorption of water by the small-diameter fiber during cooling. After applying a voltage of 1.14 V cm⁻¹ for ~6 s, its temperature increased to ~40 °C and it generated a contraction strain of 0.4% under a load of 150 times its weight. However, those conductive polymer fiber-based ETAs show low actuation strokes due to their relatively small thermal expansion/shrinkage coefficient and the use of straight beam configuration. Inspired by coiled CNT yarn ETAs, twisting and coiling treatment of conductive polymer fibers are also expected to enhance their actuation stroke and work capacity.

Conventional insulating polymer fibers have large TECs but can provide axial contractions due to the conformation change in the preoriented noncrystalline region when they were heated. For instance, a high-strength nylon 66 monofilament showed a reversible contraction strain as large as 4% at 240 °C. Therefore, polymer fibers have a great potential in fabricating low-cost, scalable soft ETAs, where electrical heating can be provided by a conductive layer coated on the fiber or an extra conductive wire/fiber. The actuation performance (e.g., maximum stroke, work capacity, and power density) of...
polymer fiber-based ETAs can be greatly enhanced by twisting and coiling treatment and tuned by adjusting the spring index of the coil and is also dependent on the modulus of the fiber.[18] Haines et al. found that a coiled nylon 66 monofilament showed a dramatically higher contraction of 34% at 240 °C.[18] After wrapping a CNT sheet on the surface the resulting CNT@nylon yarn ETA (Figure 10d) generated a stable contraction stroke of ≈10.5% for 1.2 million cycles under a 22 MPa loading driven by a 1 Hz square wave potential of 30 V cm−1. Again, their actuation response and recovery rates are limited by the low thermal conductivity of the polymer fiber.

Elastomer fiber-based ETA can provide a large contraction strain/stroke without the need of twisting or coiling treatment due to their large TEC, and can also work as strain sensors simultaneously.[165,191] CNT-coated Spandex yarn (CNT/SPX) textiles were prepared by direct knitting from a CNT forest and a stretchable Spandex yarn with n (n = 1, 4, 8, 12) fibers (Figure 10e).[165] A CNT/SPX5 textile ETA generated a highest tensile contraction of 33% when prestretched to 100% strain and heated to 70 °C by an on–off voltage of 20 V and 0.2 Hz due to the axial contraction of the Spandex fiber, while a CNT/SPX1 textile ETA provided the maximum work capacity of 0.64 J kg−1 and power density of 1.28 W kg−1 under the same condition except a 2 Hz voltage. A stretchable, self-sensing ETA prepared by adhering several CNT sheets onto a prestretched natural rubber fiber can produce a tensile contraction of 23.2% under a 0.8 Ma stress when heated to 125 °C by a power of 0.7 W cm−1 (150 V) and a maximum work capacity of 1.06 J kg−1 under a 1.6 MPa load.[191] However, the low modulus of the elastomer fiber will lead to a low actuation force/stress. In addition, elastomer polymer can be used the thermoactive scale objects such as cells and characterize mechanical techniques and were used to manipulate nanoscale/micro-grippers based on ETAs can be fabricated by microfabrication techniques and were used to manipulate nanoscale/micro-scale objects such as cells and characterize mechanical performance of nanomaterials.[193,194] Macroscopic soft grippers based on ETAs are easier to be fabricated in various configurations (e.g., with multiple arms or suckers) for manipulating small/fragile/sensitive objects,[169,174] and more functionalities such as sensing can be integrated with the ETA to produce intelligent interactive grippers.[180,184]

A trigger-activated gripper/artificial flytrap using a self-sensing PDMS/liquid metal/LCE ETA (Figure 11a) was made by rationally designed liquid metal circuit layer functioned as the electrical heater and mecha-sensor simultaneously.[180] Under a constant applied current, the artificial flytrap is open because most current flows through the middle line due to its low resistance. Once a pressure was applied on the middle, the current flowing through the side circuits was increased to enhance the Joule heating, resulting the bending of two sides and closure of the artificial flytrap. An intelligent gripper was also demonstrated by attaching a glass rod tip onto the flytrap, which can automatically grip a cherry tomato when touching it and release it after lifting for 28 s.[180] However, there is a waste of energy of the intelligent flytrap/gripper before being triggered because they require the voltage to be applied all the time.

Biomimetic soft robots inspired by plants or animals can be constructed from soft ETAs and achieve various functions such as flower-like blooming,[168,176] curling/winding,[168,169] crawling,[169] wing flapping,[160,166] color changing,[168,184] and shape morphing.[166] Potential applications can be found in unmanned patrolling or detection especially in harsh environments. It is challenging to control the motion of those robotics especially in a remote fashion. Wang designed a self-controlled walking robot driven by a CNT-wrapped rubber fiber ETA based on the resistance feedback sensed by itself, which can autonomously walk continuously on a ratchet surface when the voltage is always on.[191] A wave-like crawling robot based on a programmed PI/Ni–Cr wire/LCN ETA with two parts with opposite actuation directions and different shapes (Figure 11b) can crawl toward one direction on a flat plane or in a tube under an on–off potential.[169] Another crawling robot by integrating two photo detectors into the heating layers of two ETAs allows the remote control of the movement by shining laser beams of two different colors onto those detectors that control the working of two ETAs separately.[167] A flower-like ETA based on PI/graphene/PDMS was demonstrated to be controlled by human gestures sensed by electromyography (EMG) sensors on the fore arm of the operator (Figure 11c).[119] Color changing ETAs by depositing thermochromic pigments on their surface (Figure 11d) displayed different color while changing their shape under a low-temperature increment and are promising in camouflage robotics/devices.[168] Yi et al. demonstrated a shape-morphing microorigami robot by integrating the origami system with soft Au/SU8 micro-ETAs (Figure 11e), which can realize fast morphing due to the elastic folding of the actuators under electrical heating.[166] Interestingly, the 3D geometries were reprogrammable by overheating those actuators to allow plastic folding. However, all those robotics were tethered; realization of untethered operation and control is critical for practical application.

Soft ETAs also show promising applications in tactile displays, which can provide both haptic and thermal information/feedback. A typical example for tactile display is refreshable braille display. Lee developed a refreshable 3 × 2 dot braille cell composed of six micromachined silicon containers, each filled with wax and sealed by an elastic silicone diaphragm, and six gold microheaters at the bottom (Figure 11f).[116] Each dot can be raised up by 654 μm under electrical heating due to a 15% volume expansion of the melted wax. Under an input power
of 0.3 W (only \(\approx 7\) V) and a duty factor of 0.8, the dot height can be maintained at 50% of its maximum and the total cycle time was \(\approx 50\) s. The operation voltage is significantly lower as compared with the counterpart based on dielectric actuators, but the refreshing rate is limited to the slow cooling process, which might be improved by new materials or new configuration designs.
Fiber/yarn/textile-based soft ETAs can find potential applications in lightweight exoskeletons, smart clothing, and medical devices owing to their high contraction stroke and work capacity, all of which directly interacts with human body. For example, Haines et al. showed that a textile ETA, woven from 12 coiled nylon filament as the weft and Ag-plated nylon, polyester, and cotton yarn as the warp, produced a contraction of 17.6% while lifting a 3 kg load under electrical heating, rendering it promising in lightweight exoskeletons for rehabilitation patients and manual workers/soldiers. They also demonstrated that a braid with a coiled Ag-coated nylon ETA inserted in the middle opened its pores under electrical heating (Figure 11g) and it was possible to close the pores by changing the braid bias angle and chirality of the yarn ETA. Therefore, fiber ETAs are promising in comfort-adjusting smart clothing. However, the undesirable high temperature may bring discomfort to the wearer. A pressure-adjustable compression bandage was woven from a PDMS/Cu/PI yarn ETA as the weft and a polyester yarn as the warp, and can increase its pressure by 44.7% when electrically heated from 25.6 to 56.2 °C under 2 W and may find applications in compression garments and adjunctive therapy for varicose vein. Kongahage et al. showed that an artificial heart muscle “sleeve” (based on silicon-coated, coiled, Ag-plated nylon yarns) (Figure 11h) was able to generate a 120 mmHg pressure and a high blocking stress to support the left ventricle of a heart under a voltage of 32.5 V. This makes it possible to greatly reduce the total mass and volume of the current ventricular assist device. To address this, various fabrication methods have commonly been used to achieve thin elastomeric films. Generally, DEAs are composed of a thin elastomer film that is sandwiched between two compliant electrodes (Figure 12a). To induce actuation, an electric field is applied across the electrodes, generating an electrostatic pressure that compresses the elastomer in the thickness direction and expands in area. Fundamentally, this electrostatic pressure otherwise known as the Maxwell stress is intrinsically related to actuation performance of the DEA and can be expressed by the Equation (5)

\[ P = \epsilon_0 \epsilon_r \left( \frac{V}{d} \right)^2 \]  

where \( \epsilon_0 \), \( \epsilon_r \), \( V \), and \( D \) represent the vacuum permittivity, dielectric constant, voltage applied, and the thickness of the elastomer, respectively. Furthermore, based on Hooke’s law of an ideal elastomer that is assumed to have an incompressible nature with Poisson ratio of 0.5 and an isotropic elastic modulus \( Y \) independent of strain, the actuation strain in the thickness direction \( s_z \) can be expressed from Equation (6)

\[ s_z = -\frac{P}{Y} \]  

While this expression is true for linear elastic elastomers and at small strain, it provides an insight on the impact of the elastomer’s mechanical properties on the actuation performance. Therefore, when designing high-performing DEAs, it is critical to select elastomers with a low elastic modulus and a high dielectric constant. Also, to generate the largest possible actuation at high voltages, a high dielectric breakdown strength is favorable for DEAs. Today, elastomers used for DEAs have included silicone, acrylic elastomers, polychloroprene, natural rubber, and styrenic-based block copolymers.

### 4. Soft Dielectric, Electrostatic, Ferroelectric, and Plasticized Gel Actuators

In contrast to ECAs that generally require a wet state or solid electrolyte that are catered for the mobility of ions to drive actuation electronic EAPs are bulk polymer insulators that are driven by electric fields or Columbic forces. Generally, these electronic EAPs are able to sustain activated displacements under DC voltages and often show higher energy densities, forces, rapid response times, and can be easily operated in air. Particularly, actuators composed of electronic EAPs include DEAs, electrostatic actuators, ferroelectric polymers, and plasticized gel-based actuators. However, these actuators often require high driving voltages that are close to the breakdown field of the material that poses as a safety hazard for iHMI. Table 3 shows a comparison over the key performances of electronic EAP-based soft electrical actuators. In this section, we review the various efforts that have been utilized toward lowering the high driving voltage of electronic EAP actuators as well as their applications within iHMI.

#### 4.1. DEAs

DEAs are a class of EAPs that have achieved high actuation strains, fast response speeds, lightweight, quiet operation, large energy densities, and self-sensing properties, proving to be advantageous for iHMI applications. Generally, DEAs are composed of a thin elastomer film that is sandwiched between two compliant electrodes (Figure 12a). To induce actuation, an electric field is applied across the electrodes, generating an electrostatic pressure that compresses the elastomer in the thickness direction and expands in area. Fundamentally, this electrostatic pressure otherwise known as the Maxwell stress is intrinsically related to actuation performance of the DEA and can be expressed by the Equation (5)

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where \( \epsilon_0 \), \( \epsilon_r \), \( V \), and \( D \) represent the vacuum permittivity, dielectric constant, voltage applied, and the thickness of the elastomer, respectively. Furthermore, based on Hooke’s law of an ideal elastomer that is assumed to have an incompressible nature with Poisson ratio of 0.5 and an isotropic elastic modulus \( Y \) independent of strain, the actuation strain in the thickness direction \( s_z \) can be expressed from Equation (6)

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#### 4.1.1. Designing Low-Voltage DEAs

The main drawback of DEAs centers around its need for high driving voltages, typically above 1 kV. Therefore, in this section, we focus on the various strategies to lower the driving voltages such as techniques to lower the film thickness, raise the dielectric constant, and reduce the elastic modulus to ensure low-voltage requirements while maintaining its favorable actuation performance.

Reducing Thickness: To generate sufficient electric fields to drive DEAs at low voltages, scaling down the elastomer thickness is required. Not only does this provide greater safety to users but allow DEAs to be compatible with simple commodity driving electronics. Nonetheless, fabricating these thin elastomers poses the challenge of higher defect density from thickness inhomogeneity or contaminants that leads to local weak zones that cause premature breakdown. To address this, various fabrication methods have been proposed to design these thin DEAs.

Blade-casting and spin-coating techniques have commonly been used to achieve thin elastomeric films. To avoid damage from forces used to release these thin films from substrates, films are often coated onto a water-soluble sacrificial layer that is removed through immersion of hot water. Using a water-soluble
sacrificial layer for release, Ji et al. fabricated a blade-casted 23 μm-thick PDMS film, at which after prestretch became only 6.5 μm thick.\textsuperscript{[20]} By stacking three layers of these films alternating between four thin single-walled carbon nanotube electrodes, a low-voltage DEA was designed and was used to drive a soft robotic insect at only 450 V. Another technique, pad printing, has been used to achieve DEAs comprising thin PDMS elastomers and μm-thick carbon black elastomer electrodes.\textsuperscript{[214]} With PDMS thickness of 3 μm, the DEA realizes a lateral actuation strain of 7.5% at only 245 V. Although actuation strains due to premature breakdowns were half of a thicker DEA (30 μm), the driving voltage was reduced from 3.3 kV to 245 V.

For further reduction to the driving voltage to a few volts, nanometer-thin films are required. Regarding these nanometer-thin films, stacked DEA configurations are preferred as freestanding films may be too fragile to handle. One method that is promising to achieve such films is molecular beam deposition (MBD) of PDMS.\textsuperscript{[215,216]} By tuning the polymer chain length of PDMS, elastic properties and deposition rates of the films were being tailored to form nanometer-thick cantilever actuators.\textsuperscript{[215]} As such, driven at only 12 V, fabricated 200 nm-thin devices could achieve bending curvatures of 1.25 m\(^{-1}\). This was a drastic reduction compared with spin-coated Elastosil-based actuators of 4 μm thick that obtained similar curvatures at 500 V. However,
deposition rates from MBD are generally slow (130 nm h⁻¹), making it challenging to be adopted commercially. In contrast, electrospaying has been proposed for fabricating low-voltage DEAs where nanometer-thin films can be obtained at growth rates of 0.02–5.4 nm s⁻¹. While a functional DEA has not been demonstrated with this technique, the smaller roughness achieved compared with spin coating highlights the potential for fabricating thin films with lesser local weak zones that contribute to early breakdown of low-voltage DEAs.

The design of electrodes of thin film DEAs becomes critical with the reduction of thickness. Generally, electrodes utilized for DEAs are assumed to have little effect on the stiffness of the entire device. However, this is not the case when the thickness of the elastomer matches with the electrode, at which the combined stiffness limits the actuation strains obtained. Therefore, electrode thickness needs to be reduced to the nanometer-scale while retaining its conductivity when stretched. Concurrently, electrodes are required to have good adhesion to the elastomer and electrode patterning techniques should not damage the thin elastomers during deposition. To meet these requirements, horizontal Langmuir–Schaefer method has been utilized to fabricate monolayer electrodes of between 2 and 30 nm thickness. Composed of MWCNT and poly(alkylthiophene) that imparted stable electrical conductivity and mechanical properties, respectively, electrodes remained conductive for up to 100% strain. As such, when patterned onto a 1.4 μm-thick PDMS membrane, fabricated DEAs achieved a linear strain of 4.0% at 100 V. When designing thin stretchable electrodes, it is critical to reduce the electrode resistivity which influences the electrical charging time and, thus, the DEA response speed. Alternatively, other deposition techniques such as supersonic cluster beam implantation show promise of reducing the electrode thickness for thin DEAs.

Increasing Dielectric Constant: By increasing the dielectric constant of the elastomer, DEAs are able to generate a larger Maxwell stress or actuation at a lower voltage requirement. Currently, various methods have been adopted to improve the dielectric constant such as blending with high dielectric constant fillers (extrinsic enhancement) or chemical modifications of the elastomer (intrinsic enhancement).

Blending with high dielectric constant fillers provides a rapid and facile approach to achieve elastomers with improved dielectric constants. This is ascribed to interfacial polarization effects (Maxwell–Wagner–Sillar relaxation) from the accumulation of charges at the filler/matrix interface. Generally, extrinsic enhancement approaches adopted include 1) the addition of high dielectric ceramic powders such as barium titanate (BaTiO₃) and titanium dioxide (TiO₂), 2) conductive particles such as aluminum nanoparticles, polyaniline, and graphene; 3) polymer blends; and more recently 4) liquid fillers such as liquid metal and ionic liquid that are mixed with the elastomer matrix. While extrinsic enhancements have been successful to enhance the elastomer dielectric constant, they often come at a trade-off of either increased elastic modulus, higher dielectric loss, lower dielectric breakdowns and greater leakage currents. Moreover, it is crucial to consider the filler size introduced as it limits the possible thicknesses that can be achieved.

In these composites, spontaneous filler aggregation due to surface energy differences between filler and elastomer matrix should be avoided as it could lead to premature failures from interfacial defects and field localization. For improved interfacial compatibility, surface modification of fillers has been undertaken to chemically or physically anchor these fillers to the elastomer matrix. Alternatively, simultaneous addition of two differing fillers have been explored, at which one acts as a physical barrier to avoid agglomeration of the other.

The addition of ceramic fillers has been successful in enhancing the dielectric constant of elastomers, but this is often achieved at higher filler contents that increase the elastic modulus. As this may raise voltage requirements for actuation, plasticizers have been introduced to counteract these effects. In addition, by utilizing polar plasticizers such as diocetyl phthalate (DOP) epoxyized soybean oil and tri-n-butyl phosphate, the overall dielectric constant can be further increased. For instance, Ni et al. introduced DOP to modified TiO₂ natural rubber composites that reduced the elastic modulus from 0.91 to 0.49 MPa due to weaker filler enforcement and plasticizer swelling effects. The polar nature of DOP led to a rise in dielectric constants from 3.4 to 4 at 1 kHz which, in combination with modified TiO₂, resulted to a 3.6 times enhancement in actuation strain at 40 V µm⁻¹. Modification of TiO₂ with γ-methacryloxypropyl trimethoxy silane ensured well-dispersed fillers with high interfacial compatibility that improved interfacial polarization effects. However, the usage of plasticizers often leads to a reduced dielectric breakdown strength from plasticizer-induced defects. Other efforts to reduce to elastic modulus of DEAs to counter the effect of high dielectric ceramic fillers can be adopted and are discussed in detail in Section 2.1.3.

On the contrary, conducive fillers can achieve higher dielectric constants at loadings based on classical percolation theory. In addition to interfacial polarization contributions to the dielectric constant, these conductive fillers further improve the dielectric constant through microcapacitor mechanisms where fillers act as electrodes that sandwich the insulating elastomer. However, this approach tends to be sensitive to the formation of conductive pathways that lead to large dielectric losses and low breakdown strengths. Therefore, to achieve the coexistence of high dielectric constants, low leakage loss, and high breakdown strengths, insulating shell structures have been a promising approach where conductive fillers are coated with thin insulating layers. The success of this approach was demonstrated by Zhang et al., who introduced 0.9 wt% of core–shell CNT-Al₂O₃ to PDMS with minimal changes to the elastic modulus. Inclusions of Al₂O₃ nanolayer suppressed charge mobility and resisted propagation of electrical tree channels within the material leading to low dielectric losses and high breakdown fields, respectively. Therefore, the collective contributions enabled actuation strains achieved to be at least 200% higher than PDMS, while diaphragm-mode actuators vibrated at only 425 V.

Intrinsic enhancements to raise the dielectric constant of elastomers involve covalent bonding of additional dipole moments to the elastomeric network. The dielectric constant enhancement can be attributed to the introduction of orientation polarization effects at which permanent dipoles align with the applied electric
field. In addition, as greater dissimilarity between positive and negative centers of the polymer chains arises, further contributions from atomic polarization improve the dielectric constant. Various polar groups such as cyanoester, azobenzene, sulfonyl, polyethylene glycol, thioether, nitroaniline, and nitrite have been introduced to the polymer backbone to enhance the dielectric constant. Following this strategy, Perju et al. synthesized a silicone elas- tomer with large amounts of nitroaniline functionalities that raised dielectric constants to 18.7 at 1 kHz. Mechanical properties were tuned by varying the content of cross-linkers to obtain low modulus (0.248 kPa) with minimal hysteresis. Eventually, these properties enable DEAs with film thicknesses of 107 μm to achieve lateral actuation strains of 7.2% at 900 V. The process-

ability of these polar elastomers into thinner films was explored by Sheima et al. through a two-step synthesis process of nitrile functionalized silicone (Figure 12c). Owing to high dielectric constants of ≈18 at 10 kHz and low storage modulus of 350 kPa, thin DEAs (35 μm) could obtain stable actuation strains of 4% and 9.7% at 200 and 400 V, respectively, over 100 cycles. While intrinsic modifications have shown great promise for lowering the voltage, these modifications are often performed on polysiloxanes with low dielectric constants. This is attributed to the increase in glass transition temperatures \( T_g \) from modifications with polar functionalities and thus polysiloxanes with low \( T_g \) are often selected. Alternatively, chemical mod-

ifications of polar elastomers such as polyurethanes and polyacrylates can be considered to further enhance maximum dielectric constants. However, high driving voltages are still required for these elastomers owing to their higher elastic modulus that needs to be reduced.

**Reducing Elastic Modulus:** Recently, various efforts have been dedicated toward lowering the driving voltage of DEAs through developing elastomers with low elastic modulus. Strategies include molecular designs, addition of plasticizers, and thermal softening.

Bottlebrush architectures have been shown to be a promising molecular design platform to achieve low elastic moduli. Essentially, these bottlebrush elastomers are a form of grafted elastomers with polymeric side chains attached onto its linear backbone (Figure 12d). By tuning the side chain length, a wide range of elastic moduli can be achieved from 1 MPa to 100 Pa. Furthermore, as such architectures possess inherently strained polymer networks, electromechanical instabilities are suppressed, avoiding the need for prestrains that require rigid components. Owing to their ultralow moduli, bottlebrush elastomers have achieved actuation strains larger than 300% at low electric fields up to 10 V μm\(^{-1}\). However, owing to their ultralow modulus and strength, these bottlebrush DEAs can only be utilized under low-stress environments. Another unique molecular architecture adopted for DEAs are slide-ring materials that possess freely movable cross-links (Figure 12e). Composed of a necklace-like architecture with highly polarizable α-cyclodextrin, slide ring materials have high dielectric constants of 9–10 at 1 kHz. Unlike conventional elas-

tomers whose elasticity is mainly governed by conformational entropy, the low elastic modulus of slide ring materials has been attributed to the additional dependence on entropic elasticity.

By reducing the cross-link agents, the modulus of slide-ring mate-

tials can be decreased dramatically from 0.24 to 0.051 MPa, leading to actuation strains of 12% at 10 V μm\(^{-1}\). However, with these unique molecular designs, further efforts are still required to lower voltage requirements as elastomer thickness used for DEAs remains large. Conventional approaches that can be undertaken to reduce the elastomer moduli include tuning chain entanglement density, reducing cross-linking densities, and increasing molecular weights.

Apart from molecular design strategies, a direct approach to lower the elastic modulus is through thermal softening. At elevated temperatures, increased molecular mobility and disruption of intermolecular interactions lead to a reduction to the elastic moduli of elastomers. Moreover, orientation of dipoles is made easier at higher temperatures leading to a greater contribution to the overall dielectric constant. It has been reported that beyond a certain temperature, random thermal motion of molecules occurs causing the reduction in orientational polarization effect that results in a decline in dielectric constant. Nonetheless, capitalizing on these effects, compared with measurements at ambient conditions, DEAs composed of acrylic, silic-

one, and polyurethane realize higher isometric forces and strains at 100 °C.

**4.1.2. Application Toward iHMI**

Over the years, DEAs have achieved high actuation displacements, moderate bandwidths, silent operation, rapid response, and low power consumption. Supplemented with growing efforts to reduce actuation voltages of DEAs, this cutting-edge technology has found success toward applications within the sphere of iHMI such as haptic interfaces, medical devices, and optics.

**Haptic Interfaces:** Haptic devices have been developed to recreate tactile sensations toward human–machine interactions such as in augmented reality or virtual reality applications; braille displays for the blind and surgical tools. To generate these tactile sensations, soft actuators have been utilized as their mechanical properties are closer to the human skin, providing realistic and natural sensations. While a diverse range of soft actuators have been applied, the favorable performances of DEAs have resulted to promising haptic interfaces designs that have seen success with human subjects.

The bottleneck of adopting DEAs for haptic application is its high driving voltages that may pose a safety hazard if breakdown occurs. Therefore, devices have adopted designs to avoid direct contact with DEAs through an insulating layer or introducing a coupling medium that transmits actuation forces. Also, continuous application of high voltages to sustain deformations tends to reduce the operation lifetime of these devices from leakage currents at high stains. To address this, DEAs with bistable actuation allow actuated shapes to be fixed without voltage application. Ultimately, these challenges can be addressed by lowering the driving voltages of DEAs. Ji et al. designed an untethered haptic device composed of a 18-μm-thick silicone DEA that achieves an in-plane strain of 25% at lower operating voltages of 450 V using onboard electronics and power (Figure 13a). Owing to the DEA soft and reduced thickness, the device stays in intimate contact with the skin, compressing the skin in the off state without causing discomfort to users. Tactile sensations generated upon voltage input cause the DEA to expand, which, in turn, stretches the skin and moves...
Figure 13. Application of DEAs in iHMI. a) Digital photo of low-voltage DEA (top left) and untethered low-voltage DEA with onboard power supply and electronics (top right). Schematic of DEA in-plane strain generated during voltage application, resulting in haptic feedback as the skin gets stretched (bottom). Reproduced with permission. Copyright 2020, Wiley-VCH. b) Schematic of a rolled DEA deforming the skin to induce haptic feedback and the digital image of a fabricated haptic device composed of 2 × 2 array of rolled DEA (top left). Schematic of the components that make up the rolled DEA (top right). Digital image of haptic device tested on human participants. Reproduced with permission. Copyright 2020, Mary Ann Liebert, Inc., Publishers. c) Digital photo of tubular DEA (left) and optical image of its cross section. Digital photo of a pressurized DEA at rest and WITH voltage application (middle). Digital photo of in vitro experiment that tested DEAA under simulated physiological aortic pressure and flow (right). Reproduced under the terms of the Creative Commons CC-BY license. Copyright 2021, The Authors. Published by Wiley-VCH. d) Schematic of the focus tuning principle of the human eye and the soft expansion-tuneable lens controlled by a DEA. Reproduced with permission. Copyright 2018, Mary Ann Liebert, Inc., Publishers. e) Schematic of soft tuneable lens composed two DEAs and segmented electrodes that can be activated independently (top left). Digital photo of the tuneable lens (bottom left). Real-time performance of EOG signals based on up, down, left, and right directions to control planar motions of the lens and double blink to tune the focal length (right). Reproduced with permission. Copyright 2019, Wiley-VCH.
it in the direction normal to the DEA plane. When mounted onto the finger, the haptic device could achieve normal deformations by up to 6 μm and even provide a wide range of feedback between 1 and 500 Hz. Notably, when tested on 11 volunteers, a success rate ranging between 73% and 97% was achieved, as they were tasked to identify five different signals composed of different frequency profiles.\textsuperscript{[257]}

Majority of wearable haptic devices have focused on stimulating the fingers and hand owing to its high tactile sensitivity. However, other regions such as the arm possess lower sensitivity that demands higher forces and displacements from these devices.\textsuperscript{[266]} Several methods have been proposed to amplify the actuator performance for haptic devices including stacking of DEAs and using hydrostatic or rigid couplings.\textsuperscript{[259,260]} Recently, rolled DEAs have been found to achieve axial forces and displacements of 1 N and 1 mm at operational bandwidths and voltages of up to 200 Hz and 1000 V.\textsuperscript{[264]} Matching the ranges required to stimulate major of mechanoreceptors, these rolled DEAs, essentially a stack of DEA layers that is rolled up, were applied to an arm-based haptic device (Figure 13b).\textsuperscript{[258]} While blocked forces were reduced when in contact with skin, human tests achieved a 82.83% and 85.3% success rate of determining actuation locations and directional cues, respectively. Safety considerations of operating at high frequencies were also considered ensuring that temperatures reached only 29.1 °C at 200 Hz.

Braille or haptic displays based on DEAs often transmit tactile information to users through independent out-of-plane deflection of each DEA within an array.\textsuperscript{[209,260]} For practical considerations, this may involve control over a multitude of actuators, making the system complex where each DEA may require a dedicated high-voltage supply or switch. For better control, Mare et al. demonstrated that each DEA within a 4 × 4 array could be driven by integrated flexible high-voltage thin film transistors that allowed switching of DEAs at low voltages.\textsuperscript{[265]} While displays with arrays of evenly spaced DEAs are successful in transmitting numerous tactile cues to users, they remain limited toward generating surface textures of different roughness. To tackle this, rectangular void patterns are introduced to silicone-based DEAs to achieve tuneable surface textures.\textsuperscript{[266]} Furthermore, by tuning the distance between deformed void lines, four different surface textures from smooth to rough were produced. Alternatively, surface textures have been achieved through thickness mode actuation.\textsuperscript{[267]} In this mode, non-active regions deform out-of-plane from shear stresses generated from the actuation of active regions that expands in area. In both cases, the high transparency of these haptic devices allows for future adoption with displays as an external add-on for programmable surface textures.\textsuperscript{[266,267]}

Medical Devices: DEAs are often coined as artificial muscles because of their similar performance with their natural counterparts. For instance, stacking multiple DEAs composed of UV curable acrylic elastomers and ultrathin CNT electrodes achieved a maximum energy density of 19.8 J kg⁻¹, reaching the upper range of natural muscles (0.4–40 J kg⁻¹).\textsuperscript{[211]} Therefore, it is only natural that researchers have explored the development of DEA-driven medical devices such as rehabilitation devices and medical implants that interact with our body.

The recovery of patients with impairments is intrinsically related to the frequency of therapeutic treatments.\textsuperscript{[3]} As such, rehabilitation robots have been gaining interest for providing treatments that are portable at which therapy exercises can be performed at the comfort of the patient’s home. However, current rehabilitation devices such as exoskeletons are found to be rigid and bulky causing discomfort to patients.\textsuperscript{[31]} In contrast, soft rehabilitation devices driven by soft actuators such as DEAs are promising for providing patients with comfortable interactions with machines due to their lightweight and compliant nature.\textsuperscript{[268]}

DEAs have been explored as tremor suppression actuators, providing user comfort and scalability.\textsuperscript{[269–272]} Generally, DEAs suppress tremors through a mechanical approach where an actuated torque is generated in the opposing direction of the tremor-producing muscle torque. To address the low actuation levels of DEAs, actuation requirements are lowered through adopting tremor-active control methods.\textsuperscript{[269,272]} This implies that active voluntary motion is no longer tracked and instead DEAs can be focused on generating actuation torques to suppress tremors. This is corroborated with simulations and scaled experiments of stacked DEAs where tremor-active controls allow the remarkable ability to suppress tremors, bringing DEAs closer toward clinical adoption.\textsuperscript{[269,270]} Also, a hand rehabilitation system that utilizes DEAs as a smart actuator has been reported.\textsuperscript{[273]} In this system, cables and spring-rolled DEAs are used, resembling natural tendons and muscles, respectively, where voltage application causes finger flexion while voltage removal returns the finger to the extended state. Similarly, the complaint nature of DEAs has led to the development of active compression bandages (ACB) that targets venous pooling disorders at the body’s lower extremities.\textsuperscript{[274,270]} Compared with commercial compressive stockings with fixed pressures, ACB designed with DEAs could generate higher pressures that could be modulated based on the applied electric fields.

For medical implants, compliant interactions with the body are highly critical where mismatch in mechanical properties can lead to stress concentrations and redistribution of internal forces that have a negative impact on the body.\textsuperscript{[6]} Furthermore, this mismatch can lead to foreign body reactions that lead to implant failure. While coating implants with soft materials have resulted to reduced inflammation, the adoption of soft DEAs for medical implants proves to be highly promising.\textsuperscript{[276]} A dielectric elastomer aorta (DEAA) composed of a multilayer tubular structure was developed to assist the pulsatile behavior of the heart (Figure 13c).\textsuperscript{[277]} Based on in vitro experiments, the success of DEAA was presented with 5.5% reduction to the work done by the heart. Achieved through electrical control of the DEAA stiffness, aorta deformations can be relieved while recoil forces were enhanced during ventricle systole and diastole, respectively. Also, to address the lesions of heart failure patients, Wu et al. proposed to replace the middle layer of the heart wall with a polyphosphazene (PPZ)-based DEA.\textsuperscript{[278]} Design based on a dipole-conformation-actuated strain model, the PPZ achieved 80% actuated strains at 54 V μm⁻¹ without prestrains. Owing to its high performance, an artificial ventricle composed of PPZ was fabricated and generated similar hydraulic pressure waveforms with human ventricles, indicating their comparable functionalities.

Optical Devices: The sense of sight has been another avenue at which machines have interacted with humans. Particularly, DEAs have been applied toward tuneable lenses that are capable of controlling its optical focus, zoom, and astigmatism.\textsuperscript{[279–282]} These lenses are often composed with a soft lens structure.
and a DEA with annular electrode patterns. Mimicking the human eye, tunability over a large focal length range is achieved when a voltage is applied to cause radial expansion of the DEA that changes the lens radius of curvature (Figure 13d).[279] In addition, Ghilardi et al. designed a tuneable lens that was capable of tuning the defocus or astigmatism through applying different electrical pattern that changes the curvature and causes out-of-plane bending of the lens.[282]

The application of these tuneable lens for iHMI can be represented by Li et al. who utilized electrooculographic (EOG) signals from eye motions to tune the focal length of a soft lens (Figure 13e).[283] Motion of the lens is controlled by surrounding segmented DEAs that are individually actuated from EOG signals. Particularly, planar motions of the lens were determined from up, down, left, and right directions of the eye, while double blink activated focal lengths change. Owing to the DEA characteristic rapid response, easy synchronization between the eye and the lens can be achieved, providing us with greater collaborations between humans and machines.[283]

### 4.2. Electrostatic Actuators

Similar to DEAs, electrostatic actuators are activated based on the generation of electrostatic forces between two opposite charged surfaces. Therefore, electrostatic actuators are also known for its rapid response and low power requirements.[284–286] These actuators omit the use of a soft dielectric layer, minimizing the role of elasticity on actuation with lower restrictions to its force outputs from the low mechanical properties of the elastomer. Electrostatic actuators have therefore replaced the elastomer with either air or a shell filled with dielectric liquid.[285,286] Generally, the driving electrostatic force \( F \) can be expressed in terms of a parallel plate capacitor by Equation (7)

\[
F = \frac{\varepsilon_0 AV^2}{2d^2}
\]

(7)

where \( A \), \( V \), \( \varepsilon_0 \), and \( d \) represent the electrode area, applied voltage, vacuum permittivity, and air gap.[287] However, when the two electrodes are separated by an insulator and a medium such as in hydraulically amplified self-healing electrostatic (HASEL) actuators, the electrostatic force is represented in Equation (8)[285]

\[
F = \frac{1}{2} \varepsilon_{\text{medium}} \varepsilon_0 AV^2 \left( \frac{t_{\text{insulator}}}{t_{\text{medium}}} + \frac{t_{\text{medium}}}{t_{\text{insulator}}} \right)^2
\]

(8)

where \( \varepsilon_{\text{medium}} \) and \( \varepsilon_{\text{insulator}} \) represent the dielectric constant of the medium and insulator, respectively, while \( t_{\text{medium}} \) and \( t_{\text{insulator}} \) represent the thickness of the medium and insulator, respectively. At a critical voltage, electrostatic actuators experience a sudden increase in actuation strains where a mechanical instability causes the two electrodes to be pulled close together. This phenomenon is known as the pull-in instability where the electrostatic force overcomes the elastic restoring force of the structure and this critical voltage is termed the pull-in voltage.

Apart from the typical parallel plate capacitor electrostatic actuation, electrostatic zipping has often been used to achieve large actuation strains and forces.[288–293] Zipping begins at the hinge or edge of the actuator where electric fields are the highest, bringing the electrodes progressively closer. This zipping mechanism was utilized by an electroribbon actuator to enable it to carry 1000 times its weight and contract 99.8% of its length (Figure 14a).[285] The zipping effect was improved by adding a small amount of dielectric liquid at the hinge which allowed higher dielectric breakdowns, lower pull-in voltages, and closing forces to be obtained. Moreover, the dielectric liquid bead was retained by dielectrophoretic forces allowing amplification of zipping effects as it is driven along and is termed dielectrophoretic liquid zipping.

HASEL actuators have recently emerged as a promising electrostatic actuator that have attained high actuation forces and strains, self-healing capability, and self-sensing through coupling electrostatic and hydraulic forces.[284,286] The basic structure of these actuators is composed of a shell with two opposite facing electrodes that is filled with dielectric liquid (Figure 14b).[284,286] When a voltage is applied, electrostatic forces pull the electrode toward each other to cause the liquid between them to be displaced to the surrounding volume. To tune the actuation strain and force output, the size of the electrodes with respect to the shell should be carefully selected at which a larger electrode size would generate high actuation strains but low forces. Generally, the operation of the HASEL actuator can be classified under three regimes. At low voltages, small actuation strains are observed as it experiences a resistive force from the shell stiffness and the hydrostatic pressure within the dielectric liquid. At the pull-in voltage, a drastic increase in actuation strains is displayed. After which, further voltage application led to a gradual increase in actuation strains. Based on this concept, various configuration of HASEL actuators have been designed, including the donut, linear contracting HASEL (Peano-HASEL), and high strain Peano-HASEL.[284,289,293]

By introducing a liquid dielectric, hydraulic amplification allows larger forces and displacements to be achieved for imparting tactile sensations.[294,295] Leveraging on this approach, a large-area haptic display was designed to impart dynamic tactile feedbacks over a large area of skin (Figure 14c).[294] By targeting a larger area, better mechanical transmission to the user can be achieved at which directional tactile motion was successfully recognized by participants. To achieve higher displacements, Leroy et al. designed a hydraulic amplified taxel (HAXEL) with a central interaction membrane composed of an elastomer (Figure 14d).[295] Surrounding this is an inextensible shell that is covered with electrodes, analogous to HASEL actuators. As such, when electrostatic zipping is activated, large out-of-plane displacements can be achieved as the dielectric liquid is directed to the stretchable interaction membrane. Furthermore, by segmenting the electrodes into separate parts, in plane motions can be attained to impart shear forces to users, broadening the range of tactile sensations offered. Nonetheless, operational voltages of these electrostatic actuators range from several hundred volts up to several kilovolts and should be lowered to provide greater safety to users.

A direct way to lower the operational voltage of electrostatic actuators is through the reduction of the thickness of the film coated with electrodes.[296,297] By doing so, the bending rigidity of the film that resists against the electrostatic forces is reduced. Demonstrated with an electrostatic actuator with two films separated by an arch-shaped gap, changing the thickness from 127 to
25 μm minimized the activating voltage from 700 to 200 V.\[296\] Similarly, Hartmann et al. designed a soft subkilovolt tuneable lens based on electrostatic zipping.\[297\] This was achieved through adhering to theoretical models developed which comprised tuning actuator design parameters including the dielectric constant and thickness of the plastic foil coated with electrodes along with the opening angle. For instance, through designing lenses with smaller electrode opening angles, it could be operated at lower voltages.\[297\] To reduce the pull-in voltage of HASEL actuators, a model guided approach that highlights the importance of selecting the optimum electrode size and fluid dielectric constant.\[298\] For example, the pull-in voltage is first reduced with an increase in the electrode size as the outer shell is stiffens at a lower rate compared with the electrode area. However, beyond certain electrode size, the inverse effect takes place as the stiffness of the outer shell dominates. On the contrary, unlike efforts to improve the performance of DEAs and ferroelectric polymer actuators by increasing the dielectric constant, this is only true for HASEL actuator till electrode displacement effects dominate.\[298\]

4.3. Ferroelectric Polymer Actuators

Ferroelectric polymers are semicrystalline polymers that have a noncentrosymmetric structure with polar side group. When
an electric field is applied to these polymers, the dipoles align with the field at which a thermodynamic stable polarization state is maintained. It is only when an opposite electric field is applied or after heating above the material’s curie temperature the induced polarization is lost. Generally, actuation strains achieved by ferroelectric polymers are governed by the electrostrictive effects that originate from the change in dipole density within the material under an applied electric field (Figure 15a). The actuation or electrostrictive strain ($S_3$) along the thickness direction for linear dielectrics can be expressed from Equation (9)

$$S_3 = Q P^2 = Q(\varepsilon_r - 1)^2 \varepsilon_0^2 E^2 \quad (9)$$

where $Q$, $P$, $\varepsilon_0$, $\varepsilon_r$, and $E$ represent the electrostrictive coefficient, polarization, vacuum permittivity, dielectric constant, and electric field, respectively. Relaxor ferroelectric polymers (RFP), a subcategory of ferroelectric polymers, have been introduced at which large ferroelectric domains are reduced to nanopolar domains. This is achieved through the introduction of defects that destroy the stability of ferroelectric domains, lowering and broadening the ferroelectric to paraelectric phase transitions. As such, RFP often have lower hysteresis and coercive fields that translate to decreased energy losses and actuation voltages. To induce these defects, high energy irradiation or the introduction of bulky comonomer such as chlorotrifluoroethylene (CFE), chlorotrifluoroethylene (CTFE), and hexafluoropropylene (HFP) to PVDF-TrFE have been demonstrated. More recently, Zhang et al. designed a RFP that converted CTFE groups of PVDF-CTFE to less bulky CH═CF bonds that acted as molecular scale defects (Figure 15b). The resultant material, PVDF-DB, was strained by 500% to induce larger crystallinity that was largely in the relaxor ferroelectric phase followed by its fabrication into a cantilever actuator. Owing to its enhanced dielectric constants of 75 at 10 Hz, the actuator could be activated at low electric fields of 30 MV m$^{-1}$ (210 V) and achieve a maximum electrostrictive strain of 13.4% at 275 MV m$^{-1}$.
Based on Equation (9), a high dielectric constant is favorable to lower the driving voltage of these ferroelectric polymers actuators. Similar to DEAs, this can be achieved through the addition of high dielectric ceramic powders and conductive particles to ferroelectric polymers.\(^{1310-1316}\) Also, plasticizers have been added into ferroelectric polymers to enhance dielectric constants through interfacial polarization effects.\(^{1317-1319}\) However, while these approaches have shown to increase the dielectric constant and electrostrictive strain, this is often at the expense of a reduced dielectric breakdown strength.\(^{1321-1325,1318}\) To address this, a thin conductive layer, AgNW was sandwiched between two RFP layers, PVDF-TrFE-CTFE (Figure 15c).\(^{1304}\) Through this approach, charge accumulation at the AgNW/PVDF-TrFE-CTFE interface results in an enhancement to the dielectric constant over a broad frequency range, for instance, from 34.5 to 65.5 at 1 kHz. The distance between two conductive components is lengthened compared with ferroelectric polymers with dispersed fillers, requiring a higher electric field for dielectric breakdown. Consequently, unimorph actuators could operate at a low voltage of 250 V (25 V \(\mu\)m\(^{-1}\)) to generate a force of 1.92 mN, double that of pristine PVDF-TrFE-CTFE.\(^{1804}\)

Owing to the favorable characteristics of ferroelectric actuators, these actuators have been utilized for iHMI applications such as haptics or medical devices.\(^{1320-1326}\) Localized tactile feedbacks have been realized through fretting-vibrotactile displays that are composed of a RFP and a top electrode while separated from the bottom electrode on a substrate by a spacer (Figure 15d).\(^{1320}\) To activate the display, normal pressures from the human finger push the RFP to contact the bottom electrode upon which a locally concentrated electric field is generated below the finger to induce localized electrostrictive strains. As such, localized multitouch feedbacks can be realized locally with favorable performances under static pressures. Furthermore, the display can be easily integrated with various sensors and can be fabricated on the large scale.\(^{1320}\) Owing to the high flexibility and fast response of ferroelectric polymer actuators, these devices have been considered for steerable guide wires used for minimally invasive surgeries.\(^{1326}\) To achieve steerability, Ganet et al. modeled and designed PVDF-TrFE-CTFE tube actuators with segmented electrodes that allowed two degrees of freedom (Figure 15e).\(^{1326}\) Notably, the use of PVDF-TrFE-CTFE enabled the device to be configured into a force sensor as well, providing greater functionalities as a guide wire. The prospect of using these actuators toward medical devices for iHMI was further established by investigations that showed no changes to actuation performances under \(\beta\) irradiation sterilization procedure.\(^{1327}\)

### 4.4. Plasticized Gel Actuators

PVC has often been seen as electrically inactive though this changed with the introduction of plasticizers that causes the formation of PVC gels. Through extensive research on these materials, these PVC gel actuators are found to display fast actuation response and large deformations at medium range voltages (typically < 600 V) together with flexibility and durability, making it highly attractive for application toward iHMI.\(^{1328}\) While PVC gel actuators currently dominate these class of actuators, an effort to reduce the halogen content that may be harmful for the human body and environment has led to plasticized thermoplastic polymers to be recently proposed as an alternative.\(^{329,1330}\)

These PVC gel actuators share similar configuration to DEAs, at which the polymeric material is sandwiched between two electrodes. However, after voltage application, PVC gel actuators differ significantly from DEAs where asymmetrical deformations occur instead of expanding in area.\(^{1328,1331}\) The driving mechanism behind these deformations has been attributed to electrical charge injection from cathode to anode, migration of plasticizer, and the Maxwell force from the applied electric field.\(^{1328,1332,1333}\) PVC gels are loosely connected through physical cross-links with large mesh sizes in the network, allowing plasticizer molecules to be packed in the spaces between the chains. Under an electric field, the dipole on PVC chains rotates in the direction of the anode, while the plasticizer molecules become charged (Figure 16a). Subsequently, plasticizer molecules migrate toward the anode and physical cross-linking points of PVC chains may be pulled accordingly to cause actuation where the gel creeps on the anode.\(^{332,1333}\) Nonetheless, with the rediscovery of plasticized actuators further efforts are required to elucidate the deformation mechanism.\(^{1328}\)

The type and amount of plasticizers within PVC gel actuators play a critical role toward achieving a low actuation voltage.\(^{1334-1336}\) For instance, higher content of dibutyl adipate plasticizers showed higher actuation displacements of PVC gel actuator at lower voltages due to a reduction of elastic modulus.\(^{1334}\) Similarly, when tested against four different plasticizers (bis(2-ethylhexyl)phthalate, di-n-butylphthalate, bis(2-ethylhexyl) adipate, and tris(2-ethylhexyl)trimellitate), the influence on the mechanical properties showed a strong influence on the actuation.\(^{1335}\) While this work did not indicate a dependence on the dielectric constant of the plasticized film, Ali et al. showed that higher dielectric constants due to easier dipole reorientation led to favorable electromechanical performance.\(^{1336}\) Particularly, smaller molecular weights of these plasticizers provided less restrictions on molecular motions that generate higher dielectric constants. However, the addition of high contents of plasticizers may reduce the elastic restoring force and affect dynamic performances.\(^{1337}\) Additives such as GO that display electrophoretic mobility under electric fields can be introduced (Figure 16b).\(^{1338}\) Attributed to its polar functionalities of GO, electrophoretic mobility was enabled to allow greater pulling of PVC cross-link points toward the anode generating larger creeping motion. Ionic liquid additives have also been considered to improve the low-voltage performance through increasing Maxwell stresses from adsorption capacitance contributions.\(^{1339}\) Moving forward, it will be critical to investigate other factors that can improve the actuation performance and lower its driving voltage.

One of the unique features of PVC gel actuators is the unnecessary need for compliant electrodes, allowing higher durability and easier manufacturing of devices. For instance, a nonpetroleum-based and eco-friendly PVC/acetyl tributyl citrate gel was easily formed into a wave shape for maximum displacements (Figure 16c).\(^{1337}\) This structure was applied for haptics devices where an optimal ratio of PVC and acetyl tributyl citrate plasticizers could generate vibrotactile forces that exceeded the human vibrational threshold over a broad range of frequency (1–300 Hz) at 1 kV. Further improvements to these vibrational forces based on similar wave shapes were achieved with the introduction of silicon dioxide nanoparticles (SiO\(_2\)).\(^{1340}\) With SiO\(_2\),
lower leakage currents allowed for efficient energy consumption during actuation at which the capability of the improved device was successful to induce vibrotactile sensations on human participants. However, these wave-shape structures may not be suitable for wearable haptic devices as frictional forces between the ridges of PVC gel actuator and the electrodes may lead to detachments after prolonged usage. Instead, for improved wearability, PVC gel actuators were fabricated into a knitted fabric (Figure 16d). In the design of a knitted PVC gel actuator, the dielectric constant, mechanical properties, and length of PVC strings are considered. Furthermore, the optimized device showed high stability of only a change of 7% (reduced to 0.415 G) in vibrational amplitude after continuous operation (1.5 kV at 90 Hz) for a week. When tested on participants, vibrations generated by knitted PVC gel actuator provided clear perceptions.

Over the years, PVC gel actuators have interacted with humans through providing medical support. Li et al. designed a wearable assist-wear for hip joint support (Figure 16e). Basically, the actuator is activated to provide an assistive contraction-expansion output force that generates a torque around the joint for reduced burden on the leg muscles. At 400 V, the PVC gel actuator-based assist-wear could achieve high forces of 94 N on the hip joint and had lower power consumptions than pneumatic systems. In combination with sensor and control systems, electromyograms showed a reduced muscular burden of the lower limbs. However, in various examples of the adoption of PVC gel actuators are presented, further improvements on the fabrication methods that mostly rely on solution casting are required for large-scale commercialization.

Figure 16. Strategies to lower the voltage requirements of PVC gel actuators and its application in iHMI. a) Optical image of PVC gel actuation before and after actuation (top) Schematic of working principle of PVC gel actuation (bottom). Reproduced with permission. Copyright 2011, ACS Publications. b) Digital image of bending motion of PVC gel actuator before and after actuation (top). Schematic of working principle of PVC gel actuator enhanced with graphene oxide (bottom). Reproduced under the terms of the Creative Commons CC-BY license. Copyright 2019, The Author(s). Published by Springer Nature. c) Schematic of working principle of wave-shaped PVC gel actuator for haptic devices. Reproduced with permission. Copyright 2016, IOP Publishing Ltd. d) Schematic of fabrication process of knitted PVC gel actuator. Reproduced with permission. Copyright 2019, IEEE. e) Schematic overview of PVC gel actuator-based system for wearable assist-wear composed of multilayered PVC gel actuator (left), controller and power supply (top right), and Flexiforce sensor (bottom right). Reproduced with permission. Copyright 2017, IOP Publishing Ltd.
5. Challenges and Outlook

ECAs can be driven at very low voltages and are promising for a variety of applications, especially in soft robotics, tactile displays, and haptic feedback sensing. Though tremendous efforts have been devoted to ECAs, their actuation strain, stress, speed, and durability are still lower than other actuators and need to be improved at a large degree. Many kinds of active materials and electrolytes have been explored for ECAs, but the exact actuation mechanism for the active material and device is not well investigated, which limits the design of high-performance ECAs. To explore the mechanism especially for new material systems, it is necessary to monitoring the dimensional and microscale structural evolution of active electrodes during charge/discharge in two/three electrode cells by characterization techniques such as in situ optical microscope/SEM/XRD/Raman. The strain of active electrodes is still limited to the available surface area for ion storage, and are expected to be increased by using new nanomaterials of high electrical conductivity and specific surface area, such as graphynes,[344,345] MXenes,[115,346–349]

Table 3. Comparison of key performances of electronic EAP-based soft electrical actuators.

| Material/device type               | Electrode              | Device type | Dielectric thickness [μm] | Dielectric constant [MPa] | Elastic modulus [MPa] | Actuation strain/displacement | Actuation force [mN] | Ref. |
|-----------------------------------|------------------------|-------------|---------------------------|---------------------------|-----------------------|-----------------------------|-----------------------|------|
| Nitrile-modified silicone          | Carbon black powder    | DEA         | 35                        | 18 @ 10^4 Hz              | 0.462                 | 4% @ 200 V^a                 | —                     | [203]|
| Polyurethane acrylate             | Carbon grease          | DEA         | 430                       | 5.52 @ 10^3 Hz            | 0.337                 | 66.7% @ 6.4 kV^a              | —                     | [209]|
| Silicone                           | Carbon black-elastomer composite | DEA     | 3                         | —                         | —                     | 7.5% @ 245 V^b               | —                     | [214]|
| PDMS                              | Gold                   | DEA^6       | 0.2                       | —                         | 1.685                 | 1.43 m^-1 @ 12 V^c            | —                     | [215]|
| TiO2-PDMS/dimethylsilicone oil    | Carbon black powder    | DEA         | 100                       | 3.5 @ 5 x 10^4 Hz         | 0.095                 | 13% @ 3 kV^d                  | —                     | [222]|
| Silicone/liquid metal nanodroplets| EGaIn                  | DEA^6       | 264.2^j                    | 7.62 @ 10^3 Hz            | 1.98                  | —                           | 21.16 @ 14 kV^e         | [227]|
| Bottlebrush elastomer             | Carbon grease          | DEA         | 440                       | 2.94 @ 10^4 Hz^f          | 0.022^f               | 300% @ 3.5 kV^g               | —                     | [246]|
| Slide ring gel                    | Powdered graphite      | DEA         | 1000                      | 9.28 @ 10^3 Hz            | 0.051                 | 12% @ 10 kV^i                 | —                     | [248]|
| Circular planar HASEL actuator    | Polyacryl-amide/LiCl   | Electrostatic | —                         | —                         | —                     | 46% @ 11 kV ^f                | —                     | [284]|
| Electroribbon actuator            | Steel                  | Electrostatic | 130 (2 layer)             | 4.62^k                   | —                     | 99.84% @ 10 kV^i              | 12900 @ 10 kV^j       | [285]|
| Quadrant donut HASEL actuator     | Polyacryl-amide/LiCl   | Electrostatic | 18                        | —                         | —                     | 118% @ 12 kV^l                | 60 000 @ 12 kV^m      | [356]|
| Polyimide                         | Gold                   | Electrostatic | 25                        | —                         | —                     | 2.7 mm @ 500 V^o               | —                     | [296]|
| Peano-HASEL                       | Polyacryl-amide/LiCl   | Electrostatic | 18                        | 3.2^l (2.2)^m            | 2500                  | 15% @ 8 kV^p                  | 45 000 @ 8 kV^q       | [357]|
| P(VDF-TrFE-CTFE)/AgNW interlayer  | ITO-coated PET/PEDOT/PSS | Ferroelectric | 10                        | 65.5 @ 10^3 Hz            | —                     | —                           | 2 @ 250 V^r           | [304]|
| P(VDF-DB-500)                     | Gold                   | Ferroelectric | 7                         | 51.61 @ 10^3 Hz           | 320                   | 13.4% @ 1.9 kV^s              | —                     | [305]|
| P(VDF-TrFE-CTFE)/carbon black     | Silver                 | Ferroelectric | 8                         | 71 @ 10^3 Hz              | —                     | 248.1 mm @ 90 V^t              | —                     | [312]|
| P(VDF-TrFE-CTFE)/ diisononyl phthalate | Gold              | Ferroelectric | 80                        | 437 @ 10^-1 Hz^f          | 38.3^g               | 0.521@ 800 V^h                | —                     | [317]|
| TPU/DBA                           | Metal foil             | Plasticized gel | 350                       | 1000 @ 1 Hz^i             | 0.00237              | 13.22 mm @ 1.04 kV^p          | 1.399 @ 700 V^q       | [330]|
| PVC/DBA gel                       | —                      | Plasticized gel | 500                       | —                         | 300 mm @ 1 kV^q       | 4 @ 800 V^r                  | —                     | [333]|
| PVC/DBA gel                       | Stainless mesh/foil    | Plasticized gel | 300–1500                  | 2096 @ 1 Hz^i             | —                     | 0.548 mm @ 1 kV mm^-1^i       | 0.85 @ 1 kV^j         | [334]|
| PVC/GO gel                        | Copper tape/stainless steel mesh | Plasticized gel | 1000                      | —                         | 165 mm @ 500 V^k      | 28 @ 1.5 kV^l                | —                     | [338]|

—a)Lateral actuation strain; ^b)—, Not available; ^c)Actuated area strain; ^d)Unimorph cantilever configuration; ^e)Actuated curvature; ^f)Estimated values; ^g)Apparent shear modulus; ^h)Vertical displacement; ^i)Measured frequency not available; ^j)Actuation/recurrence; ^k)Linear strain; ^l)Dielectric constant for Cargill Envirotex FR3 liquid dielectric; ^m)Dielectric constant of BOPP; ^n)Electrostrictive strain; ^o)Vibrational Amplitude; ^p)Bending displacement; ^q)Creep displacement.
and covalent organic frameworks. The aggregation and orientation of anisotropic active materials in electrodes should be highly noted and well controlled for effective actuation. The strain of ECAs is usually determined by the strain difference between the cathode and anode and could be enhanced by using electrolytes with large size difference between the (solvated) anion and cation as well as designing new configurations such as asymmetric structure with different active materials in the anode and cathode. The stress of ECAs is generally low due to the use of soft gel electrolytes and soft electrodes and could be enhanced by improving their modulus while maintaining an effective ion diffusion network in both. The operation of ECAs is normally conducted at frequencies less than 10 Hz due to the slow electrochemical processes in both the electrodes and electrolyte membrane. It is essential to explore electrolyte membranes with superior ionic conductivity and active electrodes with short ion diffusion distance for the ECAs to deliver fast response and large displacement/strain simultaneously.

The durability of ECAs under repetitive bending and long-time operation is another issue/challenge, which are usually limited by the cracking of electrodes, delamination between the electrode and electrolyte, and the evaporation/leakage of electrolyte. The cracking of active electrodes can be eliminated by toughening them via addition of polymer binders or fibrous nanomaterials. The delamination issue can be avoided by enhancing the electrode–electrolyte interface interaction via covalent/noncovalent functionalization of the active electrodes or introducing strong chemical bonding at the interface. The evaporation/leakage of electrolytes can be inhibited by using nonvolatile or highly hygroscopic solvents/electrolytes as well as by utilizing high-barrier, thin, and soft encapsulations, which can also avoid the influence of humidity change on the actuation performance. Hydrophobic packaging with a thin barrier layer (e.g., Al, SiO₂, and Al₂O₃) or filled with 2D nanosheets (e.g., organic clay, graphene, and boron nitride) can be considered, with their thickness and modulus being minimized to reduce their constraining of the actuation deformation. Currently, the fabrication process of ECAs is a bit complicated and the device shape is monotonous. It will be interesting to explore novel fabrication technologies such as 3D printing or patterning to fabricate 3D/patterned ECAs with new geometries, which can generate complex/customized actuation modes, such as multiple freedom motions and torsions. It may be promising to design biocompatible ECAs from biocompatible electrodes and electrolytes for biomedical applications such as robotic surgery. It will be necessary to develop response models of ECAs under different conditions (e.g., potential, frequency, and humidity) by analysis/numerical modeling and analysis for controlling their actuation as required. It is also encouraged to construct multifunctional systems integrated with ECAs for practical applications. Electrochemical–mechanical–optical coupling is one example, which is capable of simultaneously deforming and changing color/reflecting infrared light (Figure 17).

Soft ETAs have a dry and relatively simple structure that is easy to fabricate and can generate relatively large actuation strain/stress in a wide temperature range under low driving voltages. However, challenges still remain for those ETAs, including slow response and recovery rate, high temperature during working, low electrical-to-mechanical transduction efficiency, possible delamination between layers, unable to keep deformation when power is off, and the integration of power and sensors for remote control and interaction. Using an efficient electrical heating layer with a high specific conductivity and low specific capacity is highly desirable to improve the response rate. Au, Ag, and Cu are highly conductive but heavy and expensive. Metallic Al with a higher specific electrical conductivity and much lower cost could be an alternative but may suffer from thermal oxidation in the air. Therefore, effectively protected Al with a higher stability is expected for future investigations. Highly conductive ultrathin films of CNT and graphene can also be considered due to its low density, high stability, and high transparency, which are desirable for designing lightweight/transparent ETAs. The high temperature of ETAs during operation may be over the safe temperature limits for human skin/organs and is undesirable for tactile/wearable/implant applications. Using thermally active materials with very high thermal expansion/contraction efficient (or difference) is expected to reduce the operation temperature and driving voltage of ETAs. Sandwiching a flexible conductor heater between a highly thermal shrinkage film (e.g., hydrogel films) and a highly thermal expansion film (e.g., phase-change material, phase-change-material-filled lms) and a highly thermal expansion film (e.g., phase-change material, phase-change-material-filled lms) and a highly thermal expansion film (e.g., phase-change material, phase-change-material-filled lms

![Figure 17](https://example.com/figure17.png)

**Figure 17.** Future directions for developing high-performance soft electrical actuators.
polymer) to construct a multilayered ETA would maximize the expansion difference and generate a large bending/curling of the device. Fast recovery could be achieved by improving the thermal dissipation by enlarging the surface emissivity (especially the out surface of the metal layer) and surface area of both layers. Improving the thermal conductivity of the insulating layers by filling efficient thermal conductive materials (e.g., carbon fibers, large graphene/boron nitride nanosheets, and long CNTs) may increase both the response rate and cooling rate via efficient thermal conduction. The electrical-to-mechanical energy efficiency could be enhanced by reducing the operation temperature and harvesting the dissipated heat by coupling with thermoelectric generators. It will also be necessary to develop actuation and recovery models of ETAs for controlling their actuation under different conditions (e.g., size, temperature, potential, and frequency). Integrating soft ETAs with flexible batteries, micro sensors, and wireless control circuits into smart soft robotics may provide a convenient remote control for various applications.

As for other soft electrical actuators such as dielectric, electrostatic, ferroelectric, and plasticized gel ones, the driving voltage has yet to be lowered, preferably below 10 V. The various strategies adopted have significantly reduced the voltage to safer levels below 1000 V (hundreds of volts). The prospect of reaching these lower voltages is shown by adopting nanoscale technologies adopted have significantly reduced the voltage to safer levels below 1000 V (hundreds of volts). The prospect of reaching these lower voltages is shown by adopting nanoscale technologies. Alternatively, to alleviate the need for nanometer-scale thickness, a combination of material strategies can be adopted, such as thinning down films with high dielectric constants or low elastic modulus. Concurrently, to easily handle and realize higher actuator performances of these thin devise, multilayer designs can be used. Owing to the wide bandwidth, rapid response times, high forces, rapid response, and energy densities of these actuators, there has been much success to its adoption for iHMI. However, further improvement to the control of these actuators can be implemented through the development and adoption of analytical models that address hysteresis behaviors from viscoelastic and polarization effects. Moving forward, with these electroactive actuators having similar configurations, the various mechanisms can be combined for higher actuation performances. For instance, Li et al. coupled DEAs with electrostatic zipping actuation to achieve an actuator with faster response times by fivefold and 87% larger stable force outputs compared with DEAs operating alone. Similarly, the combination of these two mechanisms has been applied toward tactile displays at which an improvement to displacements and blocking forces by 19.1% and 26%, respectively, was demonstrated. While these works operate at high voltages (>1 kV), the potential of a combined mechanisms is promising especially if the strategies for low driving voltage is adopted.

Acknowledgements

S.C. and M.W.M.T. contributed equally to this work. The authors acknowledged the funding provided by Ministry of Education under project no. 2020-TI-001-165 and the 6th NRF-ANR Joint Grant, NRF2020-NRF-ANR102 MEACT.

Conflict of Interest

The authors declare no conflict of interest.

Keywords

conductive materials, human–machine interaction, nanomaterials, polymers, soft electrical actuators

Received: April 22, 2021
Revised: May 30, 2021
Published online: July 11, 2021

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Adv. Intell. Syst. 2022, 4, 2100075

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