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Dielectric elastomer artificial muscle: materials innovations and device explorations

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CONSPECTUS: Creating an artificial muscle has been one of the grand challenges of science and engineering. The invention of such a flexible, versatile, and power efficient actuator opens the gate for a new generation of lightweight, highly efficient, and multifunctional robotics. Many current artificial muscle technologies enable low-power mobile actuators, robots that mimic efficient and natural forms of motion, autonomous robots and sensors, and lightweight wearable technologies. They also have serious applications in biomedical devices, where biocompatibility – from a chemical, flexibility, and force perspective – is crucial. It remains unknown which material will ultimately form the ideal artificial muscle. Anything from shape memory alloys (SMAs) to pneumatics to electroactive polymers (EAPs) realize core aspects of the artificial muscle goal. Among them, EAPs most resemble their biological counterparts, and they encompass both ion-infusion and electric field based actuation mechanisms. Some of the most investigated EAPs are dielectric elastomers (DEs), whose large strains, fracture toughness, and power-to-weight ratios compare favorably with natural muscle.

Although dielectric elastomer actuators (DEAs) only entered the artificial muscle conversation in the last 20 years, significant technological progress has reflected their high potential. Research has focused on solving the core issues surrounding DEAs, which include improving their operational ranges with regards to temperature and voltage, adding new functionality to the materials, and improving the reliability of the components they depend on. Mechanisms designed to utilize their large-strain actuation and low stiffness has also attracted attention. This Account covers important research by our group and others in various avenues such as decreasing viscoelastic losses in typical DE materials, increasing their dielectric constant, and countering electromechanical instability. We also discuss variable stiffness polymers, specifically bistable electroactive polymers, which, notably, open DEAs to structural applications typically unattainable for soft-actuator technologies. Furthermore, we explore advancements related to highly compliant and transparent electrodes—a crucial component of DEAs capable of achieving high actuation strain. We then cover noteworthy applications, including several novel devices for soft robotics and microfluidics, and how those applications fit within other major developments in the field. Finally, we conclude with a discussion of the remaining challenges facing current DEA technology and speculate on research directions that may further advance DE-based artificial muscles as a whole. This Account serves as a stepping stone into the field of EAPs, which, through the work of researchers worldwide, are positioned as a potential challenger to conventional actuator technologies.

1. INTRODUCTION

Nature has toiled for millions of years to perfect natural muscle across a stunning array of uses and environments. Natural muscles have many desirable properties, including adaptable stiffness, large stress production (~0.35 MPa), fast response speeds, self-healing over billions of work cycles, and

energy storage and generation.[1] Developing a biomimetic, muscle-like technology would revolutionize soft robotics and human assist devices. In the early 1990's, a group of new, electroactive polymers (EAPs) emerged which exhibit substantial deformation in response to applied voltage.[2–4] Their similarity to biological muscles earned them the moniker "artificial muscles".

EAPs can be divided based on their actuation mechanisms: ionic EAPs (activated by ionic diffusions) and field activated EAPs (driven by the charge interactions).[5] Ionic EAPs consist of ionic polymer-metal composites (IPMCs), ionic gels, carbon nanotubes (CNTs), and conductive polymers, while field activated EAPs include ferroelectric polymers, polymer electrets, electrostrictive polymers, and dielectric elastomers (DEs).[5] Among EAPs, dielectric elastomer actuators (DEAs) are emerging as one of the leading technologies with regards to large strains and stresses, fast response speeds, long lifetimes, good reliability, and high efficiencies.[6,7] We anticipate that DE based, soft, and biologically inspired actuators will replace conventional motors and gearboxes in applications where mechanical compliancy and compact sizes are essential.

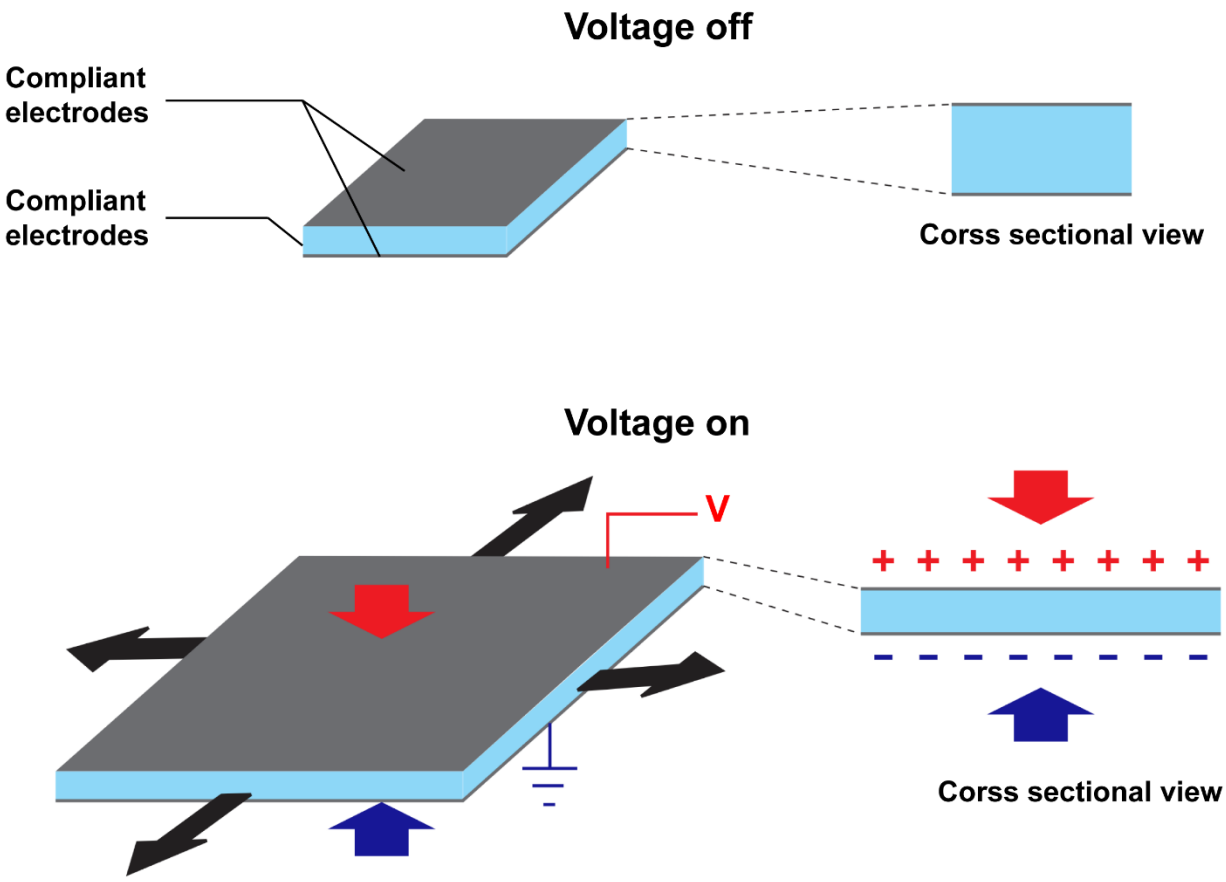

Figure 1. Operating principle of a DEA. A DEA consists of an elastomer sandwiched between two compliant electrodes. When a voltage is applied, charge separation occurs and induces an electrostatic pressure on the film. This deforms the film increasing its area and reduces its thickness.

DEAs are essentially deformable capacitors. They consist of a thin elastomeric film coated on both sides with compliant electrodes (Figure 1). When an electric field is applied, the electrostatic attraction between opposite charges generates a pressure on the film. This pressure forces the film to contract in thickness and expand in area, and thus, electrical energy is converted into mechanical work. The electrostatic pressure is given by the Maxwell stress[5] (p)

$$p = \varepsilon_r \varepsilon_o E^2 \quad (1$$

where ε_r is the dielectric constant of the DE, ε_o is the permittivity of vacuum, and E is the applied electric field. For strains below 10%, the change in thickness (s_z) can be defined by linear-elasticity and free boundary approximations[5] as

$$s_z = -\frac{p}{Y} - -\frac{\varepsilon_r \varepsilon_o E^2}{Y} = -\frac{\varepsilon_r \varepsilon_o (V/Z)^2}{Y} \quad (2$$

where Y is the apparent elastic modulus of the DE at the actuated strain. A typical DE film with ε_r of 3.0, Y of 1 MPa, and E of 100 MV/m generates a pressure p = 0.26 MPa and strain s_z = -26%. Therefore, for a manageable DE film 50 μm thick, the driving voltage is 3.7 kV. Thus, reducing the driving voltage via larger ε_r or thinner films has been an important research objective. Developing a highly compliant electrode material to reliably drive the DE films to large strains is another critical task.

This Account discusses, based on our own experience and perceptions, the key advances made in the development of new dielectric elastomers and stretchable electrodes for EAP applications. The discussion will not only cover material innovations, configurational designs, advanced

manufacturing of devices, but it will also go over representative applications of DEAs in the field of soft robotics, wearable electronics, and human-interface devices. It will conclude with a discussion of the remaining hurdles and prospects.

2. DIELECTRIC ELASTOMER MATERIALS

In 1776, Alessandro Volta credited Felice Fontana as the first to note volume changes in a Leyden jar under electric stimulation, marking the discovery that a solid material could be deformed by an electric field.[8] Later, Wilhelm Roentgen observed shape changes during charging experiments on natural rubber strips,[9] and in 1899, M.P. Sacerdote formulated the strain response to electric field activation of "electrically stretchable materials".[10] SRI International pioneered the more recent investigation into dielectric elastomer actuators in the late 1990's.[6] They reported that out of a variety of commercial elastomers, acrylic and silicone elastomers stood out for their large actuation strain, large Maxwell stress, high actuation speed, and high energy density.[6]

The unprecedented performance of these DEAs led to a scientific and technological revolution in the field of EAPs. However, their individual physical constraints and the high electric fields (~100 MV/m) required to drive them limited the widespread use of DEAs. Countering these limitations requires improving the intrinsic properties of the DEs, especially the dielectric constant and elastic stress-strain response.

2.1.Decreasing Viscoelasticity of VHB Elastomers

VHB acrylic elastomers from 3M^{TM} exhibit exceptionally large area strain, exceeding 380%, and a high Maxwell stress of 7.2 MPa (Figure 2a).[6] Furthermore, VHB's 3.4 MJ/m^3 theoretical energy density far exceeds the 0.07 MJ/m^3 of human skeletal muscle.[6] However, VHB acrylic elastomers

suffer from severe viscoelastic effects, which result in slow response speed and long-term relaxation.[11,12]

The viscoeloasticity stems from limited rotational freedom of the polymer chains, which significantly affects the actuation response of DEs because of the viscous impedance at high frequencies.[13] Zhang et al. reported that the addition of dibutoxyethoxyethyl formal (DBEF) plasticizer into VHB promoted chain rotation, which reduces viscoelastic loss. It also lowers the elastic modulus and broadens the working temperature range of the polymer.[14] Notably, the plasticized elastomer (40 wt% DBEF) can be actuated at -40 °C and generate 215% area expansion (Figure 2b).

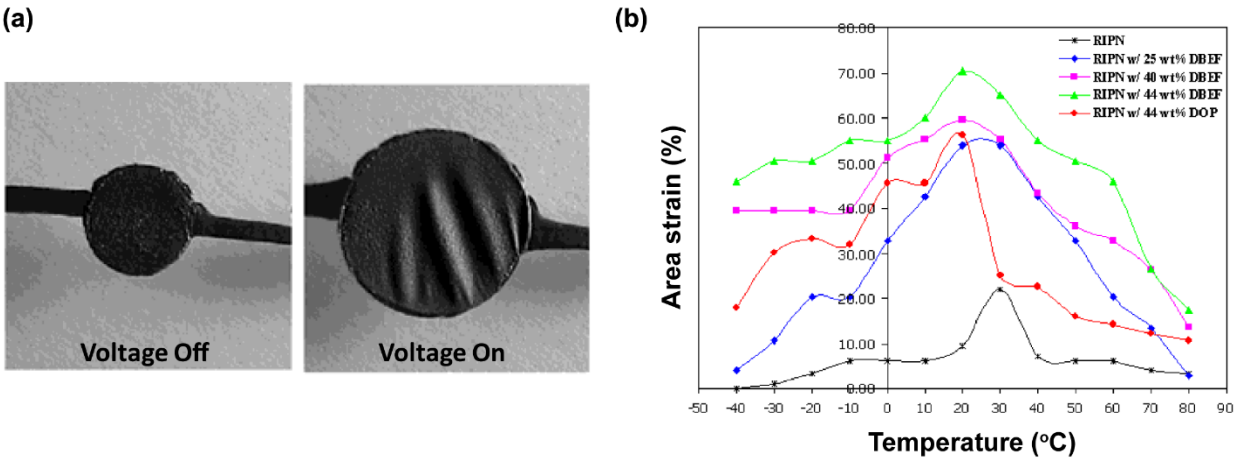

Figure 2. Actuation performance of VHB acrylic elastomers. **(a)** Actuated areal expansion of a prestretched film. Reprinted with permission from ref 7. Copyright 2000 Advanced Materials. **(b)** Comparison of temperature-dependent strain response between untreated (RIPN) and plasticizer-treated VHB films at 5 kV constant voltage. Reprinted with permission from ref 14. Copyright 2010 Polymer International.

2.2.Freestanding Elastomers with Enhanced Properties

Electromechanical instability (EMI), or pull-in effect, is one of the main failure modes of DEs (Figure 3).[15] It occurs when Maxwell stress above a certain threshold triggers a positive feedback loop of film compression until it breaks down. Suo et al. quantitatively modeled the stress-strain behavior of a typical elastomer – shown by the thin, red curve in Figure 3 – and how the soft nature of elastomers results in a long stress-strain plateau leading to EMI.[5,16] Prestretching is one method that suppresses EMI by modifying the polymer's elastic stress-strain response, shown by the red, dashed curve.[5] However, traditional prestretch techniques require rigid structures to maintain tension, adding additional load and reducing DEA work density and power to mass ratio. Additionally, the interface between the rigid frame and the soft elastomer is a stress concentrator. Thus, alternative freestanding DEs that capture the benefits of prestretch without the framing requirements are highly desirable.

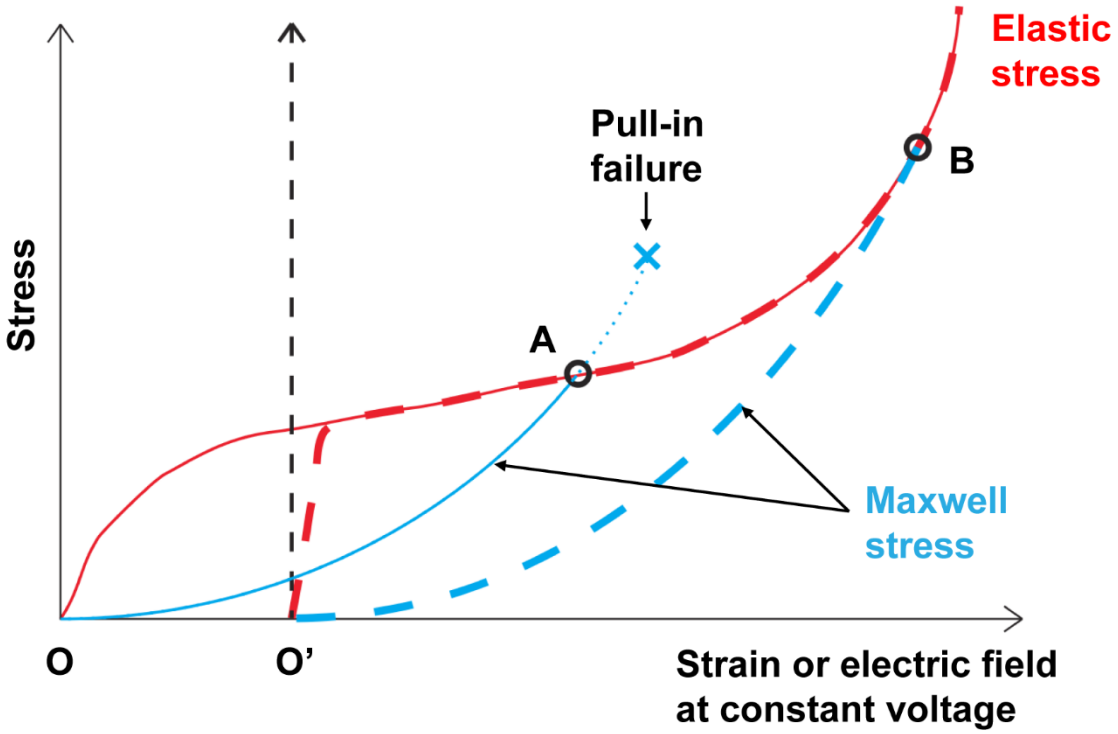

Figure 3. Characteristic stress-strain curves for DEs as a function of mechanical strain or electrical field (under constant voltage) for non-prestretched films (solid curves with origin **O)** and prestretched films (dashed curves at **O**'). The cross (**×**) indicates dielectric breakdown. Points (**A**) and (**B**) represent the apparent breakdown field and actual breakdown strength, respectively.

One method uses interpenetrating polymer networks (IPNs), synthesized by diffusing and polymerizing initiators, monomers, and crosslinkers into a prestretched polymer network (Figure 4).[17] After releasing the IPN from external support, the new network internally maintains the strained state of the original network, behaving similarly to the prestretched film. Ha et al. used 1,6-hexanediol diacrylate (HDDA) to form the new networks and prestretched the VHB films 400% biaxially.[17] With 18.3 wt% poly(HDDA) in the IPN, the resulting freestanding films retained 275% prestrain and exhibited 233% area strain.[17] Brochu et al. also demonstrated an all-silicone IPN, with a soft room temperature vulcanizing (RTV) silicone serving as the host network, and a more rigid, high temperature vulcanizing (HTV) silicone preserving the prestrain in the RTV.[18] The silicone IPN showed better actuation performance than freestanding RTV films with a linear strain of 25% and area strain of 45%.[18]

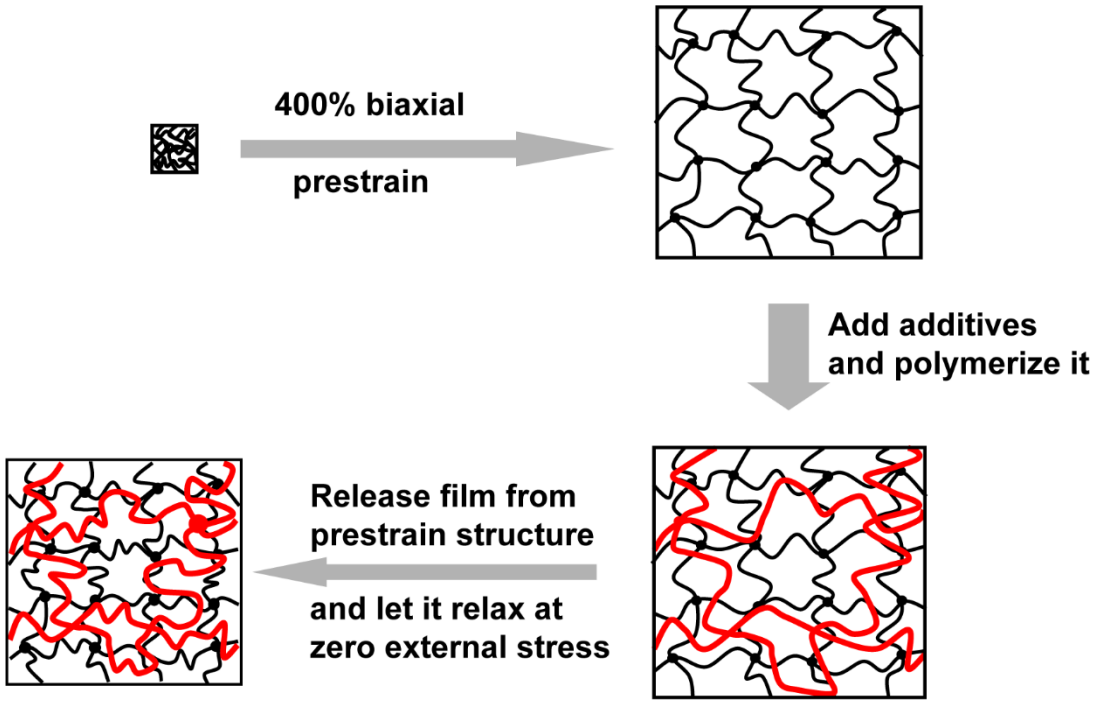

Figure 4. Freestanding interpenetrating polymer network (IPN) fabrication process. The elastomer is prestretched (black network) and sprayed with multifunctional monomer additives which are then polymerized to form the second network (red network) within the host network. Upon release, the IPN preserves most of its prestrain.

Chemically modified elastomers have also achieved high actuation strains without prestretch.[19,20] Niu et al. synthesized new dielectric elastomers that have a stress-strain behavior emulating the red, dashed curve in Figure 3 (see Figure 5).[20] By varying the crosslink density and plasticizer percentage, the modified elastomers achieved large, stable strains and total suppression of EMI. The new DEs had high area strains of up to 100% and high energy densities (>1 J/g).[20] Other teams have explored using bottlebrush elastomers as another method of generating inherent prestrain and freestanding polymers.[21]

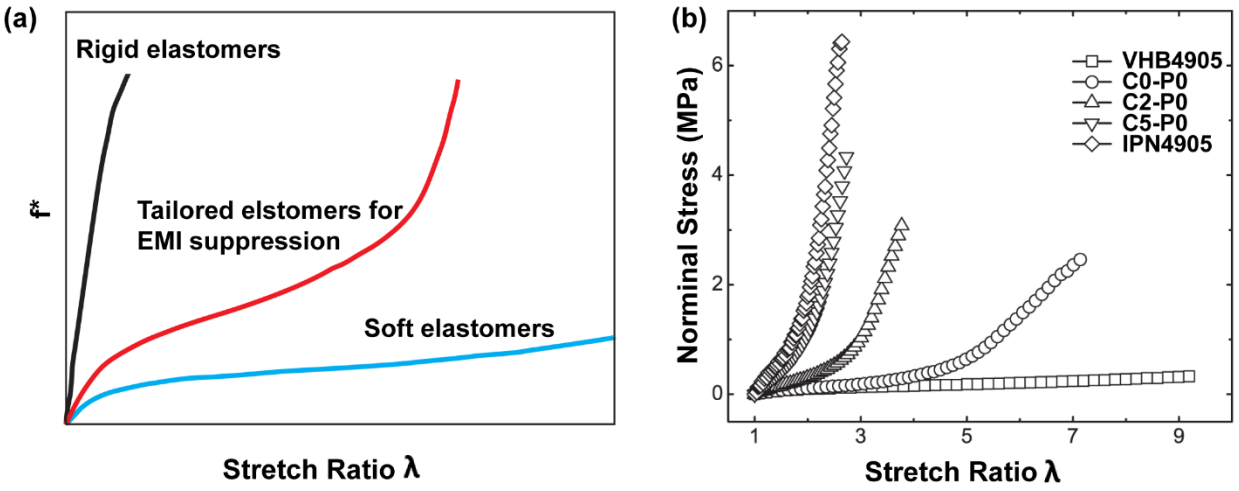

Figure 5. Chemically modified elastomers that can suppress pull-in failure without prestretching. (**a**) Characteristic modulus or stress (**f***) as a function of stretch ratio (**λ**) for rigid, soft, and tailored elastomers. (**b**) Experimental stress-strain behavior of VHB4905, its IPN, and modified elastomers (**Cx-P0**). Reprinted with permission from ref 20. Copyright 2013 Journal of Polymer Science Part B: Polymer Physics.

2.3.Increasing Dielectric Constant

A larger dielectric constant corresponds to a greater capacitance, which reduces the electric field required for actuation (Equation 2). Research has shown that incorporating organic dipole molecules,[22] adding high permittivity ceramic fillers,[23] and introducing conductive nanofillers[24] can increase the dielectric constant in DEs and improve actuation performance. However, the introduction of polar groups usually decreases the dielectric strength and energy density,[25] and ceramic-DE composites require large amounts of fillers to obtain high dielectric constants, leading to stiffening and agglomeration.[26] The use of conductive fillers, however, can achieve a high dielectric constant at relatively low filler loading.[27,28] Hu et al. synthesized an aluminum nanoparticle-acrylate composite with improved dielectric constant and low filler loading content.[28] To eliminate the possibility of agglomeration, the nanoparticles were functionalized with methacrylate molecules and copolymerized with the acrylate matrix (Figure 6a, 6b). The resulting nanocomposite, containing 4 vol% aluminum nanoparticles, improved in dielectric constant from 5.3 (for neat acrylate copolymer to 8.5 and demonstrated a maximum area strain of 56% at 140 MV/m (Figure 6c, 6d).[28]

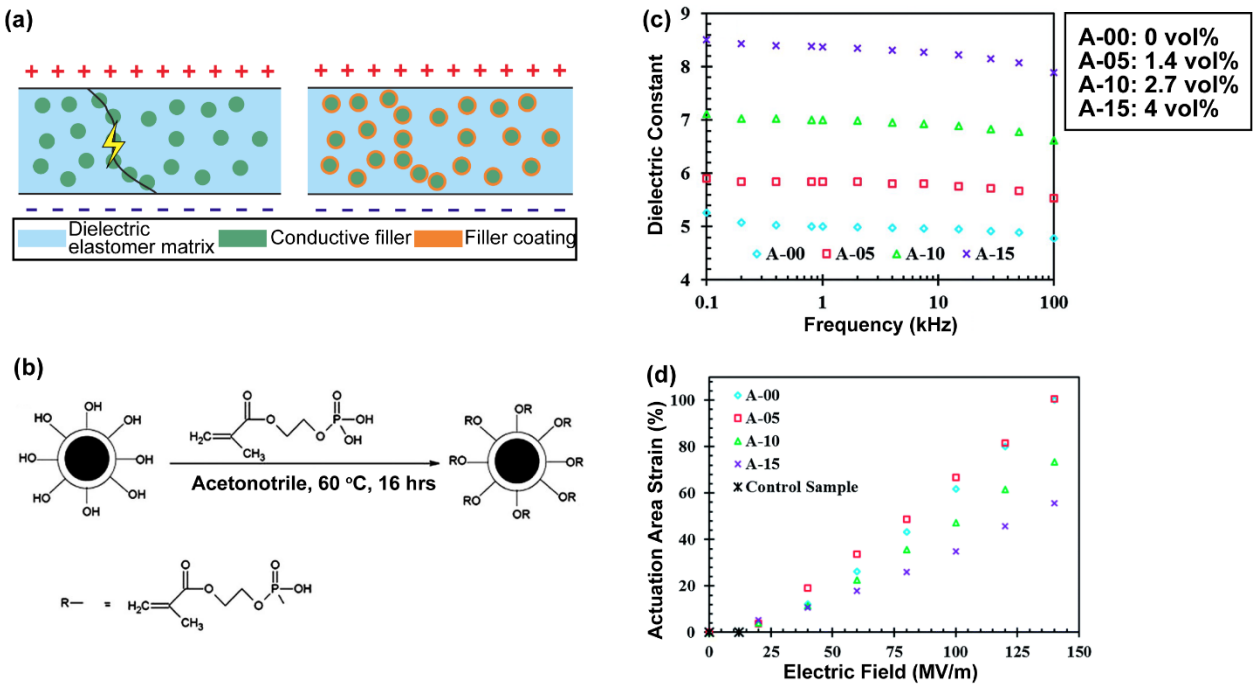

Figure 6. Conductive nanofillers improve the dielectric constant. (**a**) Illustration of a DE composite experiencing electrical breakdown due to a conductive path formed by fillers (left); encapsulated conductive fillers with hindered electrical percolation (right). (**b**) Schematic of functionalized aluminum nanoparticles. (**c**) Dielectric constant and (**d**) actuation tests of composites containing specified amount of aluminum nanoparticles. Reprinted with permission from ref 28. Copyright 2014 Journal of Materials Chemistry C.

2.4.Variable Stiffness Polymers

Most synthetic materials have a constant modulus, which limits their versatility. Materials with tunable stiffness, such as variable stiffness polymers (VSPs), can dynamically respond to changing working conditions much like natural muscles do.[29,30]

2.4.1. Polyacrylate with Diels-Alder bonds

In the theory of rubber elasticity, the tensile modulus of an ideal elastomer is related to the polymer chain segment length.[31] Using dynamic covalent bonds to change the chain segment length, or crosslinking density, makes the dielectric elastomer's stiffness adjustable. The Diels-Alder (DA) reaction can effectively form thermo-reversible covalent bonds.[32] Hu et al. synthesized polyacrylic-based VSPs that contained furan-maleimide DA adduct moieties (PADA) for tunable stiffness (Figure 7a).[33] The furan-maleimide adducts can repeatedly break and reform through a DA reaction, altering the crosslinking density and the elastic moduli. The polymer showed an incremental and reversible modulus change between 0.17 (PADA-4-s) and 0.52 MPa (PADA-4-r) (Figure 7b). The soft state polymer showed a large actuation strain of 35% area expansion at 80 MV/m, while the rigid state polymer achieved high force output of 0.55 MPa at 104 MV/m. (Figure 7c).[33]

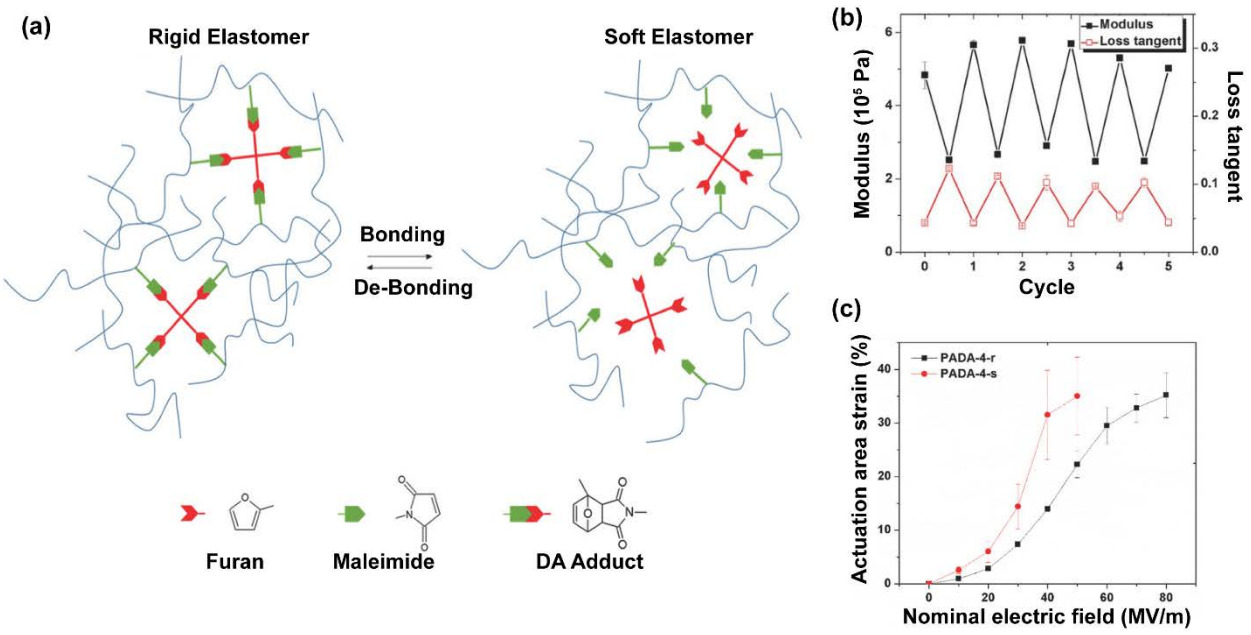

Figure 7. Variable stiffness polyacrylate with Diels-Alder bonds. (**a**) Polyacrylate crosslinked by a thermoreversible Diels-Alder reaction. (**b**) Cyclic thermal treatment for modulus reversibility. (**c**) Areal actuation strain of polyacrylic Diels-Alder VSP in the soft (PADA-4-s) and rigid (PADA-4-r) state at different electric fields. Reprinted with permission from ref 33. Copyright 2015 Advanced Functional Materials.

2.4.2. Bistable electroactive polymer

Since DEAs require a low elastic modulus for high actuation strain, they lack the stiffness required for structural applications. In addition, DEAs require a continuously applied field to maintain actuation, which consumes energy and reduces device lifetime. We synthesized bistable electroactive polymers (BSEPs) that combine the properties of shape memory polymers and DEAs, allowing for rigid-to-rigid actuation.[34–37] At room temperature, BSEPs behave like rigid plastics, but when above their transition temperature (T_r), they enter a rubbery state and can actuate like DEs (Figure 8a). Once the temperature is below T_r, the bias voltage can be removed, and the actuated deformation is preserved. The BSEPs can thus be actuated to variable stable shapes without external energy input to maintain the new shapes (Figure 8b).

BSEPs are categorized into two groups based on their physical states: glass transition BSEPs (Figure 9a) and phase-changing BSEPs (Figure 9b). The first glass transition BSEP was a linear thermoplastic poly(tert-butyl acrylate) (PTBA).[34] It exhibited a storage modulus change from 1.5 GPa at room temperature to 0.42 MPa at 70 °C (Figure 9c) and achieved a maximum area expansion of 335% at 260 MV/m (Figure 8b).[34] Although the PTBA successfully demonstrated controlled, reversible shape change and enough strength for serious mechanical loads, the actuation was unstable at strains over 100%. The modulus of linear PTBA continuously decreased with increased temperature in the softened state, which resulted in non-uniform actuation or even EMI caused by temperature fluctuations. Niu et al. resolved those issues by crosslinking the PTBA and introducing a second monomer to create an IPN.[35] The crosslinked PTBA-IPN had a constant modulus in the rubbery state and a stable area strain of 228% at 148MV/m (Figure 9c,9d).

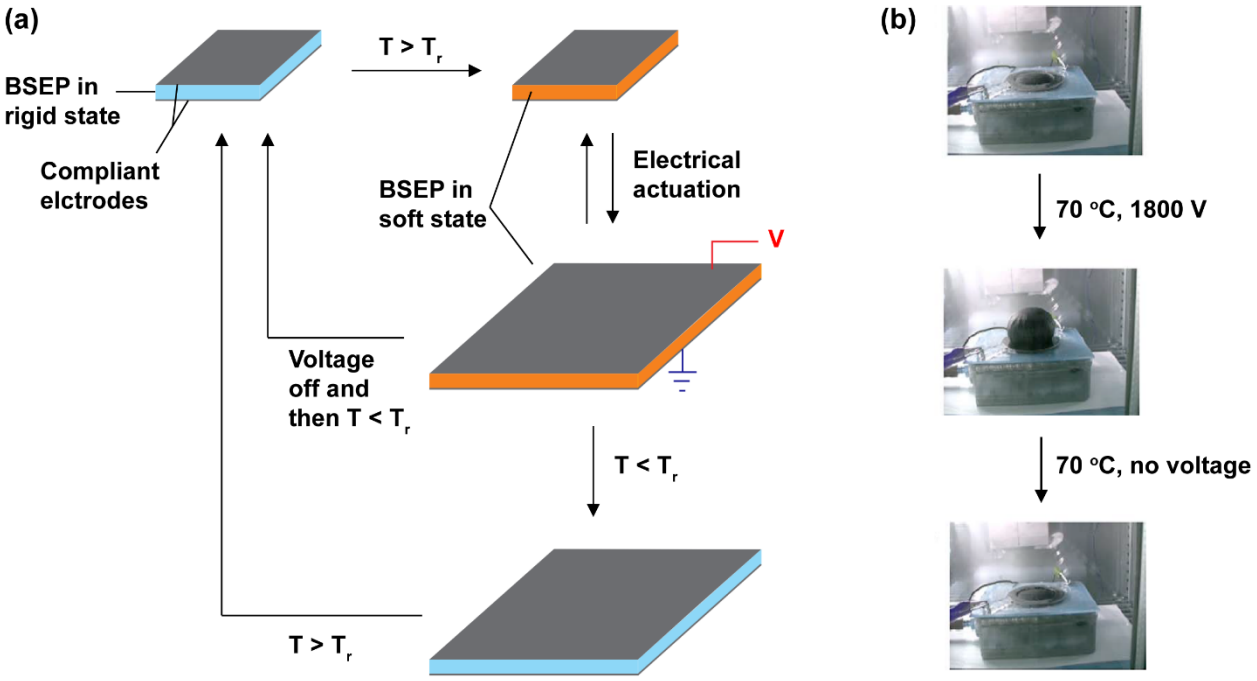

Figure 8. BSEP as DE actuators. (**a**) Working mechanism of a BSEP-based actuator. **(b)** Performance of diaphragm actuators using a glass transition BSEP. Reprinted with permission from ref 34. Copyright 2009 Applied Physics Letters.

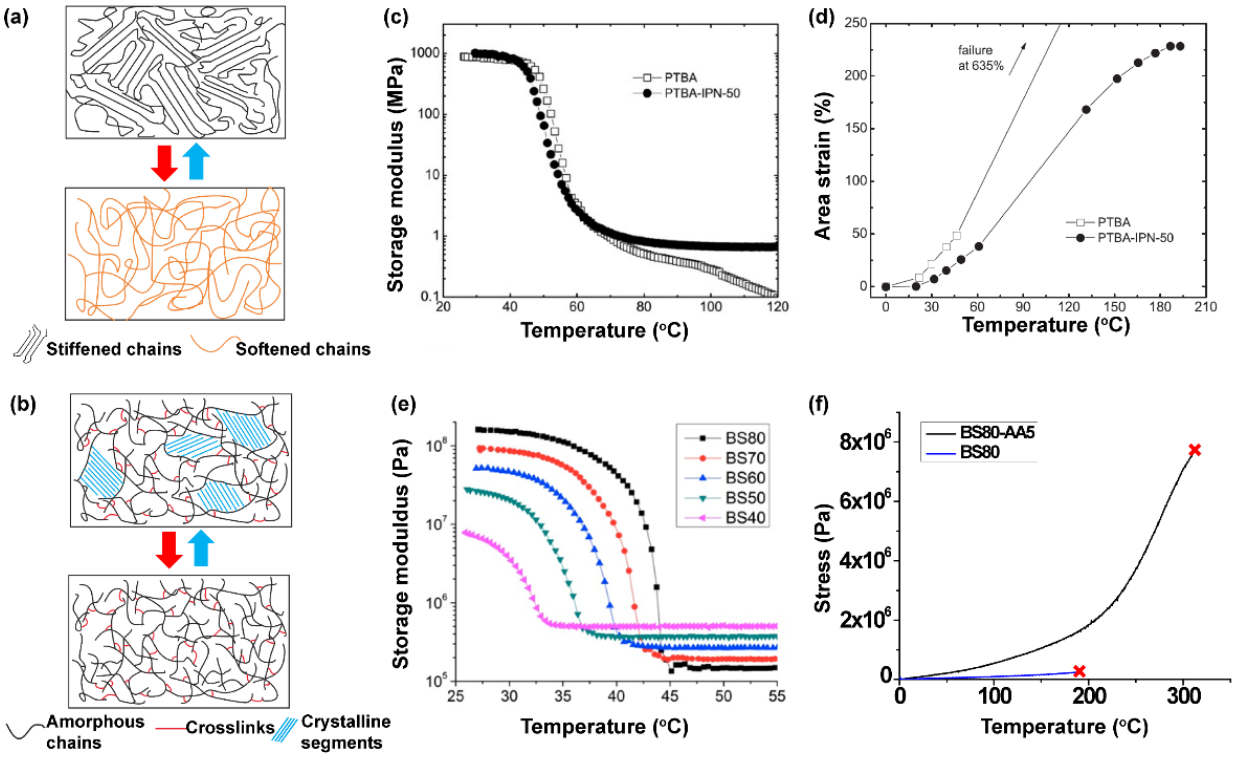

Figure 9. Improvements in the mechanical and dielectric properties of BSEP materials. (**a**) Illustration of the stiffness change of a glass transition BSEP and (**b**) phase-changing BSEP. (**c**) Comparison in modulus-temperature profile and (**d**) dielectric actuation performance between linear PTBA and crosslinked PTBA-IPN. Reprinted with permission from ref 35. Copyright 2012 Advanced materials. (**e**) Temperature dependence of phase-changing BSEP (BSxx) moduli (**f**) Stress-strain response of BS80-AA5 and BS80 at 50 °C.

The phase-changing BSEPs, introduced by Ren et al.[36], exhibit drastically narrowed T_r bands (Figure 9e) and lower inflection point below 50 °C, which is ideal for human-contact applications.[37] The narrow transition band results from the rapid crystallization and melting of stearyl acrylate (SA) moieties in the polymer network. In the softened state, the phase-changing BSEP can produce 70% area strain at 127 MV/m. To further improve the mechanical properties, Qiu et al. modified the BSEP by adding a small amount of acrylic acid to the system (BS80-AA5).[37] The hydrogen bonds from AA help increase the modulus variation to 3000-fold and form reversible cross-links to enhance the toughness, improving true tensile strength to 34 MPa (Figure 9f).

3. COMPLIANT ELECTRODES FOR DEAs

DEA electrodes must be more compliant and stretchable than the elastomer film to prevent stiffness constrain. Additionally, the electrodes must withstand large deformation, occasionally more than 200% area expansion, while maintaining their conductivity across millions of actuation cycles.[6,38] Carbon-based materials, such as carbon grease and carbon powder, are the most commonly used electrodes in DEAs due to their ease of deposition and low cost.[38] However, carbon grease has handling and patterning issues, and carbon powder has poor stretchability thus limiting their usefulness. Metallic thin films with stretchable patterns have also shown promise as DEA electrodes,[39] but their high stiffness limits the actuation strain. Some research has gone into gold, palladium, or titanium metal ion implantation stretchable electrodes.[40] And although ionic gels are useful as transparent electrodes in DEAs, exhibiting large stretchability and high frequency response,[41,42] they can suffer long term stability issues caused by drying.

In search of an ideal electrode, we developed highly compliant and conductive electrodes using one-dimensional nanomaterials arranged in percolation networks.

3.1.Silver Nanowires

Silver nanowires (AgNWs) emerged as a promising electrode material for DEAs, with advanced mechanical, optical, and electrical properties. They can sustain 160% linear strain while maintaining a sheet resistance below 10 kΩ/sq[43] and offer excellent transparency, which is vital for transparent actuators[44] and devices with an important color aspect.[45] Unfortunately, silver nanowire electrodes typically face stability issues. For example, pristine AgNW networks have shown complete fragmentation when annealed at 250 ˚C. However, a conformal coating of a few nm thick ZnO can well protect the AgNWs; networks prepared in this manner remained intact at 300 ˚C with no signs of fragmentation.[46] The ZnO coating prevents the electrical, optical, and morphological degradation of AgNW networks when under electrical and thermal stress, ensuring their longevity.

3.2.Carbon Nanotubes

Carbon nanotubes (CNTs) are also widely used as electrode materials, due to their thermal stability, mechanical compliance, and chemical resistance.[47] DEAs with ultrathin CNT electrodes have already demonstrated 200% area strain.[48] Furthermore, CNT electrodes also exhibit fault-tolerance similar to natural muscle's ability of accommodating minor damages, leading to increased operational reliability.[18,48,49] The fault-tolerance results from CNTs decomposing, referred to as self-clearing, around defects from localized dielectric breakdown. Yuan et al. demonstrated a CNT electrode that continued to actuate a prestretched VHB film despite a pin puncture through the elastomer (Figure 10a).[48] Later, Stoyanov, et al. established that silicone elastomers with CNT electrodes also attain high electromechanical durability with the same self-clearing mechanism.[49] In their tests, the actuators maintained a high strain even after more than 30

self-cleared, electrical breakdown events, with a lifetime of over 85,000 cycles at a linear strain of 25% (Figure 10b).[49]

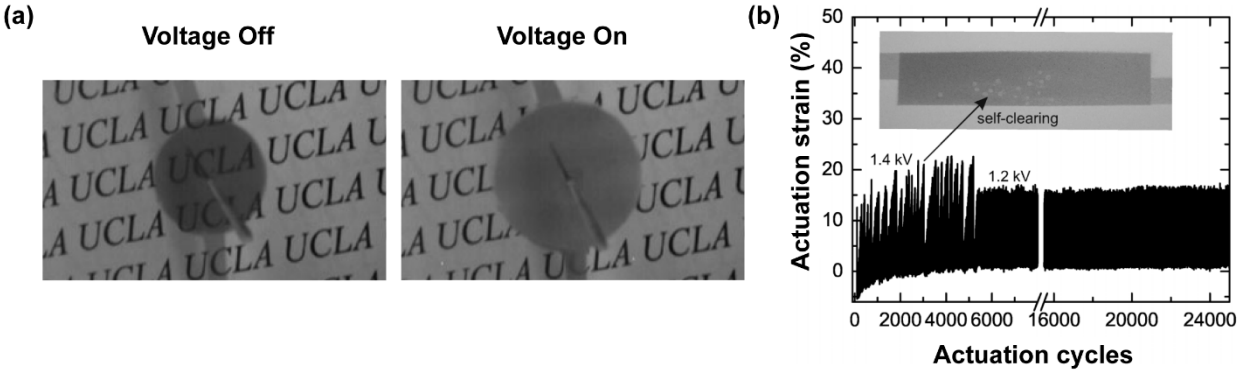

Figure 10. Fault-tolerance in CNT electrodes. (**a**) A VHB-CNT actuator demonstrated 90% actuation strain despite a pin puncture through the film. Reprinted with permission from ref 48. Copyright 2008 Advanced Materials. (**b**) Cyclic actuation tests of a silicone-CNT actuator showed prolonged lifetime due the self-clearing effect from CNT. Reprinted with permission from ref 49. Copyright 2013 RSC Advances.

However, the sharp tips of the CNTs can amplify the electric fields and potentially induce dielectric breakdown of the surrounding air. By incorporating dielectric oil over the CNT electrodes, Yuan, et al. quenched the corona discharge.[50] As a result, their prestretched VHB produced over 150% area expansion continuously for nearly 1500 minutes, which is 20 times longer than comparable DEAs with carbon grease electrodes.[50]

4. DIELECTRIC ELASTOMER DEVICES AND APPLICATIONS

DE actuators benefit largely from their simple construction, light weight, high strain nature, mechanical compliancy, and quietness, leading to many useful modes of operation. While the

agonist-antagonist DEAs mimic natural muscle organization, most DEAs utilize lateral expansion for force output.

4.1.Soft Robotics

Recreating natural forms of movement has been a long running focus for artificial muscle technology. DEAs are a common choice for biomimetic devices, where their similarity to natural muscle simplifies replicating natural systems, and their multifunctionality encourages integrating sensing and loadbearing elements into their design.[51,52] For example, Pei et al designed multiple degree-of-freedom, elastomer spring roll actuators that combined sensing elements and passive structural support (Figure 11).[53] These actuators have significant weight savings and reduced complexity compared to typical designs with external support structures and sensing circuits, and they have clear applications in walking robots, serpentine robots, and object manipulation devices mimicking elephant trunks.[54] A recent work from Acome et al. demonstrated a hydraulically amplified self-healing electrostatic (HASEL) actuator which harnesses an electrohydraulic mechanism to achieve muscle-like movement.[42] A soft gripper and a robotic arm with smart control were built based on HASEL actuators that combine versatility of soft fluidic actuators and self-sensing ability of DEAs. The use of liquid dielectrics enables self-heal from dielectric breakdown, which is novel to the field. However, it may also bring long-term stability issue caused by drying or leakage.

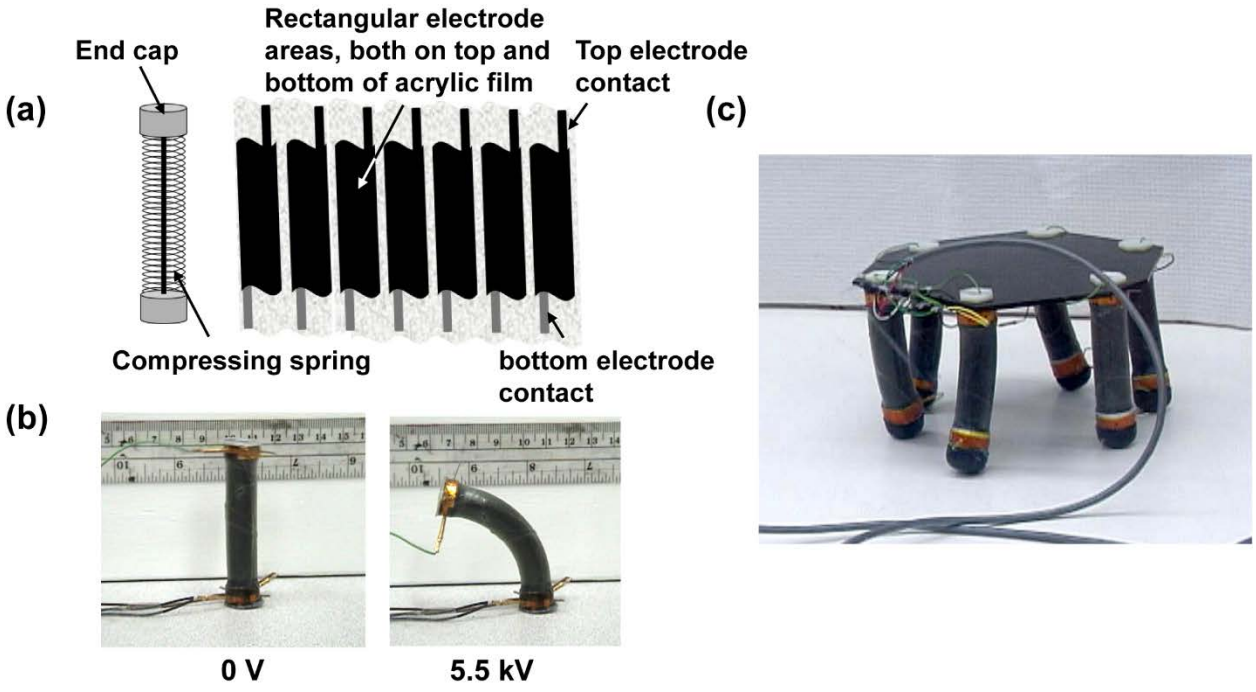

Figure 11. Spring roll actuator-based walking robot. (**a**) Schematic drawing of spring roll actuator fabrication. (**b**) A two-degrees-of-freedom (2-DOF) spring roll bends during actuation under 5.5 kV. (**c**) MERbot: a six-legged robot with 2-DOF spring rolls as legs.

4.2.Biomedicine

Beyond actuators for lightweight robotics, DEAs are also well-received as biomedical technologies. Their soft nature, low cost, and biocompatibility make them appealing for a wide variety of medical devices.[55] Microfluidic emerges as a valuable method for precisely controlling cell environments and extracting useful analysis from minimal sample volumes. Murray et al. created a hybrid device where an integrated DEA actively controls the channel cross section, dynamically tuning pressure and flow characteristics without bulky external systems.[56] Later, McCoul et al. developed a tubular dielectric elastomer valve (DEV) demonstrating an adjustable internal fluidic pressure from 0-3 kPa (Figure 12).[57] These developments have the greatest benefit in addressing clogging, a frequent problem in microfluidic devices.

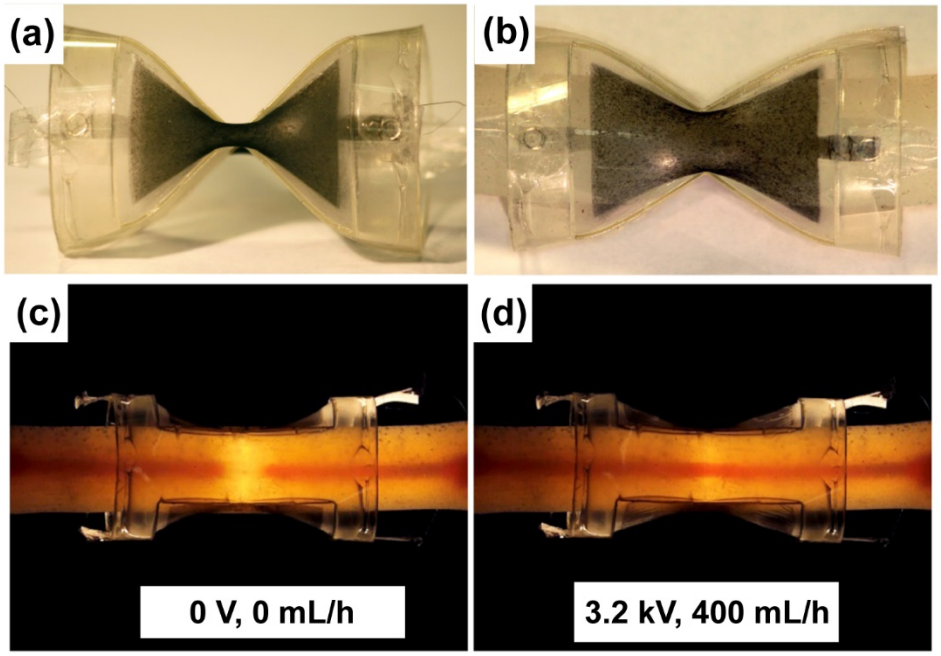

Figure 12. Dielectric elastomer valve (DEV) for microfluidic control. Photographs of a DEV at (**a**) rest and (**b**) actuated state. (**c, d**) Visual characterization of DEV fluidic control (aqueous dye in red) via backlight illumination.

4.3.Bistable Displays

BSEPs can achieve rigid-to-rigid transitions with no power required to maintain any of its stable states. One application for this technology comes in the form of a compact Braille device (Figure 13a).[35] The BSEP forms a refreshable Braille surface with more than enough stability to meet the needs of the Braille standard, and it is significantly more compact than current piezoelectric actuator designs. Later improvements to this design drastically reduced the actuation voltage to 30 V by incorporating BSEPs with pneumatic actuation (Figure 13b).[37,58]

BSEP actuators can also modulate photonic crystals embedded in the elastomer, which enables an electric field to control reflectance and transmittance properties (Figure 13c).[45] This has compelling potential as a display technology forming the basis of soft, wearable displays with low power requirements and as a “re-writable inkless paper”.

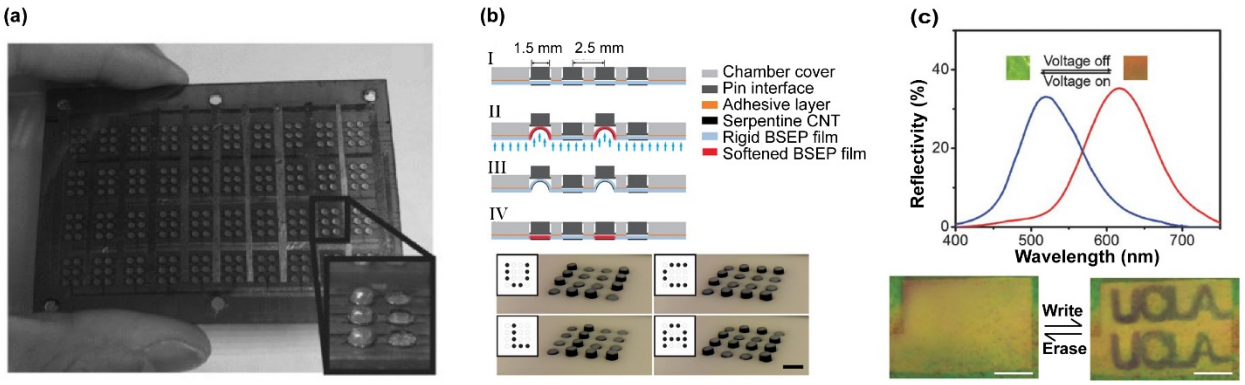

Figure 13. BSEP-based devices. (**a**) Refreshable Braille panel made of BSEP actuators. Reprinted with permission from ref 35. Copyright 2012 Advanced Materials. (**b**) Cross-sectional view of working mechanism of the BSEP pneumatic actuators (upper) and demonstration of the tactile panel showing “U”, “C”, “L”, “A” (lower). The scale bar is 2 mm. (**c**) Reversible writing and erasing of BSEP photonic nanocomposite by switching the electric field on and off. The scale bar is 3 mm. Reprinted with permission from ref 45. Copyright 2018 Advanced Functional Materials.

5. SUMMARY AND OUTLOOKS

In this Account, we surveyed the important developments and our own explorations in DE materials, compliant electrodes, and actuator devices. New DE materials with tailored stress-strain behavior, higher dielectric constants, and adjustable stiffnesses have improved the materials performance and broadened the scope of their applications. Meanwhile, AgNW, CNT, and polymer electrolyte gel electrodes have enabled large and stable actuation. As “artificial muscles”, the DE actuators have been studied for numerous applications, including soft robotics, energy generation, and human-machine interfaces.

The real-life applications of DEAs in scenarios like the one depicted in the conspectus image remain challenging. While the maximum performance of DEAs far exceeds that of natural muscles, (e.g. >1 MPa stress, >100% strain, and 100 Hz response speed), the stable operation performance metrics of DEAs have not been fully established, but should be much lower. Besides operational stability and reliability, high driving voltage has also been a major issue for safety concern and lack of compact and efficient driving circuit.

We can speculate on a few research areas that may advance the DEA technology going forward. With regards to the materials aspect, the BSEPs showed tunable stiffness similar to natural muscles37, bottlebrush elastomers demonstrated high actuation sensitivity with large strains under low electric fields21, and liquid dielectrics enabled immediate self-healing after numerous breakdown events42. The type of driving mechanism is also an area worth investigating. Pneumatic and hydraulic driven actuators are also widely employed in soft robotics due to their high force output at small scales. The HASEL actuator combined the strength of fluidic actuators and the compactness of DE actuators42. Research regarding other driving mechanisms for soft

actuators, such as magnetic field, light, and thermal energy have also gained significant traction.59 To enable smart control, efforts devoted to building dielectric elastomer switches (DES)60 may add self-sensing functionality to DEAs. DES enables safe and precise control of DEA actuation by sensing changes in the resistance of the electrodes or capacitance during actuation, which considerably reduces the complexity and size of the driving electronics.

Overall, dielectric elastomers have continually advanced since the 1990s, exhibiting previously unattainable properties and performances. The unique combinations of large-strain actuation, flexibility, and structural functions of DEAs should open new applications where conventional actuator technologies struggle. Further advancements may even see DEAs encroaching in territories where the conventional technologies have dominated for decades.

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REFERENCES

(1) Madden, J. D. W.; Vandesteeg, N. A.; Anquetil, P. A.; Madden, P. G. A.; Takshi, A.; Pytel, R. Z.; Lafontaine, S. R.; Wieringa, P. A.; Hunter, I. W. Artificial Muscle Technology: Physical Principles and Naval Prospects. *IEEE J. Ocean. Eng.* **2004,** *29*, 706–728.

(2) Zhenyi, M.; Scheinbeim, J. I.; Lee, J. W.; Newman, B. A. High Field Electrostrictive Response of Polymers. *J. Polym. Sci. Part B Polym. Phys.* **1994**, *32*, 2721–2731.

(3) Pelrine, R.; Kornbluh, R.; Joseph, J.; Chiba, S.; Park, M. Electrostriction of Polymer Films for Microactuators. *IEEE Micro Electro Mech. Syst.* **1997**, *1*, 238–243.

(4) Pei, Q.; Inganläs, O. Conjugated Polymers and the Bending Cantilever Method: Electrical Muscles and Smart Devices. *Adv. Mater.* **1992,** *4*, 277–278.

(5) Brochu, P.; Pei, Q. Advances in Dielectric Elastomers for Actuators and Artificial Muscles. *Macromol. Rapid Commun.* **2010**, *31*, 10–36.

(6) Pelrine, R.; Kornbluh, R.; Pei, Q.; Joseph, J. High-Speed Electrically Actuated Elastomers with Strain Greater Than 100%. *Science.* **2000**, *287*, 836–839.

(7) Pelrine, R.; Kornbluh, R.; Kofod, G. High-Strain Actuator Materials Based on Dielectric Elastomers. *Adv. Mater.* **2000**, *12*, 1223–1225.

(8) Montanari, G. I. Lettere Inedite Di Alessandro Volta. *Stamperia Nobili, Pesaro, Italy* **1835**, 15–17.

(9) Roentgen, W. C. About the Changes in Shape and Volume of Dielectrics Caused by Electricity. *Annu. Phys. Chem. Ser.* **1880**, *11*, 771–786.

(10) Sacerdote, M. P. On the Electrical Deformation of Isotropic Dielectric Solids. *J. phys* **1899**, *3*, 282–285.

(11) Biggs, J.; Danielmeier, K.; Hitzbleck, J.; Krause, J.; Kridl, T.; Nowak, S.; Orselli, E.; Quan, X.; Schapeler, D.; Sutherland, W.; Wagner, J.. Electroactive Polymers: Developments of and Perspectives for Dielectric Elastomers. *Angew. Chemie - Int. Ed.* **2013**, *52*, 9409–9421.

(12) Kofod, G.; Sommer-Larsen, P.; Kornbluh, R.; Pelrine, R. Actuation Response of Polyacrylate Dielectric Elastomers. *J. Intell. Mater. Syst. Struct.* **2003**, *14*, 787–793.

(13) Plante, J.-S.; Dubowsky, S. On the Properties of Dielectric Elastomer Actuators and Their Design Implications. *Smart Mater. Struct.* **2007**, *16*, S277.

(14) Zhang, H.; Düring, L.; Kovacs, G.; Yuan, W.; Niu, X.; Pei, Q. Interpenetrating Polymer Networks Based on Acrylic Elastomers and Plasticizers with Improved Actuation Temperature Range. *Polym. Int.* **2010**, *59*, 384–390.

(15) Plante, J. S.; Dubowsky, S. Large-Scale Failure Modes of Dielectric Elastomer Actuators. *Int. J. Solids Struct.* **2006**, *43*, 7727–7751.

(16) Zhao, X.; Suo, Z. Theory of Dielectric Elastomers Capable of Giant Deformation of Actuation. *Phys. Rev. Lett.* **2010**, *104*, 1–4.

(17) Ha, S. M.; Yuan, W.; Pei, Q.; Pelrine, R.; Stanford, S. Interpenetrating Polymer Networks for High-Performance Electroelastomer Artificial Muscles. *Adv. Mater.* **2006**, *18*, 887–891.

(18) Brochu, P.; Stoyanov, H.; Niu, X.; Pei, Q. All-Silicone Prestrain-Locked Interpenetrating Polymer Network Elastomers: Free-Standing Silicone Artificial Muscles with Improved Performance and Robustness. *Smart Mater. Struct.* **2013**, *22*, 055022.

(19) Vargantwar, P. H.; Özçam, A. E.; Ghosh, T. K.; Spontak, R. J. Prestrain-Free Dielectric Elastomers Based on Acrylic Thermoplastic Elastomer Gels: A Morphological and (Electro)Mechanical Property Study. *Adv. Funct. Mater.* **2012**, *22*, 2100–2113.

(20) Niu, X.; Stoyanov, H.; Hu, W.; Leo, R.; Brochu, P.; Pei, Q. Synthesizing a New Dielectric Elastomer Exhibiting Large Actuation Strain and Suppressed Electromechanical Instability without Prestretching. *J. Polym. Sci. Part B Polym. Phys.* **2013**, *51*, 197–206.

(21) Vatankhah-Varnoosfaderani, M.; Daniel, W. F. M.; Zhushma, A. P.; Li, Q.; Morgan, B. J.; Matyjaszewski, K.; Armstrong, D. P.; Spontak, R. J.; Dobrynin, A. V.; Sheiko, S. S. Bottlebrush Elastomers: A New Platform for Freestanding Electroactuation. *Adv. Mater.* **2017**, *29*, 1604209.

(22) Zhang, Q.; Li, H.; Poh, M.; Xia, F.; Cheng, Z.-Y.; Xu, H.; Huang, C. An All-Organic Composite Actuator Material with a High Dielectric Constant. **2002**, *419*, 284–287.

(23) Carpi, F.; De Rossi, D. Improvement of Electromechanical Actuating Performances of a Silicone Dielectric Elastomer by Dispersion of Titanium Dioxide Powder. *IEEE Trans. Dielectr. Electr. Insul.* **2005**, *12*, 835–843.

(24) Molberg, M.; Crespy, D.; Rupper, P.; Nüesch, F.; Manson, J. A. E.; Löwe, C.; Opris, D. M. High Breakdown Field Dielectric Elastomer Actuators Using Encapsulated Polyaniline as High Dielectric Constant Filler. *Adv. Funct. Mater.* **2010**, *20*, 3280–3291.

(25) Kussmaul, B.; Risse, S.; Kofod, G.; Waché, R.; Wegener, M.; McCarthy, D. N.; Krüger, H.; Gerhard, R. Enhancement of Dielectric Permittivity and Electromechanical Response in Silicone Elastomers: Molecular Grafting of Organic Dipoles to the Macromolecular

Network. *Adv. Funct. Mater.* **2011**, *21*, 4589–4594.

(26) Gallone, G.; Carpi, F.; De Rossi, D.; Levita, G.; Marchetti, A. Dielectric Constant Enhancement in a Silicone Elastomer Filled with Lead Magnesium Niobate-Lead Titanate. *Mater. Sci. Eng. C* **2007**, *27*, 110–116.

(27) Zhang, S.; Zhang, N.; Huang, C.; Ren, K.; Zhang, Q. Microstructure and Electromechanical Properties of Carbon Nanotube/ Poly(Vinylidene Fluoride-Trifluoroethylene-Chlorofluoroethylene) Composites. *Adv. Mater.* **2005**, *17*, 1897–1901.

(28) Hu, W.; Zhang, S. N.; Niu, X.; Liu, C.; Pei, Q. An Aluminum Nanoparticle-Acrylate Copolymer Nanocomposite as a Dielectric Elastomer with a High Dielectric Constant. *J. Mater. Chem. C.* **2014**, *2*, 1658–1666.

(29) Wang, L.; Yang, Y.; Chen, Y.; Majidi, C.; Iida, F.; Askounis, E.; Pei, Q. Controllable and Reversible Tuning of Material Rigidity for Robot Applications. *Materials Today*. 2018.

(30) Pei, Q. Variable Stiffness Materials. *Soft Robot.* **2016**, *3*, 1–2.

(31) Brazel, C. S.; Rosen, S. L. *Fundamental Principles of Polymeric Materials*; **2012**.

(32) Chen, X.; Dan, M. A.; Ono, K.; Mal, A.; Shen, H.; Nutt, S. R.; Sheran, K.; Wudl, F. A Thermally Re-Mendable Cross-Linked Polymeric Material. *Science.* **2002**, *295*, 1698–1702.

(33) Hu, W.; Ren, Z.; Li, J.; Askounis, E.; Xie, Z.; Pei, Q. New Dielectric Elastomers with Variable Moduli. *Adv. Funct. Mater.* **2015**, *25*, 4827–4836.

(34) Yu, Z.; Yuan, W.; Brochu, P.; Chen, B.; Liu, Z.; Pei, Q. Large-Strain, Rigid-to-Rigid

Deformation of Bistable Electroactive Polymers. *Appl. Phys. Lett.* **2009**, *95*, 21–24.

(35) Niu, X.; Yang, X.; Brochu, P.; Stoyanov, H.; Yun, S.; Yu, Z.; Pei, Q. Bistable Large-Strain Actuation of Interpenetrating Polymer Networks. *Adv. Mater.* **2012**, *24*, 6513–6519.

(36) Ren, Z.; Hu, W.; Liu, C.; Li, S.; Niu, X.; Pei, Q. Phase-Changing Bistable Electroactive Polymer Exhibiting Sharp Rigid-to-Rubbery Transition. *Macromolecules* **2016**, *49*, 134–140.

(37) Qiu, Y.; Lu, Z.; Pei, Q. Refreshable Tactile Display Based on a Bistable Electroactive Polymer and a Stretchable Serpentine Joule Heating Electrode. *ACS Appl. Mater. Interfaces* **2018**, *10*, 24807–24815.

(38) Rosset, S.; Shea, H. R. Flexible and Stretchable Electrodes for Dielectric Elastomer Actuators. *Appl. Phys. A Mater. Sci. Process.* **2013**, *110*, 281–307.

(39) Pelrine, R.; Kornbluh, R.; Joseph, J.; Heydt, R.; Pei, Q.; Chiba, S. High-Field Deformation of Elastomeric Dielectrics for Actuators. *Mater. Sci. Eng. C* **2000**, *11*, 89–100.

(40) Rosset, S.; Niklaus, M.; Dubois, P.; Shea, H. R. Metal Ion Implantation for the Fabrication of Stretchable Electrodes on Elastomers. *Adv. Funct. Mater.* **2009**, *19*, 470–478.

(41) Keplinger, C.; Sun, J. Y.; Foo, C. C.; Rothemund, P.; Whitesides, G. M.; Suo, Z. Stretchable, Transparent, Ionic Conductors. *Science.* **2013**, *341*, 984-987.

(42) Acome, E.; Mitchell, S. K.; Morrissey, T. G.; Emmett, M. B.; Benjamin, C.; King, M.; Radakovitz, M.; Keplinger, C. Hydraulically Amplified Self-Healing Electrostatic Actuators with Muscle-like Performance. *Science.* **2018**, *359*, 61–65.

(43) Yun, S.; Niu, X.; Yu, Z.; Hu, W.; Brochu, P.; Pei, Q. Compliant Silver Nanowire-Polymer Composite Electrodes for Bistable Large Strain Actuation. *Adv. Mater.* **2012**, *24*, 1321-1327.

(44) Liang, J.; Li, L.; Chen, D.; Hajagos, T.; Ren, Z.; Chou, S. Y.; Hu, W.; Pei, Q. Intrinsically Stretchable and Transparent Thin-Film Transistors Based on Printable Silver Nanowires, Carbon Nanotubes and an Elastomeric Dielectric. *Nat. Commun.* **2015**, *6*, 7647.

(45) Xie, Y.; Meng, Y.; Wang, W.; Zhang, E.; Leng, J.; Pei, Q. Bistable and Reconfigurable Photonic Crystals-Electroactive Shape Memory Polymer Nanocomposite for Ink-Free Rewritable Paper. *Adv. Funct. Mater.* **2018**, *1802430*, 1802430.

(46) Chen, D.; Liang, J.; Liu, C.; Saldanha, G.; Zhao, F.; Tong, K.; Liu, J.; Pei, Q. Thermally Stable Silver Nanowire-Polyimide Transparent Electrode Based on Atomic Layer Deposition of Zinc Oxide on Silver Nanowires. *Adv. Funct. Mater.* **2015**, *25*, 7512–7520.

(47) Ruoff, R. S.; Lorents, D. C. Mechanical and Thermal Properties of Carbon Nanotubes. *Carbon N. Y.* **1995**, *33*, 925–930.

(48) Yuan, W.; Hu, L.; Yu, Z.; Lam, T.; Biggs, J.; Ha, S. M.; Xi, D.; Chen, B.; Senesky, M. K.; Grüner, G.; Pei, Q. Fault-Tolerant Dielectric Elastomer Actuators Using Single-Walled Carbon Nanotube Electrodes. *Adv. Mater.* **2008**, *20*, 621–625.

(49) Stoyanov, H.; Brochu, P.; Niu, X.; Lai, C.; Yun, S.; Pei, Q. Long Lifetime, Fault-Tolerant Freestanding Actuators Based on a Silicone Dielectric Elastomer and Self-Clearing Carbon Nanotube Compliant Electrodes. *RSC Adv.* **2013**, *3*, 2272–2278.

(50) Yuan, W.; Brochu, P.; Ha, S. M.; Pei, Q. Dielectric Oil Coated Single-Walled Carbon

Nanotube Electrodes for Stable, Large-Strain Actuation with Dielectric Elastomers. *Sensors Actuators, A Phys.* **2009**, *155*, 278–284.

(51) Godaba, H.; Li, J.; Wang, Y.; Zhu, J. A Soft Jellyfish Robot Driven by a Dielectric Elastomer Actuator. *IEEE Robot. Autom. Lett.* **2016**, *1*, 624–631.

(52) Li, T.; Li, G.; Liang, Y.; Cheng, T.; Dai, J.; Yang, X.; Liu, B.; Zeng, Z.; Huang, Z.; Luo, Y.; Xie, T., Yang, W. Fast-Moving Soft Electronic Fish. *Sci. Adv.* **2017**, *3*, e1602045.

(53) Pei, Q.; Pelrine, R.; Stanford, S.; Kornbluh, R.; Rosenthal, M. Electroelastomer Rolls and Their Application for Biomimetic Walking Robots. In *Synthetic Metals*; **2003**, *135*, 129-131.

(54) Pei, Q.; Rosenthal, M.; Stanford, S.; Prahlad, H.; Pelrine, R. Multiple-Degrees-of-Freedom Electroelastomer Roll Actuators. *Smart Mater. Struct.* **2004**, *13*, N86.

(55) Carpi, F.; Smela, E. *Biomedical Applications of Electroactive Polymer Actuators*; John Wiley & Sons, **2009**.

(56) Murray, C.; McCoul, D.; Sollier, E.; Ruggiero, T.; Niu, X.; Pei, Q.; Carlo, D. Di. Electro-Adaptive Microfluidics for Active Tuning of Channel Geometry Using Polymer Actuators. *Microfluid. Nanofluidics* **2013**, *14*, 345–358.

(57) McCoul, D.; Pei, Q. Tubular Dielectric Elastomer Actuator for Active Fluidic Control. *Smart Mater. Struct.* **2015**, *24*, 105016.

(58) Besse, N.; Rosset, S.; Zarate, J. J.; Shea, H. Flexible Active Skin: Large Reconfigurable Arrays of Individually Addressed Shape Memory Polymer Actuators. *Adv. Mater. Technol.* **2017**, *1700102*, 1700102.

(59) Hines, L.; Petersen, K.; Lum, G. Z.; Sitti, M. Soft Actuators for Small-Scale Robotics. *Adv. Mater.* **2017**, *29*, 1603483.

(60) O'Brien, B. M.; Calius, E. P.; Inamura, T.; Xie, S. Q.; Anderson, I. A. Dielectric Elastomer Switches for Smart Artificial Muscles. *Appl. Phys. A Mater. Sci. Process.* **2010**, *100*, 385–389.

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