ABSTRACT

FANG, XIAOMENG. Anisotropic D-EAP Electrodes and their Application in Spring Roll Actuators. (Under the direction of Dr. Tushar K. Ghosh).

Electroactive polymers (EAPs) exhibit shape change when subjected to an electric field. They are lightweight, soft, and inexpensive, while they are easy to process, shape, and tune to offer a broad range of mechanical and electrical properties. Dielectric electroactive polymers (D-EAP) constitute a class of EAPs with great potential. D-EAPs consist of physically or chemically cross-linked macromolecular networks and are mechanically isotopic. Therefore, in most actuator applications that require directional electromechanical response, it is necessary to use other complex means to direct the stress/strain in the preferred direction.

In this work, a simple carbon nanotube (CNT) based electrode for D-EAP actuators is demonstrated that vastly improves directional strain response originating from the mechanical anisotropy of the electrode material. Using this novel approach, the mechanical anisotropy, defined as the ratio of initial modulus in fiber direction and that in cross-fiber direction, of the CNT electroded VHB actuators, ranges from 7.9 to 11.2. Hence, the CNT-VHB flat film actuators show high directed linear actuation strain in cross-fiber direction of greater than 25% meanwhile almost no strain in fiber direction at a relatively low electric field (120 V μm⁻¹).

The morphology of the CNT sheets has critical influence on their mechanical properties and resultant actuator performance. The results demonstrate the efficacy of microcombing and selective laser etching processes to improve the CNT fiber alignment to produce pure unidirectional strain of 33% at a relatively moderate electric field.

Unidirectional D-EAP composite laminates using polyurethane and polyamide monofilaments are also employed in spring roll actuators to investigate their directional mechanical and
electromechanical properties. While CNT electroded D-EAP spring roll actuators were found to have about the same performance as actuators with carbon grease electrodes (6.5% strain in CNT electroded spring roll actuators and 8.2% for carbon grease electroded actuators at 5kV), spring roll actuator made of fiber reinforced VHB composites with carbon grease electrodes showed marginal improvement in actuation strain (9.9%-11% strain in longitudinal direction at 5kV).
Anisotropic D-EAP Electrodes and their Application in Spring Roll Actuators

by
Xiaomeng Fang

A dissertation submitted to the Graduate Faculty of North Carolina State University in partial fulfillment of the requirements for the degree of Doctor of Philosophy

Fiber and Polymer Science

Raleigh, North Carolina

2017

APPROVED BY:

Dr. Tushar K. Ghosh
Committee Chair

Dr. Richard J. Spontak

Dr. Philip D. Bradford

Dr. Ericka Ford
DEDICATION

To my dear grandma, my parents, my advisor and his families, my aunt, uncle and my husband for their unconditional love and huge supports.
BIOGRAPHY

Xiaomeng Fang was born in China and she received her Bachelor’s degree in 2008 and Master’s degree in 2011 both in Textile Science and Engineering from Donghua University, China. During her master’s study, she worked on 3D orthogonal woven ramie fabrics reinforced polypropylene composites. She was awarded Major Award of Hongkong Sangma Trust Fund Scholarship, Chinese National Scholarship for Encouragement and Outstanding Academic Performance Scholarship of Donghua University in 2007, Outstanding Graduate student of Donghua University in 2009, Outstanding Graduate Student of Shanghai and Outstanding Individual of Social Practice of College Students of Shanghai in 2011.

In spring of 2013, she joined North Carolina State University to pursue her doctoral degree in Fiber and Polymer Science under the direction of Dr. Tushar K. Ghosh. She worked on aligned carbon nanotube sheets as electrodes to induce anisotropic electromechanical deformation of dielectric elastomer.
ACKNOWLEDGMENTS

I would like to thank my advisor Dr. Tushar K. Ghosh for his guidance, support and motivation throughout my doctoral studies. He encouraged me to think and learn that has been very important for me to finish my study. Dr. Ghosh has always been patient and encouraging in times of problems and setbacks in my work. I also want to express my gratitude to Dr. Ghosh’s family for being an extremely caring host for me in this foreign country.

I am thankful to Dr. Philip D. Bradford for his supports with CNT synthesis. He also kindly allowed me to share the facilities in his lab with me to conduct my experiments. I am thankful to Dr. Richard J. Spontak. After leaning in his two courses in Polymer Science and Technology, Polymer Blends and Alloys, my understanding on fundamental polymers morphologies and properties was extensively broadened and improved. My thanks to Prof. Ericka Ford for agreeing serve in my committee.

I also want to say thanks to my graduated group mates Enes, Krishna, Aylin who lead me to start my journey in our lab and trained me in using the instruments. I would like to thank my current group mates Huiqi Shao, Elena Morgan, Kony Chatterjee and Ashish Kapoor for their kind support and caring.

I really want to express my gratitude to my family members. Their unconditional love and understanding always give me motivation and happiness. I would also like to thank my husband Dr. Kun Luan. He helps me when I have problems, takes care of our families when I am busy, shares his happiness with me and takes care of me.

I also thank my friends Yavuz Caydamli, Yeqian Ge, Ozkan Yildiz, Ang Li, Chengcheng Feng, Yi Ding, Jialong Shen, Renbo Cao, Shanshan Li, Liwen Zhang and Qian Liu.
# TABLE OF CONTENTS

**LIST OF TABLES** .................................................................................................................. ix  
**LIST OF FIGURES** ............................................................................................................... x  

**CHAPTER 1** Introduction ........................................................................................................ 1  
  1.1 Introduction ......................................................................................................................... 1  

**CHAPTER 2** Electroactive Polymers ....................................................................................... 4  
  2.1 Introduction ......................................................................................................................... 4  
    2.1.1 Electric EAPs .................................................................................................................. 6  
      2.1.1.1 Electromechanical Principles .................................................................................. 7  
      2.1.1.2 Martials ................................................................................................................... 10  
    2.1.2 Ionic EAPs .................................................................................................................... 17  
      2.1.2.1 Ionic Polymer Gels ................................................................................................. 17  
      2.1.2.2 Ionomeric Polymer-Metal Composites ................................................................. 19  
      2.1.2.3 Conducting Polymers ............................................................................................ 21  
      2.1.2.4 Carbon Nanotubes ................................................................................................. 23  
  2.2 Dielectric Electroactive Polymers ....................................................................................... 26  
    2.2.1 Characteristic Parameters ............................................................................................. 27  
      2.2.1.1 Actuation Stress and Blocking Force ...................................................................... 28  
      2.2.1.2 Blocking Force ....................................................................................................... 29  
      2.2.1.3 Actuation Strain ....................................................................................................... 30  
      2.2.1.4 Elastic Strain Energy Density .................................................................................. 31  
      2.2.1.5 Electromechanical Coupling Efficiency ................................................................. 32  
      2.2.1.6 Electric Breakdown Strength .................................................................................... 33  
      2.2.1.7 Effects of Prestrain on Actuation Performance ...................................................... 33  
      2.2.1.8 Instability and Failure .............................................................................................. 40  
    2.2.2 Materials and Properties ............................................................................................... 45  
      2.2.2.1 Silicone .................................................................................................................. 46  
      2.2.2.2 Acrylics ................................................................................................................... 48  
      2.2.2.3 Polyurethane ........................................................................................................... 51
4.3.1 Sample Preparation .................................................................................. 122
4.3.2 Actuator Characterization ..................................................................... 122
4.4 Actuator Performance ............................................................................. 123
  4.4.1 Mechanical Behavior ......................................................................... 123
  4.4.2 Morphological Characteristics .......................................................... 125
  4.4.3 Electrical Behavior of Electrodes ....................................................... 127
  4.4.4 Electromechanical Properties ............................................................. 128
4.5 Conclusion .............................................................................................. 132

CHAPTER 5  Enhanced Anisotropic Response of Dielectric Elastomer Actuators with Microcombed and Etched Carbon Nanotube Sheet Electrodes ............................................. 134
  5.1 Introduction ............................................................................................ 134
  5.2 Experimental .......................................................................................... 140
    5.2.1 CNT Synthesis and Preparation ....................................................... 140
    5.2.2 Microcombing Process ..................................................................... 140
    5.2.3 Actuator Preparation ....................................................................... 140
    5.2.4 Laser Ablation Process ..................................................................... 141
    5.2.5 Actuator Characterization ................................................................. 141
  5.3 Result and Discussion ............................................................................ 142
    5.3.1 Morphological Characteristics ........................................................ 142
    5.3.2 Tensile Behavior ............................................................................. 144
    5.3.3 Electrical Behavior .......................................................................... 147
    5.3.4 Field-induced Deformation ............................................................... 149
  5.4 Conclusion .............................................................................................. 153

CHAPTER 6  Anisotropic D-EAP and Application in Spring Roll Actuator .............. 154
  6.1 Introduction ............................................................................................ 155
  6.2 Materials and sample preparation .......................................................... 158
    6.2.1 Spring ............................................................................................. 158
    6.2.2 Fiber ............................................................................................... 159
    6.2.3 Fiber/VHB composites ..................................................................... 160
6.2.4 Spring roll actuator preparation ................................................................. 161
6.3 Characterizations of fiber/VHB composites flat film actuator and spring roll actuator ............................................................................................................. 162
  6.3.1 Mechanical tests of fiber/VHB composites .............................................. 162
  6.3.2 Blocking force tests of flat film actuator made of fiber/VHB composites 163
  6.3.3 Field-induce deformation characterization of flat film actuator and spring roll actuator made of fiber/VHB composites ................................................. 163
6.4 Result and discussion ...................................................................................... 164
  6.4.1 Mechanical test results of fiber/VHB composites ................................. 164
  6.4.2 Field-induced electromechanical response tests ................................. 167
  6.4.3 Blocking force test results of flat film actuator made of fiber/VHB composites .............................................................................................................. 169
6.5 Working principle of spring roll actuator ...................................................... 171
6.6 Fiber reinforced VHB composite spring roll actuator .................................. 173
  6.6.1 Passive equilibrium statue ................................................................. 173
  6.6.2 Initial moment of activation ............................................................... 181
  6.6.3 Active equilibrium ............................................................................... 182
6.7 CNT sheet electroded spring roll actuator .................................................. 191
  6.7.1 Spring roll actuator preparation and characterization ....................... 191
  6.7.2 Result and discussion ........................................................................... 191
    6.7.2.1 Mechanical test results of CNT-VHB flat film actuator ............... 191
    6.7.2.2 Field-induced electromechanical response test results ............... 194
    6.7.2.3 Blocking force test results .......................................................... 195
    6.7.2.4 Spring roll actuator performance characterization ..................... 196
6.8 Conclusions .................................................................................................... 198
CHAPTER 7 Conclusion and Future Perspective .............................................. 201
REFERENCES ..................................................................................................... 204
APPENDIX ......................................................................................................... 257
LIST OF TABLES

Table 2.1 Comparison of the properties of EAP, SMA and EAC [2]................................. 5

Table 2.2 Maximum response of representative elastomers [7]........................................ 56

Table 2.3 Summary of electrode options for dielectric elastomers [220]............................ 68

Table 3.1 Comparison among various rolled D-EAPs in literatures(reference[309]).......... 106

Table 5.1 Comparison of mechanical, electrical and actuation anisotropy of actuator with aligned CNT sheet electrodes and actuators with fiber reinforcement. In all but one case (as noted) the D-EAP material is VHB-4905 acrylic. ........................................................ 152

Table 6.1 Spring specifications......................................................................................... 158

Table 6.2 Specifications of monofilaments used to reinforce VHB composites .............. 160

Table 6.3 Dimensions of the spring roll actuator.............................................................. 177

Table 6.4 Number of layers in spring roll actuators with constant length....................... 180

Table 6.5 Dimensions of the spring roll actuator with CNT sheet electrodes ................. 197
LIST OF FIGURES

Figure 2.1 Classification of Electroactive Polymers [2]......................................................... 6

Figure 2.2 Classification of dielectric materials [17]................................................................. 7

Figure 2.3 Schematic illustration of crystal structures in $\beta$ phase of PVDF [24].............. 11

Figure 2.4 Electrostrictive graft elastomer. (a) schematic illustration of molecular structure of
electrostrictive graft elastomer; (b) bimorph actuator within inactivated (middle) and activated
states (left and right) made from electrostrictive graft elastomer [42]. .................................. 14

Figure 2.5 Different geometries of the synthesis of LCEs. (a) side-chain elastomers and (b)
main-chain elastomers [58]................................................................................................. 16

Figure 2.6 Schematic illustration of anisotropic phase of liquid crystal part transfers to the
isotropic phase with increasing temperature and dimensional change [61].................... 16

Figure 2.7 Deformation of polyelectrolyte gels under the influence of electric field [69], (a)
schematic illustration of bending mechanism; (b) bending motion of the gel in the electric field.
............................................................................................................................................. 18

Figure 2.8 Schematic illustration of the bending mechanism of a typical IPMC [78]......... 19

Figure 2.9 Molecular conformational changes of conductive polymers while redox reactions
[98]........................................................................................................................................ 23

Figure 2.10 Schematic illustration of CNT actuators. (a) an applied potential injects charge of
opposite sign in the two pictured nanotube electrodes which are in liquid or solid electrolyte;
(b) charge injection at the surface of a nanotube bundle; (c) a cantilever actuator in which two
layer of CNT sheets are separated by a piece of scotch tape operated in the aqueous NaCl
electrolyte [104]................................................................................................................... 24

Figure 2.11 Principle of operation of D-EAPs................................................................. 27

Figure 2.12 The dimension change of D-EAP under actuation........................................ 29
Figure 2.13 Schematic illustration of circular strain set-up and photographs of acrylic elastomer under the circular strain testing. ................................................................. 31

Figure 2.14 Stress-strain curves of VHB elastomers with different strain and relaxation [122] .................................................................................................................. 35

Figure 2.15 Dielectric constant as a function of frequency of acrylic elastomers with different prestrain [122] .......................................................................................... 35

Figure 2.16 Effect of prestrain on electric breakdown strength and voltage [116] ................... 36

Figure 2.17 Comparison of actuation strain of VHB films of different initial thickness prestrained to have the same initial thickness [122] .................................................. 37

Figure 2.18 Mechanical efficiency and loss tangent of acrylic elastomers at different levels of prestrain and frequency [121] ........................................................................... 38

Figure 2.19 Schematic illustration of the fabrication process of an IPN elastomer film. (a) an acrylic film before processing; (b) after 400% biaxial prestrain; (c) curable additives are added into the prestrained film and cured, forming interpenetrating network of a highly crosslinked polymer; (d) after the external stress is removed, the interpenetrating network preserves most of the prestrain of the acrylic film [130]. .............................................................................. 39

Figure 2.20 Film surface morphology during pull-in instability [136]................................. 40

Figure 2.21 Schematic illustration of the voltage-charge curve of a layer of an elastomer dielectric where $\Phi$ is electric voltage and $Q$ is stored charged in the electrodes [137]........ 41

Figure 2.22 Working principle of dielectric elastomers transducers and three types of failure mechanisms. (a) a membrane of dielectric elastomer subject to a voltage reduces thickness and expands area. ($\Phi$ denotes applied voltage and $\lambda$ is the deformation ratio). The voltage-stretch curve typically is not monotonic. (b)-(d) three types of dielectrics are differentiated by where the two curves $\Phi(\lambda)$ and $\Phi B(\lambda)$ intersect, where $\Phi B(\lambda)$ is the breakdown voltage as a function of stretch ratio that normally is a decreasing function [150]................................. 43
Figure 2.23 Ideal stress-strain curve and molecular structure of dielectrics with Type III failure type. (a) stress-strain curve of a membrane under biaxial stress; (b) fiber embedded in a compliant matric; (c) a network of polymers with folded domains; (d) a network of polymers with side chains; (e) a network of polymers swollen with a solvent [150].

Figure 2.24 The chemical structure of silicone elastomers.

Figure 2.25 The chemical structure of acrylic elastomers.

Figure 2.26 Young’s modulus as a function of temperature for the VHB 4910 and silicone hardened with 5% hardener [125].

Figure 2.27 Cyclic actuation of (a) silicone (b) acrylic elastomer VHB F-9437PC actuators [125].

Figure 2.28 Schematic illustration of the morphology of a polyurethane elastomer where the hard segments (hatched boxes) are embedded in the matrix of soft segments (thin lines) [162].

Figure 2.29 Chemical structure of thermoplastic SEBS elastomer [214].

Figure 2.30 The equivalent circuit of film shaped dielectric elastomer actuator.

Figure 2.31 Zigzag patterned gold compliant electrodes. (a) zigzag gold electrodes undergoing large strains; (b) structured electrodes [219].

Figure 2.32 Peak isotonic transverse strain vs. electric field for the different electrode materials and a pre-stress of (a) 19.6 kPa, (b) 29.4 kPa, (c) 39.2 kPa and (d) 49.0kPa. [222].

Figure 2.33 Comparison of high-speed actuators technologies Linear Actuators [247].

Figure 3.1 Dielectric elastomer actuator configurations [4,8].

Figure 3.2 Extender configuration for linear actuation [115].

Figure 3.3 Structure of a two-phase stretched-film microactuator [250].

Figure 3.4 Antagonistically-driven linear actuator (ANTLA) and backbone shaped bending devices driven by ANTLA. (a) working principle of ANTLA; (b) four working statuses; (c)
ANTLA integrated with output shaft; (d) backbone shaped bending devices driven by ANTLA. [253]

Figure 3.5 Bowtie configuration actuator. (a) Working mechanism of bowtie actuator; (b) silicone bowtie actuator; (c) multilayer double bowtie actuator with acrylic film [8].

Figure 3.6 Bowtie D-EAPs actuator used in biomimetic applications [247]. (a) acrylic bowtie shaped actuator in self-contained six-legged robot “FLEX”; (b) flapping wing thorax-type design using D-EAPs

Figure 3.7 Diamond-shaped actuator. (a) schematic of four-bar mechanism based actuator with “negator” [258]; (b) schematic of a compliant symmetric double slider-crank mechanism with elastic joints on the slider pivot [259]; (c) assembly and prototype of acrylic diamond-shaped actuators [257].

Figure 3.8 Different designs for the flexible frame. (a) hexagon structured frame; (b) racetrack shaped frame; (c) prototype hexagonal structured actuator with and without actuation [260]; (d) stress relaxation of the nylon frame induced loss of blocking force as a function of time [258].

Figure 3.9 Stack configuration. (a) schematic of D-EAPs stacked actuator before and after actuation [262]; (b) stacked actuator consists of alternating electrodes and elastomer [261]

Figure 3.10 Tactile display with stacked configuration. (a) the schematic drawing of the elastomer stack with patterned electrodes; (b) D-EAPs contracts at the actuated point with can inducing tactile sensation of finger; (c) passive matrix arrangement of the electrodes [262,263]

Figure 3.11 Prototypes of multilayer D-EAPs actuator [267]. (a) vibrotactile display for mp3-player; (b) peristaltic pump

Figure 3.12 Rigid electrode used in stack D-EAPs actuators. (a) disadvantageous bulging out of middle layers of actuator with compliant electrode [268]; (b) D-EAPs stack actuator with fixed end [261]; (c) multilayer PDMS actuator with rigid doped silicon electrode [269]; (d)
general deformation morphology of elastomer with rigid and perforated electrodes, symmetrical setup (up) and asymmetrical setup (bottom) [270] ................................................................. 85

Figure 3.13 Stacked actuators with different configurations. (a) circular shape [271]; (b) rectangular shape [271]; (c) trapezoidal shape [274]; (d) strain versus electrical field of three types of actuators [275] ..................................................................................................................... 87

Figure 3.14 Robot finger driven by multi-stacked actuators. (a) mechanisms of one degree of freedom finger and multi-joint finger [274]; (b) performance of robot finger [272,276]........ 88

Figure 3.15 Helical “polymer spring” actuator. (a) schematic illustration of structure and operation of a helical D-EAPs actuator [290]; (b) prototype of spiral actuator, the “skeleton” consists of spiral silicone and carbon loaded silicone electrode (top); after filled skeleton with silicone (bottom) [289] ..................................................................................................................... 90

Figure 3.16 The helical actuator fabrication process. (f & a) a D-EAP tube; (b & g) cutting tube into helical shape; (c) application of electrodes; (d) assembling of a naked helix and a coated helix together to form a “tube”; (e & i) final assembly. [291].......................................................... 91

Figure 3.17 Eyeballs driven by helical linear contractile actuators. (a) prototype of eyeballs for android robot face; (b) schematic drawing of the configuration of eyeball muscles [292].... 92

Figure 3.18 Folded D-EAPs actuator configuration. (a) schematic drawing of folded D-EAPs actuator and prototype [293]; (b) axial strain as a function of the applied filed [293]; (c) folded actuator with circular cross-section [294]..................................................................................................................... 93

Figure 3.19 Application of folded D-EAPs actuators. (a) android eyeballs [295]; (b) hand rehabilitation splints [297]; (c) bi-direction tilter [296]; (d) deployable boom [296]; (d) man-machine interface ..................................................................................................................... 94

Figure 3.20 Fabrication process of spring roll actuator [12].......................................................... 96

Figure 3.21 The working principle of D-EAPs spring roll actuator[13,118]. (a) four states of the spring roll actuator based on quasi-linear mechanical behavior of D-EAP [118]; (b) the
force-displacement of D-EAP with non-linear mechanical behavior [13]; (c) passive equilibrium of loads in different directions in the spring roll actuator [13] .............................. 97

Figure 3.22 Walking robot driven by spring roll actuators [12] .............................. 99

Figure 3.23 Tube-spring actuator. (a) tube-spring actuator with silicone tubes enclosed compressed spring; (b) multi-fingered robot hand driven by tube-spring actuators [301] ..... 99

Figure 3.24 Portable force feedback glove by connecting actuator between fingers [13,118]. ................................................................................................................. 100

Figure 3.25 Prototype and working principle of the actuator bundles as the robot arm [11]. (a) wrestling robot with four actuator banks, each has 64 actuators; (b-c) the working principle of the robot arm ............................................................................................................. 101

Figure 3.26 Multiple degree-of-freedom (DOF) spring roll actuator. (a) 2-DOF [302,303]; (b) 3-DOF [303] .................................................................................................................. 102

Figure 3.27 Core-free and self-supporting roll actuator [306] ........................................ 103

Figure 3.28 Fiber reinforced D-EAPs roll actuator [307]. (a) aligning nylon filaments to reinforce VHB; (b) laying the second layer of VHB on the surface of the first fiber reinforced one; (c) applying carbon grease on both surfaces of laminates and rolling; (d) complete actuator with leads ................................................................................................................. 104

Figure 3.29 External prestretching shell and roll actuator [309]. (a) fabrication process of core-free silicone roll actuator; (b) cross-ply laminates of carbon fiber reinforced polymer (CFRP) shells with different inclinations; (c) assembly of a rolled actuator and a CFRP shell; (d) integration of actuator with a thoracic mechanism to form an insect-inspired wing flapper 105

Figure 3.30 Theoretical distribution of mechanical compressive radial pressure on the layers in rolled D-EAPs actuators consists of N film layers [11] ......................................................... 109

Figure 3.31 Structural drawbacks of spring rolls actuator. (a) free-standing spring roll actuator shows obvious bent shape [12]; (b) spring roll actuator with telescopic core [13,118] ....... 109
Figure 3.32 Progress diagram of automatic rolling up stage for assembling core-free D-EPAs roll actuators [312].................................................................................................................. 110

Figure 3.33 Fiber-like cylindrical actuator. (a) schematic of the fiber actuator;(b) axial actuation (about 10%) strain produced in uniaxially prestretched silicone based prototype [313]. .................................................................................................................................................. 111

Figure 3.34 Coextruded D-EAPs fiber actuators. (a) coextrusion mechanical schematic diagram; (b) coextruded fiber cross section; (c) an example of an assemble single fiber actuator; (d) an example of an assembled rope actuator with the ends capped with an epoxy plug. [315] .................................................................................................................................................. 112

Figure 3.35 Working principle of cone shaped D-EAP linear actuator. (a) Prototype with and without electrically stimulation [318]; (b) schematic drawing of the working principle [319]. .................................................................................................................................................. 113

Figure 3.36 Passive and active stiffness curves with various bias mechanism [316]........... 114

Figure 3.37 Biasing mechanisms in cone shaped actuators. (a) spring [316,319]; (b) weight [316]; (c) diamond-shaped four-bar linkage mechanism [323]; (d) film [316,324]; (e) flexural pivot [318]; (f) double-slider [320]; (g) parallel mechanism [325]; (h) revolute [325]; (i) rigid strut [325]; (j) bi-stable negative rate mechanism [319]................................................................................................. 115

Figure 3.38 Force and stroke operating profiles for various applications [316]................. 116

Figure 4.1 Fabrication of actuators. (a) Drawing of CNT sheet from forest with SEM image of the sheet as inset. (b) Schematic illustration of VHB-CNT actuator fabrication (top), and the operational principle of the actuator before (middle) and after (bottom) activation, (c) Optical micrograph of the electrode area of an actuator................................................................................................................................. 119

Figure 4.2 Cyclic tensile stress-strain curves of VHB-CNT actuators obtained by loading and unloading to 25% strain amplitude, (a) along the fiber (y) direction, (b) along the cross (x) direction. For clarity, data for 1st (●), 2nd (▲), and 25th (■) cycles are shown. (c) y-direction
stress-strain diagram of the 25th cycle for 6 (●), 25 (▲), and 40% (■) strain amplitudes. The solid lines connect the data in all cases.

Figure 4.3 Optical images of electrodes showing morphological changes of CNT sheet electrodes, (a) sample under 10% tensile strain in the y-direction showing fracture, the inset shows bridging fibers across lines of fracture, (b) sample after 25 cycles of 25% strain along the y-direction. Optical and SEM images (as insets) of, (c) CNT fiber clusters of tightly folded undulations and kink bands (circled area) in strain-cycled sample as in (b), (d) sample after 25 strain cycles of 25% strain in the x-direction.

Figure 4.4 Electrical resistance of the CNT sheet electrodes measured along y (■) and x (●) directions, plotted as a function of applied strain in the, (a) y-direction, and (b) x-direction. The solid lines connect the data, and the error bars represent one standard deviation around the mean.

Figure 4.5 Linear actuation strain plotted, (a) as a function of nominal electric field in the fiber (○) and cross-fiber (●) directions together with directionally uniform response of a similar actuator with carbon-grease electrode (■), (b) as a function of actuation cycle number for actuators at 50 and 80 V μm⁻¹ electric fields as labeled. The inset of (a) shows the variation or tensile modulus of the VHB-CNT samples as a function of cycle number. The solid lines represent exponential fits to the data in (a) and the same in (b) is used to connect the data. Video capture images illustrate a test specimen, (c) before actuation, (d) at 80 V μm⁻¹, and (e) at 100 V μm⁻¹.

Figure 4.6 Linear actuation strain in the cross direction (x) for the initial 220 cycles of the 370 cycles.

Figure 4.7 Self-reconfiguration of the CNT electrodes. The bar chart shows change in electrical resistance of CNT electrodes, along fiber (y) and cross-fiber directions, measured at various stages (as labelled) using a four-point probe set up. Insets (a), (b), and (e) are optical micrographs of CNT electrodes after 25 cycles of actuation, after 25 cycles of actuation and cleaning, and after 25 cycles of mechanical cycling at 25% strain followed by cleaning,
respectively. Insets (c) and (d) are SEM (surface) and TEM (cross-sectional) images of CNT electrodes, respectively, showing penetration of CNT fibers in the elastomer after 25 cycles of actuation.

Figure 5.1 Schematics illustration of (a) operational principle of D-EAP actuators, (b) microcombing (on the left) and laser ablation (on the right) processes. The laser ablation process is used to selectively remove CNT fibers along 360 μm wide lines from the CNT sheet on the surface of VHB actuator, leaving stripes of 180 μm width.

Figure 5.2 Optical and scanning electron micrographs of CNT sheet electrodes. (a), (b), and (c) are optical microscopic images of as-drawn, combed, and etched samples, respectively. (f) is optical microscopic with higher magnification of etched area in the etched sample. (d) and (e) are the SEM images of as-drawn, combed samples, respectively.

Figure 5.3 Stress-strain curves of cyclic uniaxial tensile test of actuators obtained under 40% strain amplitude. For clarity, the data for only the 1st cycle (a) along fiber (y), 2nd cycle (b) along y-directions, and 1st cycle (c) along cross-fiber (x) direction of as-drawn (●), combed (▲) as well as etched sample (■), are shown. Additionally, the 1st cycle stress-strain plot (- - -) of the pristine VHB film is shown in (a). Optical micrograph of the as-drawn sample under 20% tensile strain in y-direction shows the “turtle shell-like” morphology.

Figure 5.4 (a) Electrical resistance of the as-drawn, combed and completely etched CNT sheet electrodes, measured along the fiber (y) and cross-fiber (x) directions. Images of (b) as-drawn, and (c) completely etched CNT sheet electrodes mounted on a printed paper to demonstrate optical transparency.

Figure 5.5 (a) Linear actuation strain plotted as a function of electric field of actuators with as-drawn (●), combed (▲) as well as etched (■) CNT sheet electrodes. For comparison, actuator with carbon grease (♦) electrodes is included. (b) Magnified view of (a) in electrical range of 80 V μm⁻¹ to 120 V μm⁻¹. Video capture images of actuators before actuation with (c) as-drawn, (d) combed, (e) etched CNT electrode, and actuators at 110 V μm⁻¹ with (f) as-drawn, (g) combed, (h) etched CNT electrodes.
Figure 6.1 (a) Load-displacement curve of compression spring; (b) stress-strain curves of unidirectional tensile tests of monofilaments used to reinforce VHB composites. Inset is tensile young’s modulus results. ............................................................... 159

Figure 6.2 Schematic illustration of fiber reinforced VHB composites cross-section within 3mm length. ........................................................................................................................................... 160

Figure 6.3 Preparation of spring roll actuator. (a) Schematic illustration and (b) photographs of fabrication process. .................................................................................................................................................. 162

Figure 6.4 Unidirectional tensile test results of VHB and its composites. Stress-strain curves of strain alone (a) fiber direction (y) and (b) cross-fiber direction(x). (c) Initial modulus of composites straining in both fiber(y) and cross-fiber(x) directions......................................................... 165

Figure 6.5 Photographs of cutting off edge of composites located above the top clamp before tensile testing. (a) PA-I reinforced composites, (b) PA-II reinforced composites and (c) PU reinforced composites .......................................................................................................................................... 165

Figure 6.6 Stress supporting the prestrained VHB. It is recorded in tensile testing after mounting the sample, clamping samples and zeroing load cell, then the force was recorded after cutting off the film above the top clamp......................................................................................... 166

Figure 6.7 (a) Linear actuation strain as a function of true electric field of flat circular actuators. (b)-(e) is actuation video capture of VHB, PA-I/VHB composites, PA-II/VHB composites and PU/VHB composites actuator before actuation, respectively; (f)-(i) is capture of them at about 110 V/μm. ................................................................................................................................................. 168

Figure 6.8 Blocking force testing results of PU fiber reinforced composites. (a) force-time curve; (b-c) photographs of actuator before and after actuation, respectively. ............... 170

Figure 6.9 Blocking stress as a function of true electric field of fiber reinforced composites ......................................................................................................................................................... 170

Figure 6.10 Working principle of spring roll actuator (redrawn from reference [385])...... 172
Figure 6.11 Schematic illustration of process to preparing elastomer roll and the stresses in coordinates ........................................................................................................................................ 173

Figure 6.12 Load-length curve of spring and inactive VHB roll ........................................... 176

Figure 6.13 Photographs of (a) compressed spring and (b) spring roll actuator made of VHB elastomers. (c) Radial pressure \( \sigma_{elst} \) as a function of radius in the spring roll actuator (assuming each layer thickness \( t \) is 0.055mm with \( \sigma_h = 0.468 \text{MPa} \) and spring outside diameter \( r_i \) is 8.63mm) ................................................................................................................... 178

Figure 6.14 Cross-sectional images of spring roll actuator made of (a) neat VHB and (b) fiber reinforced composites. (c) Cross-sectional view of flat spiral with inner radius of \( r_i \), outside radius of \( r_o \), thickness of \( t \) and distance between neighboring layers is zero. ...................... 179

Figure 6.15 Radial pressure of elastomers as a function of elastomer radius position of spring roll actuators made of neat VHB and fibers (PA-I, PA-II and PU) reinforced composites. 181

Figure 6.16 Schematic illustration of activated elastomer unit cell in roll and the stresses in coordinates ........................................................................................................................................ 182

Figure 6.17 Force equilibrium in spring roll actuator at (1) passive equilibrium, (2) initial moment of activation, (3) active equilibrium, and (4) initial moment of deactivation ........... 184

Figure 6.18 Radial pressure of elastomers as a function of elastomer radius position of spring actuators in passive and active status \( (U=5 \text{kV}) \) .............................................................................................................................. 186

Figure 6.19 (a) Field-induced actuation strain in longitudinal direction of spring actuator made of VHB and its fiber reinforced composites; (b) video captures of PA-II monofilament reinforced VHB spring roll actuator before actuation and upon 4kV electric potential. ...... 187

Figure 6.20 Load-length curve of compressing spring, tensioning inactive and activated neat VHB, VHB/composites roll ............................................................................................................ 189

Figure 6.21 Schematic illustration of structure of one layer, 2 layers and 6 layers flat film actuators ........................................................................................................................................ 192
Figure 6.22 Unidirectional tensile test results of CNT sheet electroded VHB alone (a) fiber direction(y) and (b) cross-fiber direction(x). ................................................................. 193

Figure 6.23 (a) Stress supporting prestrained elastomer of neat VHB sample, 2 layers VHB with 3 layers CNT sheets electrodes sample and 6 layers VHB with 6 layers CNT sheets electrodes sample. (b) Photograph of cutting off area located above top clamp before tensile testing of 2 layers CNT-VHB sample. ............................................................................ 194

Figure 6.24 Linear actuation strain as a function of true electric field of flat film actuator of 1layer, 2 layers and 6 layers structure with (a) aligned CNT sheets electrodes and (b) carbon grease electrodes. ............................................................................................................. 195

Figure 6.25 Blocking stress as a function of true electric field of VHB actuators with CNT sheets electrodes and carbon grease electrodes. ............................................................... 196

Figure 6.26 Radial pressure of elastomers as a function of radius position of spring actuators with CNT sheets and carbon grease electrodes in inactive and active status (U=5kV) ....... 197

Figure 6.27 Field-induced actuation strain in longitudinal direction of neat VHB spring actuator with CNT sheets electrodes and carbon grease electrodes. ................................. 198
CHAPTER 1 Introduction

1.1 Introduction

Actuators are used to convert a form of input energy into motion. Typical input energy may be one of electrical, thermal, magnetic, chemical, or compressed fluid (air/water/oil). Due to the diversity of forms of input energy, actuators are highly versatile. Traditional electromagnetic motors, pneumatic, and hydraulic actuators, are used extensively in many everyday applications. However, these traditional actuators are generally bulky, heavy, noisy, and rigid, therefore not particularly suitable for many of today’s applications such as microrobotics, mobile devices, and soft electronics that often require high power/torque to mass ratio, direct-drive, conformability, and small form factor.

Materials based actuation technologies such as, piezoelectric ceramics, shape memory alloys, and electroactive polymers, seem to offer advantages that are definitely attractive in many ways. Electroactive polymers (EAP), as one of the most promising emerging materials for actuator applications, exhibit deformation when subjected to an electric potential or field. EAPs are relatively lightweight, soft, conformable, inexpensive, and are available in a broad range of mechanical and electrical properties. Unlike their inorganic counterparts (piezoelectric ceramics and shape memory alloys, etc.), they are easy to process and are more amenable to shaping and alteration of properties through various means. As a result EAPs are relatively easy to impedance-match[1,2]. In applications, EAPs have been explored as actuators as well as generators by taking advantage of their reversible electromechanical response.
Among the various EAP materials, dielectric electroactive polymers (D-EAP) offer many distinct advantages, including high energy density, remarkably high actuation strain, moderate stress, and tunability of properties [3–7]. In order to meet the actuation requirements of different applications, various configurations of actuators based on D-EAPs have been proposed. Among them, linear actuators are able to directly deliver a contractile/extensive stroke. This particular mode of actuation is required for a variety of application, such as simulation of human muscle motions [8–11]. Spring roll actuator, as one type of linear D-EAP actuators, consists of a compressed spring in the core and wound D-EAP roll as shell [12,13], is the focus of this work.

D-EAPs consist of physically or chemically cross-linked macromolecular networks [14] and are mechanically isotropic. Under an applied electric field, the in-plane expansions are uniform in every direction. However, in many actuator applications that require directional electromechanical response [15], it is necessary to use other complex means to direct the stress/strain in the preferred direction.

In this work, a simple bifunctional carbon nanotube (CNT)-based electrode is developed that serves both as an electronic conductor and a mechanically anisotropic constraint to produce directional actuation of the D-EAP. This work also explores the effect of improving alignment of CNT fibers in the sheet electrodes through microcombing and selective laser ablation of combed CNT electrodes for application in D-EAP actuators to tune anisotropic electromechanical response. Moreover, in this work, application of anisotropic D-EAP actuators in spring roll actuator is investigated.
This dissertation is organized in most parts as research papers that have been or will be published with minimum changes.

Chapter 1. Introduces the contents covered and their organization in this dissertation

Chapter 2. Contains the review of fundamental electromechanical transduction principles and material categories of electroactive polymers (EAPs), with particular emphasis on the dielectric electroactive polymers (D-EAP).

Chapter 3. Reviews the linear D-EAPs actuators on the basis to their configurations, discusses their working principle, performance characteristics, processing techniques, and potential applications.

Chapter 4. Demonstrates the bifunctional carbon nanotube (CNT)-based electrode that serves both as an electronic conductor and a mechanically anisotropic constraint to produce directional actuation of the D-EAP.

Chapter 5. Explores the effects of improving alignment of CNT sheet through microcombing and selective laser ablation for application in D-EAP actuators to enhance anisotropic electromechanical response.

Chapter 6. Investigates different types anisotropic D-EAPs and their application in spring roll actuators. Analytically and experimentally examines ways to improve actuation performance of spring roll actuator.

Chapter 7. Reports summary of this research and potential subjects of future research.
CHAPTER 2 Electroactive Polymers

2.1 Introduction

Electroactive polymers (EAP) form a distinct class of polymers within the stimuli responsive polymers that undergo dimensional change upon the application of an electric field. Over the past few decades, EAPs have attracted tremendous attention of researchers from different disciplines because of their broad range of electromechanical properties, ease of processing and diverse potential applications in contemporary technologies. [2,16].

Electroresponsive inorganic materials, such as electroactive ceramics (EAC) and shape memory alloys (SMA), have been extensively used for several decades. Although these materials produce large actuation stresses, they have many inherent drawbacks. For comparative purpose, the key properties of SMAs, EACs, and EAPs are presented in Table 2.1. SMAs, produce large stress and acceptable strain, but have long response time and the short cycle life. Similarly, EACs produce high actuation stress, but are mechanically fragile and limited by the very small strain [14]. In contrast, EAPs are generally superior in terms of relatively large actuation strain, low density and resilience. However, EAPs are limited in robustness and require high electrical field or drive voltage [2].
Table 2.1 Comparison of the properties of EAP, SMA and EAC [2].

<table>
<thead>
<tr>
<th></th>
<th><strong>EAP</strong></th>
<th><strong>SMA</strong></th>
<th><strong>EAC</strong></th>
</tr>
</thead>
<tbody>
<tr>
<td>Actuation strain (%)</td>
<td>&gt; 10 (up to 380(^a))</td>
<td>&lt; 8 short fatigue time</td>
<td>0.1-0.3</td>
</tr>
<tr>
<td>Generated stress (MPa)</td>
<td>0.03-43(^a)</td>
<td>about 700</td>
<td>30-40</td>
</tr>
<tr>
<td>Response speed</td>
<td>µsec to min</td>
<td>sec to min</td>
<td>µsec to sec</td>
</tr>
<tr>
<td>Density (g m(^{-3}))</td>
<td>1-2.5</td>
<td>5-6</td>
<td>6-8</td>
</tr>
<tr>
<td>Stimulation electric potential/filed</td>
<td>2-7 V (Ionic EAP)</td>
<td>5V</td>
<td>50-800 V</td>
</tr>
<tr>
<td></td>
<td>10-150 V/µm (Electronic EAP)</td>
<td></td>
<td></td>
</tr>
<tr>
<td>Consumed power *</td>
<td>milliwatts</td>
<td>watts</td>
<td>watts</td>
</tr>
<tr>
<td>Fracture toughness</td>
<td>resilient, elastic</td>
<td>elastic</td>
<td>fragile</td>
</tr>
</tbody>
</table>

*Note: the power consumption was estimated for devices that are driven by such actuators; \(^a\) updated according to the data in reference [7]*

The variety of EAP materials are broadly divided into two categories based on their electroresponse mechanisms: the electric type (or electronic type) and the ionic type. Within each type of EAPs there are many subclasses [2], see Figure 2.1. In the following, these EAPs are discussed in terms of their working mechanisms, dominant properties and primary applications.
2.1.1 Electric EAPs

Electric (or electronic) EAPs are a class of the polymers that response due to electrostatic or Coulomb forces developed upon application of an electric field. Electric EAPs are attractive due to their ability to serve as both solid-state actuators and sensors [2]. In this section, four fundamental electromechanical principles relevant to electric EAPs are introduced, before exploring the specific material types.
2.1.1.1 Electromechanical Principles

Piezoelectric and Electrostrictive Effects

Piezoelectric materials constitute one specific type of dielectric material (see Figure 2.2), which have non-centrosymmetric crystal structure. Dielectric crystal lattice could be considered as that consisting of anions, cations and interionic chemical bonds. Under an electrical field, anions and cations undergo asymmetric displacement which causes change in crystals dimension (except the octahedral class of crystals) [17].

![Figure 2.2 Classification of dielectric materials](image)

Piezoelectric effect is reversible and can be divided into the direct and the indirect effect according to the direction of coupling. Direct piezoelectric effect describes conversion from mechanical energy into electrical energy and vice versa. These effects are described in Equation 2.1 and Equation 2.2. Equation 2.1 demonstrates the indirect piezoelectric effect where \( S \) and \( E \) denote the mechanical strain and the electrical field, respectively. Conversely, Equation 2.2 illustrates the direct piezoelectric effect, where \( D \) and \( T \) denote the electric
displacement (or charge density $D$) and the mechanical stress, respectively. The variable $d$ in both represents the piezoelectric coefficient [17].

\begin{align*}
S &= dE \quad \text{Equation 2.1} \\
D &= d\tau \quad \text{Equation 2.2}
\end{align*}

With inherent transduction feature between electrical and mechanical energy, piezoelectric effect has a wide variety of applications such as generators and detectors of ultrasonic waves, actuators and mechanical deformation sensors etc.

Importantly, piezoelectric effect is a linear electromechanical effect that is different from the electrostrictive and Maxwell stress effects in which strain is coupled to the electrical field nonlinearly [17]. This will be elaborated further in the following.

Unlike piezoelectrics, electrostrictive materials are centrosymmetric (inversion symmetry) dielectrics. Due to this feature, movements of anions and cations offset between the nearby chemical bonds and thus in principle the deformation of crystal is close to zero. However, in practice, the chemical bonds are not perfectly harmonic and that results in the second order effect so that the lattices may have a slight deformation. This deformation is proportion to the square of electrical field and is termed as the electrostrictive effect. For instance, the increase in polarization induces a contractive deformation along the polarization direction. Since the chemical bond inharmonic structure exists universally in dielectrics, the electrostriction effect is believed to play a role in the electroresponse of all of the dielectrics. [2,17].

Equation 2.3 shows the nonlinear electromechanical relation of the electrostrictive effect where $X, Q, \epsilon_0,$ and $\epsilon_r$ are the mechanical stress, the material polarization, the free space electric
permittivity \((\varepsilon_0 \approx 8.85 \times 10^{-12} \text{ farads/meter})\), and the material’s relative permittivity, respectively.

\[
X = Q \varepsilon_0^2 \varepsilon_r^2 E^2 \quad \text{Equation 2.3}
\]

**Maxwell Stress Effect**

The electrostatic effect in dielectric media is referred as Maxwell stress effect. In other words, the mechanical response of dielectrics, upon application of an electric field that results from electrostatic force is termed as Maxwell stress. It is proportional to the square of electric field magnitude. Normally, the strain induced by the stress is express as Equation 2.4 [18]. Maxwell established this theory based on the attraction between two parallel rigid plates in a capacitor system[19].

\[
\sigma = \varepsilon_0 \varepsilon_r E^2 = \varepsilon_0 \varepsilon_r \left(\frac{V}{z}\right)^2 \quad \text{Equation 2.4}
\]

where \(\sigma\) is the Maxwell pressure, \(\varepsilon_0\) and \(\varepsilon_r\) is the free space electric permittivity and the material’s relative permittivity respectively, \(V\) is the applied voltage and \(z\) is the thickness of elastomer film \(\left(\frac{V}{z} = E\right)\).

In an isotropic free-standing D-EAP, the strain due to Maxwell stress in-plane can be expressed as Equation 2.5 where \(S_{\text{in-plane}}\) is the strain and \(s\) is the compliance of the elastomer. According to this equation, mechanical compliance or modulus of the elastomers has a tremendous effect on the responsive strain driven by Maxwell stress.

\[
S_{\text{in-plane}} = -\frac{1}{2} s \varepsilon_0 \varepsilon_r E^2 \quad \text{Equation 2.5}
\]
For anisotropic materials with the nonlinear elastic properties, however, it is necessary to account for the mechanical instability of the elastomers. These two effects in a dielectric medium; electrostrictive and electrostatic (Maxwell effect) both exhibit a quadratic dependence on the applied electric field. And their differences deserve further discussion. As mentioned earlier, electrostrictive stresses are produced by polarization of charged elements under the electrical field and therefore mostly responsible for small deformations, in D-EAPs. Maxwell stress, on the other hand, is produced by the electrostatic force and is the major response mechanism for most D-EAPs that produce large deformations under an electric field. So it has been widely used to predict or analyze electrical field induced deformation of D-EAPs. However, Zhao et al. pointed out that Maxwell stress effect is only applicable for very specific type of materials, described as “ideal dielectric elastomer,” in which the permittivity is deformation independent [20]. Molecular units in these D-EAPs are expected, in principle, to possess relatively low cross linking densities and deformations smaller than their extension limitations (polarize freely as in liquid state). The permittivity of this ideal D-EAP thus does not alter with the shape change.

2.1.1.2 Martials

Ferroelectric Polymers

Ferroelectric materials constitute a subclass of pyroelectric materials that possess both piezoelectric and pyroelectric properties. The polarization of ferroelectric materials is spontaneous and reversible [17]. In the past half century, various ferroelectric polymers have emerged, including but not limited to poly(methyl methacrylate), poly(vinyl chloride),
poly(vinylidene fluoride) (PVDF), copolymers and blends of PVDF, such as polyvinyl fluoride (PVF), polyamides (odd-numbered nylons), cyanopolymers, copolymers of vinylidene fluoride and vinyl fluoride polymers, polyureas, copolymers of PVDF with trifluoroethylene (TrFE) or tetrafluoroethylene (TFE), polythioureas, various biopolymers (polypeptides and cyanoethyl cellulose), ferroelectric liquid crystal polymers and polymer-ceramic composites etc. [21]. Among them, PVDF is the most investigated one.

One of the most common ferroelectric polymers, PVDF, has a very simple chemical structure (–CH2-CF2–). It has four known chain conformations or polymorphs. Among them, the β crystalline phase is most attractive form because of its piezoelectric and ferroelectric characteristics. It is formed when PVDF is mechanically deformed (or stretched) near its melting point. The all trans (TT) conformation of the β polymorph has a large dipole moment because all of the fluorine atoms locate at the same side of the carbon, see Figure 2.3. This non-centrosymmetric structure is ideal morphology of piezoelectric crystals. The dielectric constant of PVDF is notably high, normally in the range of 6-12 [22,23].

![Figure 2.3 Schematic illustration of crystal structures in β phase of PVDF [24].](image)

According to the theoretical simulations, it was found that in high defect concentration situation all-trans configuration of PVDF is more stable [25,26]. Copolymers of vinylidene
fluoride with trifluoroethylene, tetrafluoroethylene or hexafluoropropylene and their blends have been widely investigated and explored [21,22,24,27–30]. It is important to note that \( \beta \) phase in PVDF, does not guarantee the piezoelectric behavior unless the crystals are oriented in a certain way. Commonly, a high electric field or “poling filed” is needed to orient the crystal domains. In addition, corona discharge also has been used crystals poling. With different methods of poling, the resulting polymeric structure, ferroelectricity and chemical bond formation are different [16,31].

To further improve the electromechanical response performance, the electron irradiated poly(vinylidene fluoride-trifluoroethylene) [P(VDF-TrFE)] with improved strain response (about 4% strain) has been proposed [32,33]. After electron irradiation, the materials showed typical relaxor ferroelectric behavior. The electromechanical responses of the polar region combined with the difference of lattice strain between polar and nonpolar give the higher strain response. Later, copper-phthalocyanine (CuPc) oligomer has been introduced into P(VDF-TrFE) as fillers that dramatically enhanced the dielectric constant by about 5 times [34]. Xu et al. investigated the ferroelectric properties of poly(vinylidene fluoride-trifluoroethylene-chlorotrifluoroethylene) terpolymer [P(VDF-TrFE-CTFE)] and found that chlorotrifluoroethylene lead to the disordered ferroelectric phase, resulting in ferroelectric relaxor behavior [35]. Photo cross-linking method has also found to significantly enhance the energy density (the definition refers to section 2.2.1.4) of P(VDF-TrFE-CTFE) up to 22.5 J/cm\(^3\) at electrical field of 400 V/\( \mu \)m owing to the reduced crystal size and intensive interface effect [36].
Introduction of high dielectric constant ceramics (e.g. PbTiO$_3$, BaTiO$_3$) in ferroelectric polymers in the form of composite leads to increasing permittivity and piezoelectric coefficient [37–39]. Other filler materials such as carbon nanotubes (CNT) [28] and palladium nanoparticles [41] have also been used in PVDF for enhancing dielectric behavior.

Ferroelectric polymers, such as PVDF and its copolymers, combine pyroelectric and piezoelectric behavior and thus have great potential use in applications such as biomedicine, energy generation and storage, filtration, as well as in sensors and actuators. [24].

**Electrostrictive Graft Elastomer**

Electrostrictive graft elastomers consist of flexible backbone and grafted polar side groups, see Figure 2.4 (a) [42]. The grafted polar groups form crystalline regions and work as physical cross-links in the molecular network. For example, chlorotrifluoroethylene-vinylidene fluoride has been used as the flexible backbone and vinylidene fluoride-trifluoroethylene copolymer was employed as the grafted side chains [43]. Under application of electric field, the crystalline side chains align and the backbone chains reorient that lead to dimensional change in the polymer system [44].
Figure 2.4 Electrostrictive graft elastomer. (a) schematic illustration of molecular structure of electrostrictive graft elastomer; (b) bimorph actuator within inactivated (middle) and activated states (left and right) made from electrostrictive graft elastomer [42].

Electrostrictive graft elastomers generally have moderate to large actuation strain (~4% under electrical field of 120 V/μm) and high mechanical modulus (550-700MPa) [42]. As a result, electrostrictive graft elastomers have very high strain energy density (refer to section 2.2.1.4 for definition) (247 J/kg) compared to electrostrictive polyurethane elastomers (87 J/kg) [42]. Electrostrictive graft elastomers have been used in composites to enhance the toughness of the copolymer and thereby increasing the force output of the system [45].

**Electroactive Paper**

Electroactive paper (EAPap) was discovered serendipitously by Jaehwan Kim from Inha University, in Inchon, South Korea [46]. While they pasted two silver coated papers together, there was the displacement observed surprisingly by applying electric field between the two outside layers of silver coating [47]. This has been a very exciting find due to environmental friendly and sustainable natures of cellulose based papers [48]. The actuation of EAPap is due to a combination of electrostrictive effect and ionic migration of cellulose [48–50]. The
absorbed water plays an important role in strain response by inducing a non-uniform electric field and thus generating a dielectrophoretic force [51].

EAPap reportedly requires low stimulating electrical field (electrical field of 2 V/μm to induce a about 3mm tip displacement of a beam actuator) and consume relatively low power consumption (a couple of ten mW/cm² of electrical power) which is very promising for microwave driven actuators) [50,52]. However, EAPaps, produce limited and unstable displacement as well as force [53], low strain energy density (about 0.4 J/kg ([54])), are sensitivity to humidity, and have short life time etc. [48,55–57]

**Liquid Crystal Elastomers**

Liquid crystal elastomers (LCEs) are polymers having weakly cross-linked backbones with attached polymeric liquid crystals or mesogenic ('rod like' or anisotropic structures, with one axis appreciably longer than the other). LCEs combine the elastic properties of elastomers and the ability of self-organization of liquid crystals together and thus they manifest very unique physical properties [58]. They exhibit shape change due to change in phase and orientation of the liquid crystals under the influence of an electric field.

LCE networks routinely have two configurations based on the combination styles of mesogenic chains and backbones: side-chain elastomers and main-chain elastomers, see Figure 2.5. To orient the liquid crystal components, there are two main approaches. The first one is mechanical alignment that benefits from long chain features of polymers. With the help of mechanical stretching, a global macroscopic orientation of liquid crystal phase is formed. The second approach is applying magnetic or electric field to orient mesogenic units [58].
The electroactive response of LCEs depends on their morphology. LCEs with ferroelectric phase are triggered by the electroclinic effect in which the tilt angle of mesogens changes and hence causes deformation. In some cases, the electric field induced strain occurs in highly swollen nematic LCEs owing to nematic-isotropic (NI) phase transition (see Figure 2.6). Besides these two principles, the third actuation mechanism to use the electric power as a tool to generate thermal energy to trigger LCEs by nematic to isotropic phase transition, for instance, to add conductive wire ([59]), carbon black ([60])/CNT into LCEs.

Figure 2.5 Different geometries of the synthesis of LCEs. (a) side-chain elastomers and (b) main-chain elastomers [58].

Figure 2.6 Schematic illustration of anisotropic phase of liquid crystal part transfers to the isotropic phase with increasing temperature and dimensional change [61].
The main-chain elastomers are expected to generate larger strains compared to side-chain elastomers. This speculation was confirmed experimentally [61]. The main-chain elastomers show up to 400% strain while shape transition [62] and it followed by 70%[63] and 40% strain [64] of side-chain elastomers (side-on type and end on type, respectively). In general, the LCEs are low modulus material and the low actuation strains lead to low work density. They also suffer from high dielectric loss and low efficiency [65].

### 2.1.2 Ionic EAPs

Ionic EAPs are a class of EAPs driven by significant volumetric changes through insertion and expulsion of counter-ions during reduction and oxidation reactions (redox cycling) through exchange of ions with electrolytes. As a result, the ionic EAPs require presence of fluid in the system. Ionic EAPs require low stimulating voltage (several volts as Table 2.1 shows) in contrast to electric EAPs. However, they have long response time and do not hold strain well. Thanks to this advantage, ionic EAPs have potential to be explored in battery-driven human-friendly devices by which the high electric field safety issues are no longer an obstacle [66]. Ionic EAPs reported in the literature include ionic polymer gel, ionic polymer-metal composite, conductive polymers, and carbon nanotubes.

#### 2.1.2.1 Ionic Polymer Gels

Ionic polymer gels (IPG), consist of cross-linked elastic networks and a fluid filling the spaces of the network. IPGs undergo considerable swelling or contraction upon an external stimulation using temperature, pH, and electric field etc. IPGs are soft and wet but with solid-like appearance, like living organisms (e.g. mammalian tissues, the insider layer of skin).
Accordingly, ionic polymer gels have a huge potential to be used for synthetic organisms. The electroresponse (swelling and deswelling) of ionic polymer gels in an applied electric field is attributed to transport of hydrated ions and water in or out the polymer network [67–69].

In 1982, Tanaka et al. first reported the phenomenon of partial shrinking of rod-shaped samples of acryl acid-acrylamide copolymer gel, placed between electrodes. They explained the mechanism as a movement of negatively charged side of the gel toward the anode [70,71]. Later on it was determined that the dimensional change was indeed the result of changing ionic concentration resulting from the application of the electric field, see Figure 2.7. [69,71].

Another electromechanical mechanism of deformation of ionic polymer gels [2] has been reported as transportation of water from hydration [72] due to the cation flow or ion displacement [73]. Yet another possible mechanism is the formation of anisotropic complex between polymer gels and surfactant solution [74,75]. Doi et al. investigated, semi-quantitatively, the electric field induced strain of ionic polymer gels. They found that the
swelling speed of ionic polymer gels is proportion to square of the electric current. In addition, theoretical investigation of the electroresponse of IPGs have been reported by many [69,76,77].

2.1.2.2 Ionomeric Polymer-Metal Composites

Typical ionic polymer-metal composites (IPMC) have a laminated structure in which two metal electrode layers form the outermost layers and in the middle there is a thin layer of ionomeric polymer membrane. The metallic electrodes typically interpenetrate the polymer membrane to produce high surface conductivity. When a voltage is applied between the two electrodes solvated mobile cations diffuse toward the oppositely charged electrodes resulting in swelling near the negative electrode and shrinkage near the positive electrode, resulting in bending deformation, see Figure 2.8.

![Figure 2.8 Schematic illustration of the bending mechanism of a typical IPMC](image)

In general, noble metals such as gold, platinum and silver are preferred as electrode material due to better conductivity, high malleability and oxidation resistivity. For the ionomeric
polymers, they should be able to selectively exchange ions of a single charge with their own incipient ions. One of two main categories of polymers that has been widely used is the perfluorinated alkenes with short side-chains terminated with ionic groups (e.g. sulfonate or carboxylate) for cation exchange or ammonium cations for ion exchange [79,80]. DuPont commercialized this type of ionic polymers in the early 1997 as Nafion™ that is typically fabricated from polytetrafluoroethylene (PTFE) [81]. Other commercially available products are Flemion (Asahi Glass) and Aciplex (Asahi Chemical][78]. The other type of ionomeric polymers is the styrene/divinylbenzene-based polymer. In this type, ionic groups substitute the phenyl rings groups. These polymers with highly crosslinked structure are relatively rigid [79,80].

When a voltage is applied to Nafion, an initial fast deformation occurs due to cationic diffusion inside the ionomeric channels (the hydrophilic ionic side groups form “spherical cluster network” and connected by narrow channels). Cations, such as Li⁺,Na⁺,K⁺,H⁺, driven by the attraction from negatively charged electrode move and drag water molecules through the ionomeric channels. The side with the higher water content expands and thus the whole system bends towards the anode. The other phenomenon is the coulombic interaction between electrodes and the fixed anionic groups in the IPMC [78]. The initial reaction is followed by a slow actuation in the same direction. However, the bending deformation is followed by “relaxation” in the opposite direction as the cations are redistributed and the dipoles reorient. IPMCs tend to drift from their position due to relaxation.

The desirable features of IPMCs are light weight (1-2.5 g cm⁻³) and fast response (μs to s), low stimulation voltage (4-7 V) and the large bending strains (> 10%) [82–84].
There are many research works have been done to further improve the performance of IPMC. For example, instead of using metal electrode, carbonaceous electrodes (carbon black, CNT) also were explored to avoid problems of delamination and cracking associated with metal electrodes [85,86]. Nah et al. reported IPMC with nanofibrous Nafion mat which significantly improves ionic conductivity and strain speed [87]. To explore more available polymer types, Lee et al. used poly (styrene sulfonate)-grafted fluoropolymers as the polymer matrix and imidazolium-based ionic liquid as the inner solvent to show substantially larger bending displacement compared to Nafion-based actuators [88]. Vargantwar et al. reported a block ionomer with sulfonated midblocks and glassy endblocks to achieve a more stable molecular network with self-organizing feature and can be readily dissolved [89]. Jung et al. reported improved life-time of IPMC actuators by “doping” T\textsubscript{1}O\textsubscript{2} particles in the ionomeric polymers. Also, the mechanical properties, proton conductivity and water-uptake ratio of IPMC were dramatically improved by employing fullerene to reinforce the ionomeric polymer [90]. 3D IPMC actuators have also been developed based on the novel configurations [81,91]. Also, many works have focused on the theoretical modeling of electromechanical behavior of IPMC [92–97].

2.1.2.3 Conducting Polymers

Conducting polymers (CP) such as polypyrroles, and polyanilines, change their electrical property from semi-conducting to conducting upon doping with donor or acceptor ions. The mechanical actuators made from CPs consists of three essential components; an anode, a cathode and a separating electrolyte. CPs can be used as either of both electrodes (anode or
cathode). The dimensional change in CPs is associated with the volume changes to accommodate anions and cations diffusing into and out of the polymer from the surrounding electrolyte [98,99,65].

CPs can belong to a wide range of categories, including basic conductive polymers, substituted polymers, self-doped polymers, copolymers polymer/macron blends and composites. Every type of CPs has its own balancing counterion and solvent system [100]. Among them, several types have been widely investigated, such as polypyrrole, polyethylenedioxythiophene, poly (p-phenylene vinylene)s, polyanilines and polythiophenes [2]. In polypyrrole (PPy) during electromechanical redox reaction under an applied electric field, the electrons egress and the reorganization of double bonds takes place, see Figure 2.9. The remaining positive charges (polarons or bipolarons) are still stored in polymer chains correspondingly, the polymers have conformational movements and more free volume is generated to keep the electroneutrality. This free volume is filled with counterions as well as water molecules from the surrounding solution and thus the polymer swells. By contrast, in reduction process, electron ingress occurs and the positive charges are compensated. The double bonds recover and then water molecules as well as counterions are expelled out from the chains. The polymer accordingly shrinks [98]. These redox reactions take place in the electrolyte solution.
 Normally, a relatively low (1-10 V) electric potential is required to trigger CPs \[101\] strain is in the range of 2-10\% \[102\]. However, recent work by Kaneto et al. show strain level can be as high as 40\% \[65\]. And the work density (100 MJ/m\(^3\)) of CPs is relatively high \[65\]. In general, CPs have some drawbacks, such as low energy efficiency (on the order of 1\%) and low electromechanical coupling (less than 1\%) \[14\], and low rates of actuation due to slow diffusion of ions \[101\]. The main relative advantage of CPs over other EAPs is their low operating voltage, however, degradation of the polymer as well as the electrolyte system has been noted \[103\] at the upper limits of the voltage (\(\sim\)10V) \[99\]. With many attractive features, CP has been developed as a wide range of polymeric actuators, such as bending/linear artificial muscles, switchable membrane, drug delivery devices, biological transducers and artificial synapses etc. \[100\]

### 2.1.2.4 Carbon Nanotubes

Carbon nanotubes (CNT) can be easily thought as a layer of graphite (or graphene) rolled into a cylinder with nanoscale diameter. According to the number of layers of “graphene sheets”,

![Diagram of molecular conformational changes of conductive polymers while redox reactions](image)
there are two main categories of CNTs: single-wall CNTs (SWCNT) with only one layer, and multiwall CNTs (MWCNT) [65].

Like the conductive polymer actuators, CNT actuators need to work within electrolyte solutions. The predominant cause of actuation in CNTs is electrostatic. Unlike in dielectric elastomers (see Section 2.2) the electrostatic forces here are repulsive. Application of an electric potential, between an actuating CNT electrode and counter electrode in an ion containing solution, see Figure 2.10, leads to electronic charging of the CNTs. The interface between the CNT and electrolyte has been known as the “double layer”. The accumulated like charges on the CNTs induce repulsive forces against the stiff covalent (carbon-carbon) bonds in the CNTs causing the CNTS to deform [104,105]. Interestingly, the “double layer” works as a capacitor, in which the amount of stored charges depends not only on the capacitance of CNT/electrolyte interface but also on the magnitude of the applied potential [106].

![Figure 2.10 Schematic illustration of CNT actuators. (a) an applied potential injects charge of opposite sign in the two pictured nanotube electrodes which are in liquid or solid electrolyte; (b) charge injection at the surface of a nanotube bundle ; (c) a cantilever actuator in which two layer of CNT sheets are separated by a piece of scotch tape operated in the aqueous NaCl electrolyte [104].](image_url)
In 1999, a CNT actuator was fabricated in a “paper” form, known as “bucky-paper” [104]. A simple bimorph cantilever actuator was formed by pasting two CNT papers to a layer of double-sided scotch tape and NaCl as the electrolyte solution as shown in Figure 2.10 (c). The simple cantilever system was able to bend alternatively to produce 0.1%-1% strain under an electric potential of -0.5 V to 1.5 V [106].

Since then many interesting actuator designs have been proposed. For example, a "nanotweezer” was developed using two MWCNTs [107,108]. A CNT yarn actuator has been used to demonstrate reversible torsional movement [109]. Recently, an all-solid-state CNT two-ply yarn torsional actuator made by plying two twisted CNT yarns infiltrated with the solid gel electrolyte [110] has been reported.

Spinks et al used a new CNT actuator design termed as “pneumatic mechanism” to demonstrate very large strain of 300% in the thickness direction along with 3% in-plane strain [111]. This giant strain results from the effect termed “pneumatic mechanism”. There is some gas generated beside the connection place between the electrode and the CNT sheets. The gas inflates the sheet and form many dis-shaped pores.

Carbon nanoscrolls (CNS), formed by wrapping graphite sheet into a cylinder, also show similar electroresponse feature. Under electric stimulation, CNS exhibits the scroll unwinding response due to charge injection [112].

CNT assemblies, with very large surface area and the porous structure, help ions transfer faster with a response time less than 10 ms and strain rate up to 19%/s. Because of the lightweight nature and the extremely high elastic modulus, CNT actuators have very high power-to-mass
ratio (up to 270 W/kg). And their work density is as high as about 1MJ/m$^3$ which is similar to that of dielectric elastomers and ferroelectric polymers. CNTs are thermally stable and can survive temperature higher than 450 °C in air and 1000 °C in the inert environment. For the shortcomings, the individual CNT particles produce very limited strain due to the extremely high elastic modulus[65]. At present, CNTs normally are limited by the low electromechanical coupling and the high cost [14].

2.2 Dielectric Electroactive Polymers

Dielectric electroactive polymers (D-EAP) constitute a class of electronic electroactive polymers with great potential. They are lightweight, low cost, and are able to generate large strains at high frequency. Because of these and other desirable qualities and their potential applications in contemporary and future technologies, D-EAPs are the most investigated actuator material in recent years. In all matters, electrons and ions move in response to applied external electric field. In conductors, the electrons and ions are able to move microscopic distances. However, in dielectrics, the charged species just move within very limited distance under the electric potential. And the deformation and the polarization are coupled. D-EAPs possess the electroresponse feature of dielectrics and low elastic modulus of elastomeric polymers[113]. This section focuses on D-EAPs in terms of the working principles, characterization techniques, materials commonly used including electrodes, actuator design, and their important performance and potential applications.

D-EAP actuators are capacitors with deformable electrodes and soft dielectric elastomer as the dielectric medium. The operation principle of D-EAP, shown in Figure 2.11, is relatively
simple. In a typical actuator configuration, a layer of soft D-EAP is sandwiched between two layers of compliant electrodes. On application of an electric potential across the electrodes, the attractive force between the opposite charges and the repulsive force between the like charges result in deformation of the dielectric elastomer. This electrostatic force induced deformation is known as “Maxwell stress,” described in detail in section 2.1.1.1. In addition to Maxwell stress effect, electrostriction related to the polarization in the dielectric material also plays a role in the electroresponse of D-EAPs. Electrostriction effect is usually small and is considered insignificant in exploring the mechanism behind electromechanical response of D-EAPs.

![Figure 2.11 Principle of operation of D-EAPs](image)

2.2.1 Characteristic Parameters

The important measures of performance of D-EAP actuators are stress, strain, coupling efficiency and energy density. It is very important to note that despite promising performance in terms of these parameters, not all D-EAPs are appropriate for every application. Therefore,
it is important to clearly understand this performance metric and the trade-offs that are often necessary for many applications. In the following, these parameters are defined and discussed in terms of published data on various D-EAP materials.

### 2.2.1.1 Actuation Stress and Blocking Force

The compressive stress \( P \) generated in a D-EAPs is expressed in terms of relevant parameters [114,115].

\[
P = \epsilon \epsilon_0 E^2 = \epsilon \epsilon_0 (V/z)^2 \tag{Equation 2.6}
\]

where \( \epsilon \) and \( \epsilon_0 \) is relative permittivity of free space and the material, respectively, \( E \) is the applied electric field, \( V \) is the applied voltage, and \( z \) is the thickness of the D-EAPs.

Equation 2.6 assumes that the dielectric behavior of the elastomer remains unaltered under strain. Nevertheless, it is not always true. For example, the acrylic elastomers (3M, VHB\textsuperscript{TM}, 4910) show lower dielectric constant under a large strain (dielectric constant drops to 4.45 from 4.7 when the prestretch ration increases to 15 from 0) [116]. So it is necessary to take the electrostriction effect into consideration. Accordingly, the in-plane stress and the compressive stress can be expressed as Equation 2.7 and Equation 2.8 respectively. In these two equations, \( \alpha_1 \) and \( \alpha_2 \) denote the dielectric property change factors in shear and bulk deformations. They are related to the electrostriction effect. So components in these equations containing the two factors represent the contribution of the electrostriction and the rest is related to Maxwell effect [117].

\[
\sigma_{xx} = \sigma_{yy} = \frac{1}{2} \epsilon \epsilon_0 E^2 (1 + \frac{\alpha_1}{\epsilon}) \tag{Equation 2.7}
\]
\[
\sigma_{zz} = -\frac{1}{2} \varepsilon \varepsilon_0 E^2 (1 - \frac{\alpha_1 + \alpha_2}{\varepsilon})
\]  
Equation 2.8

2.2.1.2 Blocking Force

The force required to return a fully activated actuator to its original state is defined as blocking force. In a planar actuator, it is the in-plane force exerted toward the inactive area at the boundary. Alternatively, it is the force generated by a linear actuator held at a constant length [118]. Considering isochoric deformation of the material under the effect of Maxwell stress only, the blocking force can be calculated from the actuator’s initial and activated dimensions. If a planar actuator having dimensions, \((X_0 \times Y_0 \times Z_0)\) changes to \((X \times Y \times Z)\) upon activation, see Figure 2.12, and the ratio between \(X_0\) and \(X\), \(Y_0\) and \(Y\) are \(\alpha_x\) and \(\alpha_y\), respectively. Then the blocking force \((F_y)\) in \(y\) direction can be expressed as [116].

\[
F_y = \sigma_{Maxwell} \times A = \varepsilon \varepsilon_0 E^2 \times (XZ) = \varepsilon \varepsilon_0 E^2 \times X_0 \times \alpha_x \times \frac{Z_0}{\alpha_x \alpha_y}
\]  
Equation 2.9

\[
= \varepsilon \varepsilon_0 E^2 \frac{X_0 Z_0}{\alpha_y}
\]
2.2.1.3 Actuation Strain

The electrostatic stress through the thickness direction as well as the in-plane repulsive forces engenders in-plane actuation strain in the D-EAP. In general, the actuation strain has been expressed both in terms of linear or areal actuation strains. The linear strain is the ratio between the change in length and the original length along any direction of the active area. For example, for the geometry as shown Figure 2.12, the linear strain can be expressed by Equation 2.10. Alternatively, the thickness strain can be calculated from the areal strain.

\[ S_x = \frac{X - X_0}{X_0} \times 100\% \]  \hspace{1cm} \text{Equation 2.10}

Similarly, the areal actuation strain is the ration between the increased area and the original area of the active part as Equation 2.11.

\[ S_{area} = \frac{XY - X_0Y_0}{X_0Y_0} \times 100\% \]  \hspace{1cm} \text{Equation 2.11}

The in-plane strain is normally tested by a well-known circular actuator set-up as shown in Figure 2.13. The rigid frame is needed as the support of pre-stretched film. The circular active area just occupying a small proportion of the whole area is to eliminate the boundary arcing [119].
2.2.1.4 Elastic Strain Energy Density

Elastic strain energy density ($u_e$) is a critical performance parameter of actuator materials. It is defined as a work output capability or energy output in one actuation cycle per unit volume of the material excluding the overheads (power supply, electrode, packaging, etc.) [120]. This parameter is very useful to evaluate the performance of an actuator because it is size independent. In terms of material properties, and assuming small strain, it can be expressed as,

$$u_e = \frac{1}{2} PS_z = \frac{1}{2} Y S_z^2$$

Equation 2.12

where $P$ is the compressive stress, $S_z$ is the strain in thickness direction and $Y$ is the modulus of the materials.

For large strain situations, the active area increases and the formula of elastic energy density is expressed as [115].

$$u_e = Y[S_z - \ln(1 + S_z)]$$

Equation 2.13

The equation works well as the strain is less than 20% and the value is much higher than the actual if strain is beyond this critical point.
2.2.1.5 Electromechanical Coupling Efficiency

Electromechanical coupling efficiency \( k^2 \), following the nomenclature for piezoelectrics is another useful parameter and is calculated as a ratio of the generated work to input energy. Therefore coupling efficiency is an expression of the proportion of the electric energy converted into mechanical energy in one working cycle. Normally, \( k^2 \) is derived from the capacitance of the D-EAP actuators by using the electrostatic model assuming the dielectric constant remains the same upon actuation [115]. The calculation for coupling efficiency begins with the capacitance \( C \) of D-EAPs actuator;

\[
C = \frac{Q}{U} = \frac{\varepsilon \varepsilon_0 A}{z} = \frac{\varepsilon \varepsilon_0 V}{z^2}
\]

Equation 2.14

Where \( Q \) is the total charge stored, \( U \) is the electric potential between electrodes, \( V \) and \( A \) is the total volume and active area of the capacitor or actuator respectively.

The electric energy of the actuator \( W \) expressed in terms of capacitance is,

\[
W = UQ = \frac{Q^2}{C}
\]

Equation 2.15

Assuming state 1 denotes the stage at the beginning of the application of electric field, and 2 is the stage when D-EAPs show mechanical response. If a viscoelastic loss of the polymer is ignored, the difference of electric energy between these two states is equal to the converted electrical energy to mechanical energy.

\[
\Delta W = W_2 - W_1 = \frac{Q^2}{C_2} - \frac{Q^2}{C_1}
\]

Equation 2.16

The electromechanical coupling efficiency is expressed as the fractional change. Because strain in thickness direction is negative, the coupling efficiency is normally expressed as,
\[
\frac{\Delta W}{W_1} = \frac{Q_2^2}{C_2} - \frac{Q_1^2}{C_1} = \frac{z_2^2 - z_1^2}{z_1^2} = \frac{[z_1(1 + S_z)]^2}{z_1^2} - 1 = S_Z^2 + 2S_Z
\]

Equation 2.17

Needless to note, actuator materials with high coupling efficiency are desirable.

The electromechanical coupling efficiency is determined by the mechanical and electrical loss in the elastomers. The mechanical loss in D-EAPs is normally greater than the electrical loss [4,121]. The mechanical efficiency represents the ability of elastomers to remain their elasticity, a parameter that is inversely proportional to the loss tangent. In viscoelastic materials, loss tangent is a measure of mechanical damping in the material and is measured as a ratio of the loss modulus to the storage modulus obtained through dynamic mechanical analysis of materials.

2.2.1.6 Electric Breakdown Strength

The electric field can be applied to D-EAPs is not infinite. There is a maximum value known as the breakdown strength at which D-EAPs have a finite conductivity owing to charge generated in the materials. At the electric breakdown strength, the electric circuit short occurs and there is “burning” phenomenon on the film. This failure normally is unrepairable because that it propagates quickly and the opposite electrodes surrounded it would be conductive. So a higher electric breakdown strength is desired in principle [116].

2.2.1.7 Effects of Prestrain on Actuation Performance

Prestrain has a significant influence on performance of D-EAPs in terms of the actuation strain, the response speed, the breakdown strength and the boundary constraint [122]. All of these effects are complex and interactive. For example, the prestrain, on one hand, helps to improve
the response speed and break down strength. On the other hand, it reduces the deforming ability of the elastomers [122].

In order to systematically investigate the influence of the prestrain on overall performance of D-EAPs actuators, Choi and coworkers experimentally studied the relationships between properties of VHB elastomers (one of the most used acrylic D-EAPs, more detail can be found in section 2.2.2.2) and the prestrain [122].

First of all, they examined the effect of prestrain on stiffness of polymers. In this work, VHB was firstly stretch to a certain strain (from 100% to 500%) and kept for a while. Then in the second step, the same sample was continuously stretched by the increment of 100% strain. For a compare purpose, one uninterrupted tensile testing was conducted with maximum strain of 600%. Figure 2.14 shows the results. Interestingly, it is found that after the first step stretching the stress-strain curves of the second step are very similar to the one of sample of uninterrupted testing. This means that the prestrain almost does not have influence on the inherent elastic modulus of the materials. However, the first strain magnitude surely decides the second step young’s modulus. For example, the initial modulus in the second stretching step following a 100% strain in the first step (the black dash line) is higher than that of the sample following a 400% prestrain in first step (the yellow dashed line). This suggests that prestrain is not always stiffening elastomers. In this work, VHB elastomers with prestrains of 200%-500% have similar stiffness, which is smaller than those with prestrains less than 200% (plateau effect [123]). But if the prestrain is beyond 500%, the moduli of the elastomers increase again.
Figure 2.14 Stress-strain curves of VHB elastomers with different strain and relaxation [122]

In addition, the permittivity of acrylic elastomers also is influenced by prestrain. Figure 2.15 illustrates the experimental results: VHB with the higher prestrain has smaller dielectric constant on contrast in whole of the frequency range (1-10^5 Hz). This phenomenon has been usually neglected which is actually large enough and not negligible.

Figure 2.15 Dielectric constant as a function of frequency of acrylic elastomers with different prestrain [122]
While the prestrain increases, the thickness of elastomer decreases. The thinner elastomers commonly have a lower breakdown voltage [116,124]. However, the breakdown strength of the thinner elastomers is higher because the decrease of thickness surpasses the decrease of breakdown voltage, see Figure 2.16. In addition, researchers claimed that the electric breakdown strength in cross polymer chains direction is higher than the one along chains. Because that in the cross-chain direction there are more D-EAP chains working as obstacles of avalanching electrons. On contrast, along chain direction there are less polymer atoms perpendicularly facing to electrons. When applying in-plane prestrain, more polymer chains orient horizontally so that the electric breakdown strength of D-EAPs increases [116].

![Figure 2.16](image)

Figure 2.16 Effect of prestrain on electric breakdown strength and voltage [116].

There is experimental evidence to suggest that the actuation strain of D-EAPs decreases with higher prestrain in the same electric filed [4,125]. This is generally attributed to the increased stiffness of the elastomer [125]. However, since prestrain also improves the electric breakdown strength of D-EAPs, the maximum actuation strain is improved with larger prestrain.
Figure 2.17 shows two acrylic elastomers films having different initial thicknesses but stretched to the same thickness by applying different levels of prestrain [122]. The film with higher prestrain produced lower actuation strain at the same electric field but significantly higher dielectric strength and thereby larger actuation strain at failure.

Figure 2.17 Comparison of actuation strain of VHB films of different initial thickness prestrained to have the same initial thickness [122]

The relationship between prestrain and the electromechanical efficiency as well as the loss tangent of the materials has been examined [121]. The loss tangent and mechanical efficiency is shown as a function of prestrain and frequency in Figure 2.18. At all frequencies reported the efficiency is greater at higher prestrain values. The relation between prestrain and loss tangent have been reported for many materials [121], including gum vulcanized [126], natural rubber [127] and poly (ethylene terephthalate) monofilaments [128]. In general, prestrain is reported to increase molecular chains orientation resulting in reduced the degree of freedom of the molecular relaxation and thereby less viscoelastic losses.
Figure 2.18 Mechanical efficiency and loss tangent of acrylic elastomers at different levels of prestrain and frequency [121]

In order to apply the prestrain to elastomers, bulky and cumbersome rigid frames are commonly required. The added mass significantly decreases the elastic energy density of the actuator system. In addition, a prestrained elastomer goes through varying levels of stress relaxation that further causes the time-varying actuation response [123]. In order to address these problems, novel D-EAP materials that do not require prestrain have been reported. In what has been called an interpenetrating network (IPN) [129,130]. The prestrained dielectric elastomer is stabilized using second interpenetrating elastomeric networks, as shown in Figure 2.19. The second polymer is curable with a bifunctional or trifunctional group(s) in the chemical structure, such as poly(1,6-hexanediol diacrylate) (poly (HDDA)) [129,130], poly (trimethylolpropane trimethacrylate) (poly (TMPTMA)) [130,131], and silicone [132] etc. The resulting IPNs have been reported to produce improved actuation stress [132], areal actuation strains up to 300% [130], high elastic energy density and high electromechanical coupling.
efficiency [131]. The IPN network developed using prestrained acrylic elastomer film, VHB, has been stabilized by 18.3% second polymer network. The VHB network successfully retained 275% prestrain from the starting prestrain of 400% [129,130]. Suo and Zhu established a theoretical model to simulate interpenetrating networks with long and short chains to have a better understanding of the interlocking mechanism [133].

Figure 2.19 Schematic illustration of the fabrication process of an IPN elastomer film. (a) an acrylic film before processing; (b) after 400% biaxial prestrain; (c) curable additives are added into the prestrained film and cured, forming interpenetrating network of a highly crosslinked polymer; (d) after the external stress is removed, the interpenetrating network preserves most of the prestrain of the acrylic film [130].

In a similar effort, Shankar et al. developed polystyrene-block-poly(ethylene-co-butylene)-blockpolystyrene (SEBS) triblock copolymer swollen with midblock-selective solvent [134,135]. This nanostructured copolymer system has properties that surpass those of non-prestrained VHB elastomers in terms of actuation strain, energy density, electromechanical and coupling efficiency. More detail on this material is included in section 2.2.2.4.
2.2.1.8 Instability and Failure

The instability of D-EAPs mainly roots in the “pull-in effect” [136]. The instability or “pull-in effect” occurs in a D-EAPs when its thickness falls below a certain value as the electrostatic stress becomes larger than the materials’ compressive strength. Beyond this critical point, the pressure further flattens the film leading to a larger electrical field across the film and much higher pressure resulting in “positive feedback”. The process finally leads to instability and catastrophic failure of the film. Figure 2.20 shows the visible wrinkled morphology on the surface of D-EAPs before failure. Due to the extremely large local deformations, wrinkles eventually bring in either mechanical failure or dielectric breakdown of the materials.

![Figure 2.20 Film surface morphology during pull-in instability [136]](image)

Zhao and coworkers explained the instability in dielectric elastomers using the variation of electric potential needed to maintain charge in the DE electrodes. The applied electric voltage plotted as a function of amount of stored charges in the electrodes during actuation process is shown in Figure 2.21. When the amount of charge (Q) is small, the voltage (Φ) increases proportionally. As Q becomes large, the thickness of the elastomer film decreases significantly.
to make the true electric extremely large. At this point, the potential ($\Phi$) needs to maintain the charge drops. As $Q$ increases further, the flattened elastomer film becomes mechanically stiff in compression and thus $\Phi$ again increases with $Q$. The first peak of $\Phi$ has been known as start of pull-in instability. The process of thinning of the elastomer is discontinuous because of the non-uniform distribution of charge and the applied voltage. If $\Phi$ is held constant, the elastomer will show electrochemical hysteresis and a sudden reduction of the thickness like ferroelectrics. If $Q$ is maintained, there will be a state in which the thin and thick regions of the elastomers exist at the same time under the certain voltage. And the thin region with larger area will be constrained by the surrounding thick regions so that wrinkles appear [137]. According to the theoretical model, the thin parts tend to expand to the thick regions at a constant voltage [138].

![Schematic illustration of the voltage-charge curve of a layer of an elastomer dielectric where $\Phi$ is electric voltage and $Q$ is stored charged in the electrodes][137]

Zhao and Suo also investigated the pull-in instability of D-EAPs using free energy theory [139,140]. They concluded that the critical electrical field associated with instability becomes higher for prestrained elastomer membranes. This has also been observed experimentally [136]. Norris further simplified Zhao’s model with a two term Ogden rubber elasticity model...
Díaz-Calleja et al. further explored the pull-in effect and the electromechanical stability of neo-Hookean silicones [142]. Meanwhile, Liu et al. also modified Zhao’s model and Díaz-Calleja’s model through using Mooney–Rivlin elastic strain energy function with two materials constants [143–145]. Later, Leng et al. proposed a nonlinear expression of the permittivity as the function of strains and built the relationship between critical nominal electric field, critical true electrical field, nominal stress and principle stretch ratio [140]. Furthermore, Xu at al. used the concept of total stress to broaden the availability of expression for the critical stability electric field of Zhan and Suo which can be used for general hyperplastic material models [146]. Also, for specific shaped D-EAP devices, their instability has been investigated as well, for example, the multilayered soft dielectrics [147] and tube actuators [113].

There are two main mechanisms behind D-EAPs failure: the mechanical breakdown and the electric breakdown [148]. Polymers generally are viscoelastic. Their deformation includes the recoverable elastic component and plastic unrecoverable component. During the actuation cycling of D-EAPs, unrecoverable component of the deformation (secondary creep) is added up leading to the mechanical breakdown. In case of electric breakdown, the finite electrical resistance of the D-EAP membrane is overcame by the charge generated in the electrodes. At the dielectric strength of the D-EAP, the electric circuit is shorted. It is important to note, that the dielectric breakdown strength is not a materials characteristic. The mechanisms behind electric breakdown are not always identical [149]. For example, electrons in the polymers are accelerated and collide that may trigger the avalanching breakdown. In addition, thermal breakdown also takes place due to the joule heating and dielectric loses. In some elastomers,
conductivity increases with the temperature (e.g. polyurethane). The increase in temperature may be dramatic because of the positive feedback mentioned before. Discharge of gas at the surface and/or inside of the elastomers could also induce the electric breakdown. The defects in the form of air voids in elastomers are normally the places where sparks begin. Thickness variation in elastomers may also produce extremely high local electric field. Poor circuit connections may lead to failure due to local joule heating [148].

Zhao and Suo categorized three types failure in dielectrics based on their voltage-stretch response, see Figure 2.22 [150,151]. Type I response occurs in stiff dielectrics, such as ceramics or glassy polymers. These materials fail because of electrical breakdown; Type II is the soft dielectrics, such as elastomers, normally fail due to the electromechanical instability; Type III is a special class of dielectrics. They are able to survive the electromechanical instability and further deform to reach a giant strain level.

Figure 2.22 Working principle of dielectric elastomers transducers and three types of failure mechanisms. (a) a membrane of dielectric elastomer subject to a voltage reduces thickness and expands area. (\(\Phi\) denotes applied voltage and \(\lambda\) is the deformation ratio). The voltage-stretch curve typically is not monotonic. (b)-(d) three types of dielectrics are differentiated by where the two curves \(\Phi(\lambda)\) and \(\Phi_B(\lambda)\) intersect, where \(\Phi_B(\lambda)\) is the breakdown voltage as a function of stretch ratio that normally is a decreasing function [150].
Type III dielectrics should have the following features, (1) at small strain the materials should be soft; (2) at modest strain $\lambda_{lim}$ the dielectrics should stiffen steeply, here the modest strain is emphasized which means the stretch should not be excessive as shown in Figure 2.23 (a). In order to achieve these features, polymeric composites (see Figure 2.23 (b)) or polymers with special molecular structures were proposed (see Figure 2.23 (c-e)).

![Figure 2.23 Ideal stress-strain curve and molecular structure of dielectrics with Type III failure type. (a) stress-strain curve of a membrane under biaxial stress; (b) fiber embedded in a compliant matrix; (c) a network of polymers with folded domains; (d) a network of polymers with side chains; (e) a network of polymers swollen with a solvent [150].](image)

Besides to tailor polymer molecular structures, there are other options to eliminate the electromechanical instability. One of them is the electrode-free actuator design. Driven by sprayed-on electrical charges, such as corona discharge from two needle electrodes needle
combs [152], the applied charges are controlled. Accordingly, the pull-in effect can be effectively prevented without the positive feedback [152,153]. The other option is to dynamically control the load applied to the elastomers and the electrical voltage cross them. For example, Keplinger et al. firstly inflated a D-EAP film with air to the state close to appearance of the instability. And electrically actuate it without the air inflation. This approach successfully achieved 1692% areal actuation strain [154,155]. Recently, Zhao et al. reported a new type of instability which is called “creasing to cratering instability”. As the name described, the flat polymer film tends to form a local folded structure. These folded regions form creases as the applied electric field is high enough. While continually improving electric field, the creases finally evolve into craters in the polymers. To understand this new type of instability is helpful for designing insulating cables, organic capacitors, polymer actuators, energy harvesters and functional surfaces/patterns [156,157].

### 2.2.2 Materials and Properties

During late of 1990s and early 2000s, a wide range of D-EAP materials have been investigated, including but not limited to silicones [125,125,158], acrylic elastomers [116,159,160], polyurethanes [161–163], isoprenes [164], interpenetrating networks [129,131,133], block-copolymers [89,134,165] and fluoroelastomers [115]. Among these, silicones, acrylics and polyurethane have been explored most often. Additionally, block copolymers look promising because of their spontaneous molecular self-organization and composition-tunable dielectric and mechanical properties. So in this section, silicone, acrylics, polyurethane and block copolymers are focused.
2.2.2.1 Silicone

Silicones, having flexible silicon-oxygen backbones, are one of the most explored D-EAPs (see Figure 2.24). The side chains attached to silicone atoms are various and thus there are many different types of silicones, such as dimethyl-silicones, vinyl-silicones, phenyl-silicones, nitrile-silicones, fluoro-silicones, boro-silicones and silicone-carbides [166]. Among them, polydimethylsiloxane (PDMS) with two methyl (CH$_3$) side groups is the most common. There are many commercially available silicone products, including CF 19-2186 from NuSil, and HS3, and Sylgard from Dow Corning [167]. Most of their properties have been summarized in reference [7].

Silicones are stable over a broad range of working temperature (−50°C-200°C) and humidity. As D-EAP they have fast response speed as well as high electromechanical coupling efficiency [125]. The reliability of silicone is promising, for instance, it was reported that silicone based framed actuators worked for $10^7$ cycles within 5% strain level at a frequency of about 10 Hz, without any degradation of performance [8]. However, silicone elastomers as D-EAP show moderate actuation strain (5.6-117% linear strain [168]) with or without the prestrain. Also, due to relatively low dielectric constant (~2.8 [168]), the trigger electric field for silicones is relatively high (72-350 V/μm).
The properties of silicones are tunable through their compositions and processing conditions. The elastic modulus of silicones, for instance, can be tailored by adjusting the ratio of crosslink agents [169–174]. This thanks to the change of the polymer networks crosslinking density. In addition, crosslinking agent also influences other properties of silicone, such as the dielectric constant and the breakdown strength [125]. Efforts to improve the relatively low dielectric constant of silicone elastomers have been widely done. The most used methods are using electrically conductive and high-dielectric fillers or blending silicone with other polymers. High dielectric constant fillers have been extensively used to improve the performance of silicone elastomers. There are main two categories of fillers, one is inorganic fillers (TiO$_2$ [172,175], BaTiO$_3$ ([176,177], Pb(Mg$_{1/3}$Nb$_{2/3}$)O$_3$-PbTiO) [177], water [178]) and the other is organic fillers (poly(CuPc) [179], copper-phthalocyanine oligomer (CPO) [125], and encapsulated polyaniline (PANI) [180,181]). In addition to high dielectric constant fillers, conductive fillers also have been widely explored, such as carbon nanotubes [182,183], expanded graphite [184] and carbon black. Addition of high dielectric constant fillers to improve the dielectric behavior of the polymer is commonly often associated with increasing stiffens that in principle may induce the less actuation strain. The second problem is a dramatic lowering of the dielectric strength (for example, the breakdown strength of silicone increases by 60% after adding 9%wt of TiO$_2$ [185]) and increase in dielectric loss (for example, the dielectric constant of silicone doubled after adding 12%wt of TiO$_2$[185]). However, there are exceptions. For example, Carpi et al.
reported after adding TiO$_2$, the young’s modulus of silicones reduced to 16 kPa from 40 kPa [175].

In addition to the composites methods, the polymer blends approach is the other option to modify silicone elastomers. The blending approach is considered better because of the increasing likelihood of more uniform dispersion between components in solution state. Another likely outcome is the preservation (or reduction in cases) of softness of the silicone elastomer due to addition of a more compatible polymer as blend. For example, Carpi et al. fabricated the PDMS-based blends with a highly polarizable conjugated polymer poly(3-hexylthiophene) (PHT) [186]. The resultant blends had not only an improved permittivity (from 4.6 to 13.8) but also a reduced elastic modulus (from 100 kPa to 46 kPa). These improvements all together brought a considerable increase of the electromechanical response (transverse strain at 8 V $\mu$m$^{-1}$ increased by 2.3 times). In addition to PHT, other polymers have been explored based on PDMS, such as inherent conductive polymer polypyrrole [187], polyethylene glycol (PEG) [188] and polyurethane [177] etc. Furthermore, copolymer approach is another option to modify the polymer performance which benefits from developed polymer synthetic technologies [189].

2.2.2.2 Acrylics

Acrylic elastomers, known also as acrylate rubber or polyacrylate rubber, have the chemical structure shown in Figure 2.25. The most well-known commercially available products are the “very high bond” (or VHB) structural adhesive elastomers manufactured by 3$M^TM$. These
crosslinked amorphous polyacrylate networks sold in the form of films and can be used as D-EAP without any further processing[116].

\[
\begin{align*}
\left(\text{CH}_2\text{CH}_2\right)_n \\
\text{C} \equiv \text{O} \\
\text{OR}
\end{align*}
\]

Figure 2.25 The chemical structure of acrylic elastomers.

VHB produces very high actuation strain (maximum area strain up to 380%), and high elastic energy density (up to 3.4 MJ/m$^3$). They can sustain very high strains up to 36 times in area without breaking. This acrylic elastomer has been extensively explored in many D-EAP applications. However, these are not as good as silicones in terms of environmental tolerance [190]. Figure 2.26 shows that the young’s moduli of acrylics and silicone as a function of temperature. Note the dramatic change in modulus at temperatures below 0°C, as a comparison silicones perform more stably in this temperature range.

Figure 2.26 Young’s modulus as a function of temperature for the VHB 4910 and silicone hardened with 5% hardener [125]
Additionally, acrylic elastomers have a lower respond speed and a larger viscoelastic loss (hysteresis) compared to silicones [125], see Figure 2.27. Note that while silicone elastomer only took approximately 3 seconds to reach the maximum strain, the acrylic elastomer kept growing for 30 seconds and still did not reach a stable state (the experiment results not included here showed after 3 mins the strain still kept increasing). In general, acrylic elastomers show significantly higher creep deformation compared to silicones [191].

![Cyclic actuation of (a) silicone (b) acrylic elastomer VHB F-9437PC actuators](image)

Figure 2.27 Cyclic actuation of (a) silicone (b) acrylic elastomer VHB F-9437PC actuators [125].

In particular, at high frequency, acrylics have lower dielectric permittivity, higher young’s modulus and lower electromechanical response [192]. This agrees well with the suggestion that VHB is more suitable for low frequency applications (10-100Hz) [7,159]. Pei et al. proposed a method of using low molecular weight additives to reduce the viscoelastic losses of VHB. The glass transition temperature of VHB dropped to -62°C from -33°C by introducing additives [6]. In section 2.2.1.7, the interpenetrating polymer networks (IPNs) based on acrylic elastomers are discussed.
2.2.2.3 Polyurethane

Polyurethane (PU) elastomers are block copolymers that include alternating rigid and soft segments as shown in Figure 2.28. The hard segments dominate the manifested properties of the copolymer due to their rigidity and hydrogen bonds, and work as the physical crosslinks [161].

Figure 2.28 Schematic illustration of the morphology of a polyurethane elastomer where the hard segments (hatched boxes) are embedded in the matrix of soft segments (thin lines) [162]

PU has relatively high dielectric constant (~6-7) and larger actuation force output (0.14-1.6 MPa) [168]. During late 1990s to early 2000s, investigate great deal of work aimed at elucidating electromechanical response of polyurethanes [162,163,193–195]. PU suffers from limited actuation strain so it soon fell into sidelines while interest in silicone and acrylic elastomers grew[168]. More recent works related to PU are more focused on their composites. Fillers employed in PU based composites are normally conductive species in order to improve their permittivity and actuation performance. Similar to silicone elastomers, carbonaceous fillers such as graphite [196], carbon nanotubes [197] and carbon black [198–200] etc., have also been investigated. As mentioned earlier, the composite approach is associated with increased dielectric loss and elastic modulus which may result in higher leakage current, power consumption and a reduction of actuation strain [168]. Beside carbon-based fillers, the
nanosized montmorillonite (MMT) [201] and carbon-coated SiC nanowires [202] also have been explored as fillers.

In addition to adding fillers, to tailor the molecular structure or composition of PU is the other option to modify its performance. For example, Huang et al. proposed chemically compatibilized fillers which in principle can improve performance of the silicone composites. They fabricated three component functionalized polymers with polyaniline (PANI), copper phthalocyanine oligomer (PoluCuPc) and PU. Encouragingly, the resultant multicomponent polymers have improved dielectric constant, breakdown field and reliability as well as lower dielectric loss [203]. With the similar idea, Wang et al. synthesized PU-PolyCuPc copolymers. They found the nanosized PolyCuPc dramatically increased the dielectric constant of the composites by about 60 times[204].

In a recent work, the polarizability and dielectric constant of PU were reportedly improved through the disruption of formation of hydrogen bonds. A proton donor, diaminonaphthalene (DAN) was used to disrupt the hydrogen bonds between thermoplastic PU chains. With reduction of modulus and increase of dielectric constant, this treated PU produced higher actuation strain (areal strain increase from ~0.5% to ~2.5%) at a relatively low electric field of 20 V/μm [205]

### 2.2.2.4 Block Copolymers

Block copolymers consist of two or more long sequences of chemically different repeat units that are covalently linked together to form a single macromolecule [206]. One of the most unique features of a block copolymer is the ability of self-organization. Due to thermodynamic
incompatibility, blocks in a block copolymers undergo spontaneous molecular self-assembly into various nanoscale morphologies. Their performance is tunable by tailoring the block materials and their concentrations. For example, by optimizing the proportion between the rigid and soft blocks, the mechanical properties of copolymer can be altered.

Because of their potential, a diverse group of block copolymers have also been explored as electroactive polymers. The block copolymers investigated as electroactive polymers include, diblock copolymer polyurethane[162,193], various triblock copolymers, such as poly(styrene-b-(ethylene-co-butylene)-b-styrene) (SEBS) [134,135,206], poly (methyl methacrylate)-b-poly(n-butyl acrylate)-b-poly(methyl methacrylate) (PMMA-PnBA-PMMA) [207–209], poly (styrene-isoprene-styrene) (SIS) [210], styrene-butadiene rubber (SBR) [210], styrene-acrylic copolymers (SAR) [210], poly (methyl methacrylate (MMA)-dodecyl methacrylate (DMA)-MMA) (PMDMs) [211], other triblock copolymers with cyanobiphenyl moieties and poly(n-butyl acrylate) segments [212] or polydimethylsiloxane (PDMS) segment [213] etc.

SEBS is a triblock copolymer consisting of glassy end block of styrene segments (S segment) and the rubbery midblock segments (EB segment) as illustrated in Figure 2.29 [206]. This physically cross-linked micellar networks can be swollen with low-volatile, aliphatic-rich solvent to controllably decrease the stiffness of the copolymer (7-163 KPa). Because of this low stiffness and the polarizability of the styrene blocks, SEBS show excellent displacement under an applied electric field. SEBS showed a considerable areal strain (>200%) at a relatively low electric field (<40 V/µm) with the high electromechanical coupling efficient (92%) [206].
Among the many material parameters that can be adjusted to tailor the electromechanical performance of SEBS, are the molecular weight of the copolymers, the concentration of the solvent, and the fractional composition. All of these parameters significantly influence the actuation strain, dielectric constant, electric breakdown strength and the electromechanical coupling coefficient [134]. This flexibility offers great potential to satisfy the diverse requirements of advanced contemporary engineering, biomimetic, biomedical [135] and optical applications [214].

Compared to the benchmark VHB elastomers, SEBS demonstrates the less dependency on the prestrain to achieve acceptable electroresponse [134]. Published data suggests SEBS having the optimum composition and concentration has higher strain and coupling efficiency as well as considerably improved energy density than VHB without prestrain [134]. To improve the relatively low dielectric constant (≈2) of SEBS, chemical grafting of electrically conductive macromolecules [215] and addition of porphyrin metal complex [165] have been considered. Stoyanov et al. grafted the π-conjuncted polyaniline (PANI) to the flexible backbone of (poly-styrene-co-ethylene-co-butylene-co-styrene-g-maleic anhydride) (SEBS-g-MA) and the permittivity was increased by 7 times [215,216]. Another shortcoming of SEBS is the low
actuation force which in principle can be improved by increasing the Young’s modulus of
SEBS [135], if lower strain is not a concern. In addition, the solvent used to swell the
copolymer network may accelerate degradation of the polymers over time due to leaching [7].

In summary, Table 2.2 lists the representative performance parameters of commonly used D-
EAPs. In general, the requirement of high trigger electric potential limits the practical
applications of D-EAPs [217]. Especially in bulk devices, it is necessary to have safety
precautions, such as to use foolproof packages similar as those used in fluorescent lights [8],
or to employ a switch-model amplifier (it can build very small electrical circuits that may
supply high voltage) to reduce the risk [116]. From the materials perspective, there are two
general methods to lower the required electric voltage: (a) to reduce the thickness and thus the
corresponding electric field will increase under the same voltage; (b) to improve the dielectric
constant so that the low electric field is required to achieve same response. Thickness reduction
requires more careful processing in order to avoid dielectric breakdown due to local
inhomogeneity or variability in structure. As a result, improving the permittivity remains an
area of active research in D-EAPs [7].
<table>
<thead>
<tr>
<th>Polymer</th>
<th>Elastic energy density (J/cm³)</th>
<th>Actuation pressure (MPa)</th>
<th>Areal strain (%)</th>
<th>Young’s modulus (MPa)</th>
<th>Electric field (V/µm)</th>
<th>Dielectric constant (@ 1kHz)</th>
<th>Coupling Efficiency, ( k^2 ) (%)</th>
</tr>
</thead>
<tbody>
<tr>
<td>Silicone (Nusil CF19-2186)</td>
<td>0.091-0.75</td>
<td>0.6-3</td>
<td>33-64</td>
<td>1</td>
<td>160-350</td>
<td>2.8</td>
<td>0.05</td>
</tr>
<tr>
<td>Silicone (Dow Corning HS3)</td>
<td>0.026-0.16</td>
<td>0.13-0.4</td>
<td>69-93</td>
<td>0.1-0.135</td>
<td>72-128</td>
<td>2.8</td>
<td>0.05</td>
</tr>
<tr>
<td>Silicone (Dow Corning Sylgard 186)</td>
<td>0.082</td>
<td>0.51</td>
<td>47^b</td>
<td>0.7</td>
<td>144</td>
<td>2.8</td>
<td>54^a</td>
</tr>
<tr>
<td>Polyurethane (Deerfield PT6100S)</td>
<td>0.087</td>
<td>1.6</td>
<td>12^b</td>
<td>17</td>
<td>160</td>
<td>7</td>
<td>0.08</td>
</tr>
<tr>
<td>Fluorosilicone (Dow Corning 730)</td>
<td>0.0055</td>
<td>0.39</td>
<td>39^b</td>
<td>0.5</td>
<td>80</td>
<td>6.9</td>
<td>-</td>
</tr>
</tbody>
</table>
Table 2.2 Continued

<table>
<thead>
<tr>
<th>Material Type</th>
<th>Thickness Strain</th>
<th>Thickness Modulus</th>
<th>Dielectric Strength</th>
<th>Dielectric Loss</th>
<th>Cost</th>
<th>Lifespan</th>
</tr>
</thead>
<tbody>
<tr>
<td>Fluoroelastomer (Lauren L143HC)</td>
<td>0.0046</td>
<td>0.11</td>
<td>9&lt;sup&gt;b&lt;/sup&gt;</td>
<td>2.5</td>
<td>32</td>
<td>12.7</td>
</tr>
<tr>
<td>Isoprene natural rubber latex</td>
<td>0.0059</td>
<td>0.11</td>
<td>12&lt;sup&gt;b&lt;/sup&gt;</td>
<td>0.85</td>
<td>67</td>
<td>2.7</td>
</tr>
<tr>
<td>Acrylic (3M VHB 4910)</td>
<td>0.0057-3.4</td>
<td>0.13-7.2</td>
<td>40-215</td>
<td>2.3-3.0</td>
<td>17-412</td>
<td>4.2-4.8</td>
</tr>
<tr>
<td>SEBS161 (5-30 WT% copolymer)</td>
<td>0.141-0.151</td>
<td>-</td>
<td>30-180</td>
<td>0.007-0.163</td>
<td>32-133</td>
<td>1.8-2.2</td>
</tr>
<tr>
<td>SEBS217 (5-30% copolymer)</td>
<td>0.119-0.139</td>
<td>-</td>
<td>47-245</td>
<td>0.002-0.133</td>
<td>22-98</td>
<td>1.8-2.2</td>
</tr>
<tr>
<td>ACN rubber</td>
<td>0.084</td>
<td>0.3</td>
<td>25&lt;sup&gt;2&lt;/sup&gt;</td>
<td>4</td>
<td>50</td>
<td>14</td>
</tr>
<tr>
<td>IPN (VHB 4910-HDDA)</td>
<td>-</td>
<td>-</td>
<td>233</td>
<td>2.5</td>
<td>300</td>
<td>-</td>
</tr>
<tr>
<td>IPN (VHB 4905-TMPTMA)</td>
<td>0.68</td>
<td>1.51</td>
<td>146</td>
<td>3.94</td>
<td>265.4</td>
<td>2.43</td>
</tr>
<tr>
<td>IPN (VHB 4910-TMPTMA)</td>
<td>3.5</td>
<td>5.06</td>
<td>300</td>
<td>4.15</td>
<td>418.05</td>
<td>3.27</td>
</tr>
</tbody>
</table>

a. refers to [115]; b. Calculated from thickness strain

In summary, the materials research in the area of D-EAPs is ongoing. Discovery or adaptation of new polymer types including the composites and material processing technologies are key areas of research. In composites approach, a wide range of organic and inorganic high dielectric...
constant fillers are being explored, nanoscale materials and processing techniques are also being employed. For example, the reported performance of poly(propylene oxide) (PPO) compares well with that of VHB elastomer [218].

### 2.2.3 Compliant Electrodes

Electrodes used in elastomers play an important role in actuation performances of D-EAPs. In general, there are some essential features that electrodes used in D-EAP actuators must have. These include high compliance (the young’s modulus less than 100MPa or than that of the D-EAP), low electrical resistance (sheet resistance $< 1k\Omega/\square$) even under high strain, lower thickness than the elastomer (a few $\mu m$ or less), high charge density under large strain, good adhesion to the elastomers, long lifetime, and ability to be patterned with high resolution. Because electrostatic devices are normally required to work under high voltage and low current, electrodes materials do not need to be highly conductive [219]. A wide range of materials have been used as electrodes with D-EAPs [220, 221]. This section will briefly summarize the commonly used materials and related techniques.

The amount of charge stored in the electrodes at a given electric potential is key parameter in the performance of electrode materials. In principle, the charges stored in the actuator and its equivalent capacitance can be calculated based on some measurable parameters [222]. Assuming a film shaped D-EAP actuator is a capacitor, its equivalent circuit can be represented by resistors $R_1$ and $R_2$ in series and parallel, respective, with the capacitor as shown in Figure 2.30. $R_1$ denotes the electrode resistance and $R_2$ denotes the resistance of capacitor.
Figure 2.30 The equivalent circuit of film shaped dielectric elastomer actuator.

The capacitance of this actuator is expressed in terms of the relevant parameters as [222],

\[
C = \frac{Q}{V_C} = \frac{1}{V_C} \int_{t_1}^{t_2} I_C(t) dt
\]

Equation 2.18

Where, \(Q\) is stored charge, \(V_C\) and \(I_C(t)\) are the electrical potential and current through the capacitor, respectively. \(t_1\) and \(t_2\) represent the initial and final stimulation time, respectively. \(I_C(t)\) can be calculated as,

\[
I_C(t) = I(t) - \frac{V_{R_2}}{R_2}
\]

Equation 2.19

Where, \(I(t)\) is the overall current, and \(V_{R_2}\) is the electric potential over \(R_2\). \(V_{R_2}\) is expressed,

\[
V_{R_2} = V - I(t)R_1
\]

Equation 2.20

Equations 2.22-2.24 contain two measurable parameters, \(V\), and \(I(t)\). Parameters \(R_1\), \(R_2\) and \(V_C\) are unknowns. Equivalent resistance values, \(R_1\) and \((R_1 + R_2)\) are defined at time, \(t_1^+\) when capacitor just starts charging and at \(t_2\) when it is in fully charged or in steady state, respectively. Therefore values of \(R_1\) and \(R_2\) can be calculated from;

\[
R_1 = \frac{V}{I(t_1^+)}
\]

Equation 2.21
The parameters in these two equations are all measurable except $V_C$. It is known that the electrical potentials between the capacitor, $C$, and equivalent resistance, $R_2$, are same, thus the voltage between $C$ at steady state can be expressed as Equation 2.23.

$$V_C = V_{R_2}(t_2) = V - I(t_2)R_1$$  \hspace{1cm} \text{Equation 2.23}

Finally, all the parameters in Equation 2.18 can be substituted with the measurable parameters. As a result, the capacitance as well as the charge stored in the electrodes can be calculated. These are important for further estimation of the Maxwell stress and the electrostriction effect.

### 2.2.3.1 Carbonaceous electrodes

Carbon particle based electrodes are the most common in D-EAP actuators. The most often used form of the carbonaceous material is carbon grease [116] [219]. Other forms of carbon such as carbon black, graphite etc. have also been explored [220]. With the development of nanosized materials, carbon nanotubes have also been explored as electrodes for D-EAP devices [223–225]. Table 2.3 summarizes some of the properties of commonly used carbonaceous electrodes.

Carbonaceous electrodes have many unique advantages, including the ease of application, commercial availability at low cost, stability at high strain, and good electromechanical coupling efficiency etc. However, there still remain a few issues. For example, uniform dispersion of particles especially in the multilayer configurations is often difficult [219]. The “wet” state carbon grease usually smudges and tends to dry out so that it has relatively short

$$R_2 = \frac{V_C}{I(t_2)}$$  \hspace{1cm} \text{Equation 2.22}
working life. The graphite powders/spays require additional adhesive binder system to work. The binders may stiffen the elastomers [220]. The elastomers loaded heavily with carbon fillers also have stiffening influence [221].

### 2.2.3.2 Metallic electrodes

Various forms of metals are also used as electrodes in D-EAPs. Though they possess high conductivity, metallic materials generally have two problems in use as electrodes. The first obstacle, obviously, is the stiffness of metals that is much higher than those of elastomers (50-100 GPa compared to 0.2-1 MPa). The rigid metal electrodes may significantly constrain the deformation of elastomers upon actuation. For instance, an 8 nm thick gold layer was sputtered on a 30 μm thick PDMS film increased the Young’s modulus of the elastomers from 0.77 MPa to 4.2 MPa. The second problem of metal electrodes is the low elasticity (normally is 2-3%) and therefore tendency to crack under a high strain [221].

Metal electrodes have been applied to D-EAP actuators through various techniques including sputtering [226], and vapor deposition [227]. In microelectronics and microelectromechanical systems (MEMS) industry, there are many technologies to fabricate patterned conductive thin films, including electron beam evaporation, cathodic sputtering, and electroplating and photolithography. These patterning technologies also show potentials in D-EAPs actuators[221].

One interesting example of patterned metal electrodes is the zig-zag shaped gold electrodes processed by the sputter-deposition technique [219]. In this particular instance, gold was first sputtered uniformly on a silicone film. In a subsequent step, patterning was achieved through
photolithography with a resolution as high as 5 μm (see Figure 2.31 (a)). This zigzag patterned gold electrodes worked up to 80% strain. Bi-component electrodes made with metallic materials with varying conductivities as shown in Figure 2.31, reportedly improved the reliability and robustness of actuators. While the electric breakdown takes place, the current has to go through electrodes with low conductivity (the bottom electrode in Figure 2.31 (b)) to the one with high conductivity (the zig-zag electrode in Figure 2.31 (b)). The less conductive electrode may limit the current and localize the breakdown effect. It theoretically can avoid catastrophic failure. Less conductive electrodes also help to limit the current leakage.

![Figure 2.31 Zigzag patterned gold compliant electrodes. (a) zigzag gold electrodes undergoing large strains; (b) structured electrodes [219].](image)

In addition to directly integrate metals to the elastomers surface, a metal reduction reaction was also proposed to fabricate the photo-patternable electrode. In this approach, a transparent metallic salt was first mixed with UV curable elastomer before printing on the target surface.
After curing, this electrode was immersed into liquid chemical reduction solution where the salt reduction took place at the polymer/liquid interface to form thin metal electrodes on the elastomer surface. The reported surface resistance of the platinum electrodes was less than 10 Ω/□ and the electrodes remained effective at strains up to 30%. [228].

Filtered cathodic vacuum arc (FCVA) has been used to implant metal ions to elastomer surface as electrodes [229–231]. In contrast to sputtered or evaporated metal electrodes that lose the conductivity at about 3% strain, ion implanted electrodes have a longer life time at the relatively higher strain level. The implanted metal ions formed nanosize clusters in the first 50nm below the elastomers surface and tend to move toward each other, in-plane, upon stretching of the elastomers. Hence, this electrode remained electrically conductive at strain up to 175%. Rosset et al. fabricated low-energy metal ion implantation on PDMS with three different materials (gold, titanium, and palladium). The gold implantation resulted in the most stable electrode with lowest surface resistance. All electrode types survived more than 10^5 cycles of strain amplitude of 30%. Additionally, electrodes processed through ion implantation generally have semi-transparent optical feature and are considered well-suited for some specific applications, such as tunable lens. However, the ion implantation degree should be highly controlled in order to avoid metal ion penetrate through the film and result in shorting [230,231].

The electrostatic self-assembly approach also has been explored in electrodes fabrications for D-EAP actuators [232–234]. In this method, the surface layer of the D-EAP substrate is functionalized or charged. The substrate is then dipped into a charged solution that has the counter-charges. The counter-charges in the solution self-assemble on the surface of the D-
EAP substrate driven by the electrostatic force. Subsequently, stabilization is obtained with a polyanion and polycation solution. The process is repeated many times, to build up a multilayer thin film on the D-EAP surface. The resultant films have organometallic complex and have been known as “metal rubber”. The unique advantages of metal rubber are the low modulus (Young’s modulus less than 1-10 MPa), low electrical resistance ($10^{-5}$ $\Omega \cdot cm$), good extensibility (may be strained to larger than 100%-1000%) and optical transparency.

In a recent work, the potential of nanosized metals as D-EAP electrode has been explored. Yun et al. fabricated a silver nanowires/polymer composites compliant electrode. The nanowires located in the surface are highly conductive (electrical resistance is less than 10 $\Omega/\square$) and maintain conductivity (electrical resistance ranges of $10^2 - 10^3 \Omega/\square$) at strain levels as high as 140% [235].

### 2.2.3.3 Conductive Polymer Electrode

Conductive polymers (CP) have the inherent potential to impart the soft nature of polymeric materials in electrodes while being electrically conductive. Among the more common CPs are polypyrrole, polyaniline and poly(3,4-ethylenedioxythiophene) doped with poly(4-styrenesulfonate). CPs are generally more mechanically compatible with DEs, however, CP based electrodes suffer from the short lifetime and the low reliability [220]. Polypyrrole has been used as electrodes in polyurethane based DE actuators [194]. This all polymer actuators were flexible with electrode conductivity of 3000 $\Omega/\square$, and the adhesion between the electrodes and elastomer was reportedly very strong.
Lam et al. reported 97% areal strain with the use of polyaniline (PANI) nanofibers, with high aspect ratio, moderate conductivity, as spray electrodes on VHB elastomers. PANI, however, is light sensitive and its electrical resistance increases from 50 kΩ to 300 kΩ after exposure to light [236]. The electrode showed a fault tolerance discussed in more details in section 2.2.3.6. Okuzaki et al. recently used Poly(3,4-ethylenedioxythiophene) doped with poly(4-styrenesulfonate) (PEDOT:PSS) as the electrode of ionic liquid/polyurethane elastomer (IL/PU) composite gels actuator. PEDOT:PSS is sticky and soft after adding xylitol and subsequent heating. With this feature, PEDOT:PSS is able to be conveniently integrated with D-EAPs [237].

2.2.3.4 Electrolyte Solution Electrode

In 2003, Carpi et al. reported large actuation strains at relatively low electric field (20-25V/μm), with NaCl electrolyte as electrodes in VHB actuator [222], see Figure 2.32. It was reported to be difficult to achieve uniform thickness and the evaporation of the electrolyte solution was a problem [220].
Figure 2.32 Peak isotonic transverse strain vs. electric field for the different electrode materials and a pre-stress of (a) 19.6 kPa, (b) 29.4 kPa, (c) 39.2 kPa and (d) 49.0 kPa. [222]

2.2.3.5 Ionic Conductor Electrode

Hydrogels and gels swollen with ionic liquid are commonly used ionic conductors. In contrast to electronic conductors, they are optically transparent, very flexible, and able to sustain large strain magnitudes. All of these features make them promising for D-EAP applications. Recently, polyacrylamide hydrogel containing NaCl electrolyte electrodes with VHB actuators[238] has reportedly produced 167% area strain. A transparent speaker based on this DE-electrode combination was found responsive in a wide frequency range from 20 Hz to 20 kHz.
Although ionic conductors have the higher electric resistance than electronic conductors, they are still promising in application where their ability to sustain large strain and optical transparency may be of value. Furthermore, hydrogels offer choice of diverse, inexpensive, and biocompatible material options to serve a wide range of soft electronics [238].

2.2.3.6 Self-healing Electrode

Self-healing (or fault tolerant) electrodes for D-EAP applications have been reported for a number of materials. “Self-healing” in the context of D-EAP implies that in case of a premature local breakdown of the electrode, the electrode material are evaporated or transformed into an insulator without any melt-through of the DE avoiding any catastrophic failure of the actuator[219,223]. A wide variety of electrodes based on various materials have been observed as self-healing, for example gold[219], inherently conductive polymers [236,239] and carbon[223]. In order to achieve self-healing, the relative thickness of the electrodes compared to the elastomer is important. Ideally, the electrode thickness should be as small as possible so that it is easier for the electrode material to evaporate without creating enough heat necessary for burn-through and breakdown. [219].

Yuan et al. proposed a fault-tolerant electrode based on single-walled CNT [223,239]. The CNTs were dispersed in an aqueous solution and then sprayed at the surface of the elastomer films. As a result of local breakdown, the CNTs located at the edge of the area of breakdown lost conductivity. This self-cleaning feature enabled the actuators to survive with a localized failure.
In summary, different electrodes materials have their own advantages and limitations. The role to select electrodes of D-EAP actuators is based on the elastomer materials used, the configuration of the actuator and the goal of the actuators performance. Table 2.3 summarizes the electrode materials for D-EAPs and representative properties.

Table 2.3 Summary of electrode options for dielectric elastomers [220]

<table>
<thead>
<tr>
<th>Electrode</th>
<th>Resistance per unit length</th>
<th>Application</th>
<th>Advantages</th>
<th>Limitations</th>
</tr>
</thead>
<tbody>
<tr>
<td>Carbon grease [222]</td>
<td>50 kΩ/cm</td>
<td>Thin film deposition</td>
<td>Commercially available, good electromechanical coupling efficiency</td>
<td>Difficult to obtain uniform thickness, subject to evaporation</td>
</tr>
<tr>
<td>Graphite powder [222]</td>
<td>80 kΩ/cm</td>
<td>Screen printing</td>
<td>Moderate performance</td>
<td>Difficult to obtain uniform thickness, requires adhesive surface or binding agent</td>
</tr>
<tr>
<td>Graphite spray [222,240]</td>
<td>20 kΩ/cm</td>
<td>Spray</td>
<td>Good performance at high voltages, easy to apply, uniform coating</td>
<td>Requires adhesive surface or binding agent</td>
</tr>
<tr>
<td>Method</td>
<td>Thickness</td>
<td>Deposition Method</td>
<td>Conductivity</td>
<td>Notes</td>
</tr>
<tr>
<td>------------------------</td>
<td>-----------</td>
<td>------------------------------------------</td>
<td>--------------</td>
<td>--------------------------------------------</td>
</tr>
<tr>
<td>Thickened electrolyte</td>
<td>15</td>
<td>Thin film deposition</td>
<td>Good</td>
<td>Not commercially available, difficult to obtain uniform thickness, subject to evaporation</td>
</tr>
<tr>
<td></td>
<td>[222]</td>
<td></td>
<td>performance at low voltages</td>
<td></td>
</tr>
<tr>
<td>Patterned metallic</td>
<td>-</td>
<td>Sputtered deposition and patterned photolithography</td>
<td>Metallic electrode capable of strain up to 80%</td>
<td>Requires specialized equipment, more labor-intensive</td>
</tr>
<tr>
<td></td>
<td>[115]</td>
<td></td>
<td></td>
<td></td>
</tr>
<tr>
<td>Corrugated metallic</td>
<td>-</td>
<td>Micromachining, physical vapor deposition</td>
<td>Anisotropic, limit strain to a single direction</td>
<td>Requires specialized equipment, limits strains to 33%</td>
</tr>
<tr>
<td></td>
<td>[227]</td>
<td></td>
<td></td>
<td></td>
</tr>
<tr>
<td>Sputtered gold</td>
<td>2.4</td>
<td>Sputtered deposition</td>
<td>Thin electrode layers are possible</td>
<td>Degradation in short period of time, cracking at critical strain ( &lt; 20%)</td>
</tr>
<tr>
<td></td>
<td>[240]</td>
<td></td>
<td></td>
<td></td>
</tr>
<tr>
<td>Silver paste</td>
<td>1.6</td>
<td>Thin film deposition</td>
<td>Good</td>
<td>Difficult to obtain thin layers, subject to evaporation</td>
</tr>
<tr>
<td></td>
<td>[240]</td>
<td></td>
<td>conductivity</td>
<td></td>
</tr>
<tr>
<td>Conductive polymer [240]</td>
<td>51 kΩ/cm</td>
<td>-</td>
<td>Good compliance and elastic properties</td>
<td>Poor long-term performance and reliability</td>
</tr>
<tr>
<td>-------------------------</td>
<td>------------</td>
<td>---</td>
<td>---------------------------------</td>
<td>---------------------------------</td>
</tr>
<tr>
<td>Platinum salt [241]</td>
<td>$10^6 - 10^9$ μΩ/cm</td>
<td>Photolithography</td>
<td>Amenable to micro-scale, low-strain D-EAP applications, photo patternable</td>
<td>Limits strains to 40%, film resistance decreases significantly with strain, inconsistent manufacture</td>
</tr>
<tr>
<td>Self-healing electrodes [115]</td>
<td>-</td>
<td>Thin film deposition</td>
<td>Limits breakdown, exploratory-potential for future advantages</td>
<td>Requires thin layers, poor long-term performance, poor adhesion to elastomer materials</td>
</tr>
</tbody>
</table>

### 2.2.4 Actuators Design and Applications

D-EAPs, with large electric field induced strain, high electromechanical coupling efficiency and fast response, have been employed to develop a wide range of actuators. The soft nature of D-EAPs and the compliance of electrodes together enable the design and fabrication of actuators that are highly flexible. In the late 1990s and early 2000s, the Stanford Research Institute (SRI) developed many D-EAP-based actuators with different configurations, such as
unimorph, bimorph, bowtie, diamond, roll, tubular and folded types [168]. Chapter 3 will focus more on the linear actuators that can generate the actuation motions in the straight line manner. Although the configurations of the actuators are different, they possess some common design elements[8]. Firstly, in all of the designs the stiffness of the elastomer is a major design parameter to control actuation strain and the elastic strain energy density. Secondly, the dimension of the actuator is one other crucial parameter. It should match the requirements of the application and be able to produce the required stress/strain output. For example, the magnitude of stroke and specific elastic energy density (usually normalized by the mass of the actuator) are very dependent on the size of the actuator. In addition, it is necessary to consider the mechanical damping and creep of the elastomeric material. For example, the mechanical loss tangent of silicone is less than 0.05 (tested at 80Hz) but that of acrylic is 0.18 (20 Hz). Computer modeling is a powerful tool to assist design of actuators. It helps to anticipate the actuation performance before fabrications. Zhao and Suo developed a computer simulation of programmed 3D deformation of D-EAP actuators on the basis of the fundamental theory of elastic dielectrics [242]. The program uses finite element modelling (ABAQUS) to predict a eigen-strain from the prescribed nominal electric field input based on the free-energy function. The sample simulation of this model successfully predicted the formation of wrinkles, movement of actuators (e.g. hand like actuator) and nonlinear oscillation of a balloon shaped actuator. Interestingly, D-EAPs also have also been explored to work with a secondary system. This approach broadens the variety of D-EAPs actuators design and realizes safe electrically transmission of forces to loads [217,243]. For example, Carpi et al. proposed a system consists
of D-EAP actuators as an incompressible fluid. The fluid works as the media to transit the motion to a passive component, acting as the end-effector. On the basis of this idea, a millimeter sized hydrostatic coupling bubble-like actuators were built [244]. Later, a tunable lens was proposed with two layers of D-EAP films and the fluid sealed inside [245].

In application of D-EAP actuators, various representative devices have been explored. These include motors, loudspeakers, robotic [3], animatronic or prosthetic actuators [220], tunable optical devices [246], micro electro mechanical systems (MEMS) and biologically imitations (artificial skin or muscles) etc. Most of these devices have been extensively explored based on the Most of the devices mentioned here have been already well developed based on the conventional transducers, such as electromagnetics, piezoelectrics or shape memory alloys. However, the D-EAPs give opportunities to possess high specific energy density, lighter weight, higher flexibility, lower cost, more variety of shapes and dimensions and ability of combination of sensing and actuation. Among all of the potential applications, D-EAPs actuators most have been explored in field of artificial muscles as shown in Figure 2.33.

Figure 2.33 Comparison of high-speed actuators technologies Linear Actuators [247]
CHAPTER 3 Dielectric Electroactive Polymer Linear Actuators

3.1 Introduction

Linear actuators are capable of generating motion (and force) along a straight line. With direct transmission of thrust or contractile stroke, linear actuators have been used in a wide variety of diverse applications [248]. In principle, linear motion can be converted to rotary movement, by specific transmission mechanism, which further broadens the use of linear actuators [249]. Currently, there is a wide range of commercially available linear actuators, including mechanical, hydraulic, and pneumatic types. Commercially available materials based actuators, for example, have “SmartMOVE D-EAP actuator” manufactured by Artificial Muscle Inc. and the “InLastor actuator” manufacture by Danfoss Polymer A/S company [8] etc. For obvious reasons, various D-EAP based actuator configurations have been proposed, see Figure 3.1. Many of these designs are inspired by existing actuators based on piezoelectric materials [8]. In the following, the linear D-EAPs actuators will be categorized by their configuration, such as extender, stretched film, bowtie, diamond, stacked, helical, folded, rolled, fiber-like or cylindrical, and cone shape. In all cases, the primary objective is to harness the thickness strain of the D-EAP film and convert it to directional linear displacement as efficiently as possible. In this chapter, for each type of actuator, we discuss their working principle, performance characteristics, processing techniques, and potential applications.
3.2 Linear Actuators

3.2.1 Extender Structure

The extender structure is arguably the simplest actuator design to convert the in-plane extension of the D-EAP film into linear motion, see Figure 3.2. In this design, flat layers of dielectric elastomer films are assembled in a laminated structure, with conductive layers placed between each layer to act as electrodes. In this manner, the laminates are able to extend in the horizontal plane as the electrical potential is applied with appropriate polarity to each layer simultaneously [114,115]. This structure can also act as a bending actuator when the potential
is applied such as to create a strain gradient through the thickness of the actuator. It was able to bend more than 180° [114]. The disadvantages of this design are many. The practical length of the actuator is limited by the free length of the cantilever that is able to support itself. It is also difficult to apply and maintain the necessary uniform prestrain in the D-EAP membranes.

![Diagram of Extender Configuration](image)

Figure 3.2 Extender configuration for linear actuation [115]

### 3.2.2 Stretched Film Structure

This design, takes advantage of the areal extension of the D-EAP, with a very simple design that can generate linear strain up to 20% [250] as shown Figure 3.3 shows. One unitary prestrained layer of elastomer film, with two separate active electroded areas and an inactive intermediate space with an attached end-effector, is fixed to a frame. As one side (farther) of the film is stimulated, the film expands so that the other non-actuated part contracts, producing a linear movement of the end-effector in the horizontal plane.
The smallest stretched film actuator with thickness of 200 µm has been reported as a component in a micro-electromechanical system (MEMS) [114,250]. Arrays of this actuator (3×3) were fabricated in a single monolithic structure to improve the force output and flexibility of movement [250]. Choi et al. reported a backbone-shaped robot which was driven by several antagonistically-driven linear actuators (ANTLA) of this kind [251–253], see Figure 3.4(a). Each ANTLA is capable of producing combinations of four working states based on the selective electrical stimulation. For example, the movement to the right happens when electrodes A and C (bottom electrode) are activated. Using these combinations of segmented actuators, a spine shaped bending device was designed, see Figure 3.4(d). In this system, three ANTLA devices are equidistantly located in along the circumferential direction of a cylindrical
frame. Upon actuation of an ANTLA, the corresponding elastomer expands and then pushes one flexible upper circular part. By superimposing these strokes from all of the cylindrical parts, a bending movement is realized.

Figure 3.4 Antagonistically-driven linear actuator (ANTLA) and backbone shaped bending devices driven by ANTLA. (a) working principle of ANTLA; (b) four working statuses; (c) ANTLA integrated with output shaft; (d) backbone shaped bending devices driven by ANTLA. [253]

In this design, uniformity of the prestrain affects the linearity of the movement. In the other words, the rectangular electrode area may not evenly expand due to nonuniform prestrain and thus the output shaft could move upward or downward rather than moving solely linearly along the horizontal direction. In addition, the output force may not be robust enough due to limited
thickness of the elastomers. The interface between rigid shaft and soft elastomers may also cause problem in transferring the load.

### 3.2.3 Bowtie Structure

In this design a prestrained and electroded D-EAP film is attached to and supported by a bowtie-shaped rigid spring-like frame, which connects the two halves of the bowtie with a flexible hinge in the middle. Upon application of the electric field, the elastomer film begins to extend towards the frame such that the hinges open up outward to produce a linear displacement in the cross direction, as shown in Figure 3.5a.

![Image of Bowtie Configuration Actuator](image)

**Figure 3.5** Bowtie configuration actuator. (a) Working mechanism of bowtie actuator; (b) silicone bowtie actuator; (c) multilayer double bowtie actuator with acrylic film [8]

Bowtie actuators have been employed to provide motion to a hexapod walking robot, flapping wing robots, and a variety of animated systems such as eyes, face, skin etc.[254]. For instance, a number of bowtie actuators based on acrylic D-EAP have been used as “muscles” of a hexapod walking robot as shown in Figure 3.6(a) [255]. The robot can lift its feet and change the walking direction. In another example, the bow-tie actuator has been used in a flapping wing robot, see Figure 3.6(b). A bowtie actuator is placed inside the “thorax” of the flying
robot and is intended to create up and down motion and drive the wings. The power densities of both silicone and acrylic elastomers were more than enough to lift this device, but due to relatively heavy frame (80-90%wt) the potential power density of the whole system decreased significantly. Therefore, low overhead in packaging D-EAP actuators is a key point in designs of biomimetic devices with high overall power density [247].

![Figure 3.6 Bowtie D-EAPs actuator used in biomimetic applications [247]. (a) acrylic bowtie shaped actuator in self-contained six-legged robot “FLEX”; (b) flapping wing thorax-type design using D-EAPs](image)

**3.2.4 Diamond-shaped and Similar Structures**

In contrast to bowtie actuators, the frame of the diamond-shaped actuators has two more hinges at the top and bottom vertices, see Figure 3.7. The actuation principle of diamond-structured actuators is based on four-bar linkage mechanism, with one the vertices fixed in plane, to produce displacement with an axis of symmetry. The system can provide a nearly constant actuation force and therefore ideal for many haptic devices, mobile robots and others [256].
There are mainly two types of proposed diamond configurations using D-EAP. One uses a diamond-shaped plastic frame with links of equal length, and the other makes use of a non-linear tension spring (Figure 3.7 (a)). This spring, called “negator,” provides a contractile force and enhances the actuation force. When the frame is “open” state, the stored energy in the extended spring tends to contract back to original status so it brings larger output force [257].

Alternately, a highly elastic elastomer band with low hysteresis and weight can also be used (see Figure 3.7(c)). By simply changing the spring constant or elasticity of rubber band, actuators could have the desired output force and displacement [258]. Another type of diamond-shaped configuration integrates a compliant “delta” element to serve as the “negator” (see Figure 3.7(b)) [259].

Figure 3.7 Diamond-shaped actuator. (a) schematic of four-bar mechanism based actuator with “negator” [258]; (b) schematic of a compliant symmetric double slider-crank mechanism with elastic joints on the slider pivot [259]; (c) assembly and prototype of acrylic diamond-shaped actuators [257]
Hexagonal (Figure 3.8 (a)) and racetrack-shaped (Figure 3.8 (b)) linear actuators that work in the same manner as the diamond-shaped have also been proposed. For example, a hexagonal-shaped actuator has been used to drive a rehabilitation device. In that, two the upper and bottom flexible working frames were connected by the plastic edge as a restorative force, see Figure 3.8. This monolithic structure produced an increased stress at the hinges due to their continuity. However, the stiffness of the nylon frame under compression changed as a function of time, over three days, and resulted in about 67% loss in available force, see Figure 3.8(d) [258].

![Figure 3.8](image)

Figure 3.8 Different designs for the flexible frame. (a) hexagon structured frame; (b) racetrack shaped frame; (c) prototype hexagonal structured actuator with and without actuation [260]; (d) stress relaxation of the nylon frame induced loss of blocking force as a function of time [258].

### 3.2.5 Stacked Structure

In contrast to the actuator designs discussed thus far, stacked actuators are contractile. They are designed to utilize the negative strain in the thickness direction of the D-EAP films. In this respect, stacked actuators are similar to the actuation of natural muscles.
In a stacked actuator, layers of appropriately electroded D-EAP films are stacked in a manner shown in Figure 3.9 (a). Under Maxwell stress, the D-EAPs films reduce in thickness and expand in plane. The superimposed thickness strains results in the overall contractile strain of the actuator [261][167,262].

Figure 3.9 Stack configuration. (a) schematic of D-EAPs stacked actuator before and after actuation [262]; (b) stacked actuator consists of alternating electrodes and elastomer [261]

In a tactile display based on stacked D-EAPs actuators, the electrodes on alternating layers are placed in a manner shown in Figure 3.10. Upon activation of specific stacks, dimple-like depressions are produced to create a tactile texture [262,263].
Figure 3.10 Tactile display with stacked configuration. (a) the schematic drawing of the elastomer stack with patterned electrodes; (b) D-EAPs contracts at the actuated point with can inducing tactile sensation of finger; (c) passive matrix arrangement of the electrodes [262,263]

This tactile display has been analyzed through equivalent circuits and other mathematical models, to elucidate the role of various parameters including the thickness of individual elastomer layers, pattern and dimension of the electrodes, electrical resistance of electrode materials and electrical stimulation frequency, etc. [264] A 20-layer PDMS-based stacked actuator with each layer of 50 μm thickness generated 40μm contraction under an applied 200Hz sinusoidal electrical field of 1300V. This relatively small strain was considered within the range of human sensitivity [264]. Many other practical applications of this tactile actuator, such as vibrotactile display interface of a mp3-player [265,266] and peristaltic pump have been explored (see Figure 3.11) [267].
The most challenging problem of tactile display made from stacked linear actuators is the long discharge time induced “crosstalk”. The arrangement of electrodes with periodically switched column and row lines are necessary to produce the distributed actuation points, however, because of the long discharge time, the residual charges from previous cycle interferes with the next stimulation cycle. As a result, wrong information or noise, termed as crosstalk, is produced. The same problem in organic light-emitting diodes (OLED) has been addressed through the use of active matrix to individually control each electrode with patterned thin-film transistor (TFT) [262].

The decrease in thickness in a stack D-EAPs actuator is accompanied by in-plane expansion. This is particularly evident in case of actuators with two rigid boundaries at the ends, see Figure 3.12 (a) and (b). As a result, the elastomer films in the middle tend to bulge out [268]. In order to eliminate this inhomogeneous strain distribution, rigid electrodes have been investigated. Lau et al. fabricated stack D-EAPs actuator with rigid silicon electrodes (See Figure 3.12 (c))
and reported large force and adequate strain under moderate voltages [269]. In another approach, perforated nickel plate electrodes were found suitable in stacked actuators [268,270], see Figure 3.12 (d). By substituting some perforated electrode with completely closed nickel layer, the constraining effect was reportedly improved further. This work also suggested that rigid electrodes are able to help retain prestrain in the elastomer and avoid “pull-in” instability [269,270]. The other solution for “bulging” problem is to keep some inactive edge around the electrodes in every layer [261]. But this passive area may increase the system stiffness and limit the maximum achievable actuation strain [270,271].

Figure 3.12 Rigid electrode used in stack D-EAPs actuators. (a) disadvantageous bulging out of middle layers of actuator with compliant electrode [268]; (b) D-EAPs stack actuator with fixed end [261]; (c) multilayer PDMS actuator with rigid doped silicon electrode [269]; (d) general deformation morphology of elastomer with rigid and perforated electrodes, symmetrical setup (up) and asymmetrical setup (bottom) [270]
Besides tactile displays, stacked actuators have been designed for other potential applications. Chuc et al. fabricated circular, rectangular, and trapezoidal shaped stacked actuators, as shown in Figure 3.13 (a-c) [10,271]. Figure 3.13 (a) displays the circular stacked actuator, actually the individual D-EAPs is annular. In the hollow center of the annular elastomers, there was a shaft to support flexible elastomers without incline as actuation. And a compressed spring was assembled in the end of the stack of elastomers. It is used to compress the elastomers and thus give higher prestrain and enlarge the overall displacement. In order to individually control a certain “bundle” of stacked elastomer, separating used for electric potential application were assembled as spacers in the circular shaped actuator. In the rectangular shaped actuator, each layer of electrode D-EAP film is folded from a larger piece and can be controlled independently (see Figure 3.13 (d)). The rectangular actuators showed better performance in comparison with the circular due to having less inactive area surrounding the electrode with folded design. The trapezoidal shaped has a lower fraction of inactive region due to their planar shape as well as folded design, and thereby achieved larger strain magnitude. However, this folded configuration also has drawbacks because of its asymmetrical structure. The free ends always have larger deformation than areas adjacent to the inactive areas. Additionally, the folded structure may easily bring about nonuniformity of thickness [272]. Using synthetic acrylonitrile butadiene rubber as D-EAP, these assemblies also were proposed as load sensors [273].
Figure 3.13 Stacked actuators with different configurations. (a) circular shape [271]; (b) rectangular shape [271]; (c) trapezoidal shape [274]; (d) strain versus electrical filed of three types of actuators [275]

Stacked actuators together with slider crank mechanisms have been used to design fingers and legs robots with multiple degrees of freedom, see Figure 3.14 (a) and (b) [272,274]. The flexing of joints is achieved by activating the appropriate stacked actuator to work on the slider crank at a maximum angle of about 65° (one degree of freedom (DOF)). Through a series of combinations of these actuator-crank assemblies multiple degrees of freedom can be achieved to produce multi-joint fingers. Robot fingers with electronic controllers could give fast response to simulate human-hand-like movement. The weight lifting performance of this robotic finger is illustrated in Figure 3.14 (b). Thus D-EAP based soft robot fingers with muscle-like features was demonstrated as a new and promising paradigm for future robot manufacture [275].
A wide variety of D-EAP materials have been used in stacked actuators, including acrylic elastomers [277], acrylic-based interpenetrating polymer networks (IPN) [261,278], silicone [269,279,280], and synthetic acrylonitrile butadiene rubber [273]. The acrylic IPN produce large strains (46% non-loaded) and high energy density (12.9J/Kg), however, the processing is complex and the amount is limited. Silicones produce lower strain magnitude (< 20μm per 1.2mm device). Recently, UV-cured poly (urethane acrylates) (PUA) was proposed that does not require prestrain in the actuator fabrication process and can generate approximately 11% load-free strain [281,282].

With all its promise, the fabrication of multilayer stacked actuators remains a challenge. Various approaches for improving the efficiency and the consistency of fabrication have been proposed. Alternating spin-coating of D-EAP film and electrode deposition on the surface can
be used to stack hundreds of elastomer layers in a short time with good consistency [280,283,284]. Also, inspired by processing techniques of roll-shaped polyester capacitors, a winding method was proposed in which elastomer films already deposited with electrodes were first wound on a removable core and then cut to certain lengths [285]. This cut smaller pieces can be used in stacked structure. The mandrel in winding has different shapes, including flat, octagonal and circular. For fabrication of electrodes in a certain pattern with high accuracy, spraying of electrode solutions or powders through a stencil mask onto elastomers have been widely used [272,279].

In summary, compact and the high weight-to-force ratio contractile stacked D-EAP actuators, have been designed and evaluated for a wide variety of applications such as tactile display devices, artificial muscles, gas and pneumatic valves [278,286], active suspensions [287] and adaptive absorber systems [288]. However, with the multilayer configuration, stacked actuators still have some limitations in terms of processing and interaction between neighboring layers. It is not easy to achieve high prestrain levels necessary to reduce the required electric field for activation while avoiding early electrical breakdown of the device [262].

3.2.6 Helical Structure

In a helical actuator, a narrow strip of D-EAP film with electrodes on the helical plane form a helix, akin to a spring, see Figure 3.15 (a). Upon activation, the elastomeric film contracts longitudinally and acts like an electrically actuated polymer spring. The overall displacement of this actuator is adjustable by changing the helix angle (α), see Figure 3.15 (b). For obvious
reasons, larger helix angle results in higher overall axial strain of the actuator. In contrast to stacked actuators, helical actuators use continuous electrodes and in principle have lower chance of failure due to shoring between two neighboring electrodes [289].

![Image of helical actuator](image)

Figure 3.15 Helical “polymer spring” actuator. (a) schematic illustration of structure and operation of a helical D-EAPs actuator [290]; (b) prototype of spiral actuator, the “skeleton” consists of spiral silicone and carbon loaded silicone electrode (top); after filled skeleton with silicone (bottom) [289]

A convenient way to fabricate helical actuators is shown in Figure 3.16 [291]. In the first step, a spiral “spring” (Figure 3.16 (b) and (g)) is cut from a silicone tube (Figure 3.16 (a) and (f)) and then coated with electrodes of carbon clack loaded silicone on the two faces (Figure 3.16 (c) and (h)) to form the “skeleton” of the actuator. This helical skeleton is subsequently combined with a naked helix (without electrodes), to assemble the final tubular shaped actuator (Figure 3.16 (d-e) and (i)). Using this configuration, Capri et al. reported contractile actuation strain in the range of 0.5%-5% under the electric field of 4-15 V/μm [289–291].
In an interesting application, helical actuators have been used in providing motion to eyeballs in an android face [292]. The project dubbed as, Facial Automaton Conveying Emotions (F.A.C.E.), an android face endowed with dynamic expression and artificial vision was required. Two helical D-EAP based pseudo-muscular actuators were used in agonist-antagonist configuration as “ocular muscles”, see Figure 3.17 (a). In this assembly, each eye consists of an eyeball hinged on the central vertical axis and connected to two actuators by inextensible tendon-like wires as shown in Figure 3.17 (b). While one of these two linear actuators a contract, the eyeballs rotate towards the actuated side. The preliminary
mathematical modeling show that the eyeballs driven by two helical linear actuators can rotate up to approximately ±25° with 54mm antagonist length.

![Figure 3.17 Eyeballs driven by helical linear contractile actuators. (a) prototype of eyeballs for android robot face; (b) schematic drawing of the configuration of eyeball muscles [292]](image)

### 3.2.7 Folded Structure

The folded actuator is a monolithic compact structure and functionally equivalent to the multi-layer stacked actuators discussed earlier. Because of continuity of the D-EAP film form layer to layer, folded actuators eliminate the difficult fabrication processes of stacked actuators, see Figure 3.18. The folded structure is also preferred over the helical configuration because of the ease of processing [293].

Carpi et al. built and evaluated prototype of folded actuators from strips of silicone appropriately coated with electrodes on both sides [293,294]. The folded stack was subsequently sealed along the exposed sides with electrodes, see Figure 3.18 (c). The actuator was able to generate contractile strain as high as 15% under an electrical field of 12V/µm.
Using the same fabrication technique, stacked actuators of other cross-sectional shapes could easily be assembled. For instance, with alternate circular D-EAP strips, a cylindrical folded actuator was built.

Figure 3.18 Folded D-EAPs actuator configuration. (a) schematic drawing of folded D-EAPs actuator and prototype [293]; (b) axial strain as a function of the applied field [293]; (c) folded actuator with circular cross-section [294]

The folded actuators have been evaluated in driving android eyeballs [293,295] (Figure 3.19 (a)) as well as in adjustable “smart” hand rehabilitation splints (Figure 3.19 (b)) [296,297]. The force generated by a silicone folded D-EAPs actuator is about 1N at about 6KV. Additionally, folded actuators have also been explored in other applications, such as man-machine interfaces,
deployable booms in space (solar cells, antenna systems, and cameras etc.) and bidirectional tilters as illustrated in Figure 3.19 (c-d) [296].

Mechatronic systems equipped with folded D-EAPs actuator performed with magnetic resonance imaging (MRI) have been investigated [298,299]. It highlights this contractile actuator could perform properly under harsh magnetic field and has no obvious disadvantageous effect on result images. Also, by parallel arranging stacked actuators and actuating them in sequence, a traveling wave or peristaltic motion was achieved [300]. This demonstrates arrays of linear actuators are on option to functionalize actuators.

Figure 3.19 Application of folded D-EAPs actuators. (a) android eyeballs [295]; (b) hand rehabilitation splints [297]; (c) bi-direction tilter [296]; (d) deployable boom [296]; (d) man-machine interface
3.2.8 Rolled Structure

In all of the actuator structures discussed thus far, a common problem is the difficulty in application and retention of prestrain in the D-EAP film. The roll-shaped D-EAPs actuators help address this problem. In rolled actuators, two adjacent layers could constrain each other and thus retain the applied prestrain. Rolled actuators are compact, free standing and generally capable to produce high power, and large-strain. Due to the interlayer constraint in the presence of prestrain-induced hoop stress, the rolled actuators are structurally more robust in that, small defect or crack in one layer do not trigger catastrophic mechanical failure of the whole roll system and the chances of mechanical breakdown is lower [12].

Rolled actuators proposed thus far can be categorized in two: spring roll actuators (that combine a pretensioner (spring) in the core), and core-free roll actuators.

3.2.8.1 Spring Roll Structure

In the spring roll structure, a compressed spring is incorporated inside the roll formed by the prestrained and appropriately electroded D-EAP film. The compressed spring not only helps prestrain the D-EAP roll longitudinally but also enhance strain upon activation as well as help assist retraction of the actuator as the power is turned off [12]. The fabrication process involves, two layers (usually) of bi-directionally prestrained elastomer films laminated and sandwiched with a middle layer of electrode. In the next step, electrode is applied to one of the sides of the elastomer laminate. In the final step, the laminate is rolled around a compressed spring core and the roller edge of the elastomer laminate is secured at the corresponding ends of the spring. Figure 3.20 illustrates this fabrication process. In the free standing state, the whole system
reaches a critical force equilibrium where the extensive force of the spring balances the contractile force of the elastomers [12].

Figure 3.20 Fabrication process of spring roll actuator [12]

The performance of spring roll actuators depends on a number of crucial parameters that determine their overall actuation performance. These parameters include, component dimensions (spring diameter, film thickness, etc.), spring constant, numbers of D-EAP layers, the Young’s modulus of the elastomer, level of prestrain in the D-EAP film, etc. [12]. Besides some of these intrinsic parameters, the operational conditions, such as frequency and prestrain amplitude are important to the performance of the actuator as well [12]. Acrylic spring roll actuators undergo considerable reduction of actuation strain at frequencies beyond 5 kHz and, interestingly, it has also been reported that the lifetime goes down from 1.1 million cycles at 3.3 kV, without any observable degradation to failure after 36,000 cycles at 4 kV [12].

Understanding of the force-strain equilibrium in a passive spring roll structure is the key to the design and impedance matching of the actuator. Zhang et al. analyzed the four stages of the
spring roll actuator, shown in Figure 3.21 (a) [118]. This working principle analysis assumes that elastomer is incompressible, isotropic and its mechanical behavior is quasi-linear under application of prestrain condition.

Figure 3.21 The working principle of D-EAPs spring roll actuator[13,118]. (a) four states of the spring roll actuator based on quasi-linear mechanical behavior of D-EAP [118]; (b) the force-displacement of D-EAP with non-linear mechanical behavior [13]; (c) passive equilibrium of loads in different directions in the spring roll actuator [13]

a) **Assembly stage**: the initial state is attained after applying and securing the prestrained elastomer film around the compressed spring, before the spring is released.

b) **Passive equilibrium stage**: once the applied force on the assembly is removed part of the spring’s compressive strain is released until reaching force equilibrium with the rolled film wherein the contractile force of the elastomer equals to the expansive force of the spring.”. As Figure 3.21 (c) displays the force equilibrium of this actuator. In the axial direction, spring extension force balances the contractive force of D-EAPs. And
in the circumferential direction, the prestretched elastomer is supported by the spring core.

c) *Free-strain stage*: as an electrical field is applied to the elastomer, the film layers are compressed in the thickness direction and the spring roll undergoes longitudinal extension and radial expansion. The spring extends until reaching the next force equilibrium status called “Activated equilibrium”. When the electrical field is removed, the elastomer film contracts back to original dimensions and compresses the spring until it returns to the “Passive equilibrium” state.

Later, this model was improved by substituting the linear material properties with the non-linear, see Figure 3.21 (b).

One of the most well-known applications of spring roll actuators is in the locomotion of a walking robot, see Figure 3.22 [12]. Inspired by Sprawlita pneumatic robots, Pei et al. assembled six spring roll actuators based on acrylic elastomers with one DOF as legs to drive a robot that can walk at a speed up to 7cm/s [6]. The preliminary experimental data and theoretical simulations both suggest that the walking velocity greatly depends on the strain and strain rate of the “legs”. While the actuation displacement was 10mm at 1Hz frequency, the robot walked faster at the speed of 20mm/s. The response speed of the acrylic spring roll actuators was found suitable at 10Hz or lower due to significant viscoelastic losses of acrylics.
D-EAPs spring rolls also were used in robotic hands. Jung et al. used four concentric layers of cast silicone elastomer tubes with intervening electrodes and a compressed spring in the center, to form a spring roll-like actuator [301]. The subsequently used the actuators in a multi-fingered robot hand as shown in Figure 3.23. Linked by pulleys, a pair of antagonistic parallel actuators was connected to emulate the bending motion of human fingers. The prototype finger showed an independent bending to approximately $15\text{-}30^\circ$ angle.

Figure 3.23 Tube-spring actuator. (a) tube-spring actuator with silicone tubes enclosed compressed spring; (b) multi-fingered robot hand driven by tube-spring actuators [301]
Another example of spring roll actuator is a portable force feedback device [13,118]. Figure 3.24 displays a prototype portable force feedback glove based on spring rolls. In preliminary experiments, several proof-of-principle assemblies were fabricated and tested. This device (38g) generated the blocking force up to 7.2N and the maximum longitudinal strain of 31% (5mm displacement). It highlights the potential of this force feedback device. In principle, force feedback devices should be in physical contact with human body therefore safety issues caused by the high working electrical potential remain an obstacle for the further commercialization.

![Attachment to the finger]

Figure 3.24 Portable force feedback glove by connecting actuator between fingers [13,118].

As a demonstration of the potential of D-EAP in robotic and bioengineering applications a robotic arm driven by bundles of spring roll actuators was fabricated to go against a human opponent [11], see Figure 3.25. Although the robot lost, the D-EAPs actuators demonstrated its huge potential as artificial muscle.
Figure 3.25 Prototype and working principle of the actuator bundles as the robot arm [11]. (a) wrestling robot with four actuator banks, each has 64 actuators; (b-c) the working principle of the robot arm.

So far, the discussion has been limited to spring roll actuators with one degree of freedom (DOF). The basic design was modified using patterned electrodes, for selective stimulation and thereby achieve multiple DOF. Figure 3.26 (a) shows an actuator with 2 DOF. It is fabricated by applying electrode in segments on both sides of the D-EAP film in such a manner that after rolling the electrodes form two distinct segments, see Figure 3.26 (a). When one of the segments is activated, the elastomer on that side is subjected Maxwell stress and extends. Because of the differential segmental strain, the actuator bends. Likewise, if the electrodes are patterned in four segments the actuator would have 3 degrees of freedom (Figure 3.26 (b)). Multiple DOF broadens the application potential of spring roll actuators. For instances, by substituting the 1-DOF actuators with the same with 2-DOF the walking robot is able to move in any direction as shown in Figure 3.26 (b).
3.2.8.2 Core Free Roll

In an effort to reduce the overall weight of the spring roll actuator and thereby increase the energy density, in this design the spring is removed from the core. Additionally, in absence of the spring, the inner layers do not experience extremely high compressive forces and therefore chance of early failure in the inner layers is reduced. Kovacs et al. proposed a core free roll actuator using the interpenetrating polymer networks (IPN), discussed earlier in section 2.2.1.7 [304]. This actuator was simply assembled by rolling up the IPN acrylic film without a core or only with a very small soft elastic core. Without the bulky spring, the cross section of this actuator consists only of active elastomer layers with no limitation in terms of overall number of IPN elastomers. Experimentally, however, the IPN
acrylic roll had about 10% strain under 6kV electrical potential which is much less than the maximum strain (up to 35%) of spring roll actuators [304].

Another prestrain free rolled actuator, proposed by Benslimane et al. used the corrugated metallic electrode that induce anisotropic behavior of D-EAP film [227,305], more details is described in section 4.2.2.

Figure 3.27 Core-free and self-supporting roll actuator [306].

Fiber reinforced elastomers also have been investigated for core-free roll actuators. Huang et al. used equal-spaced aligned nylon filaments to reinforce VHB films to fabricate roll actuator, see Figure 3.28 [307]. The relatively rigid filaments are used to constrain the deformation in the radial direction of the elastomer film and maximize the displacement in the other direction. The prestrained D-EAP film was supported by two rigid tube ends and rolled up to form the actuator. The cylindrical actuator could generate large strain (about 30% linear strain) even without prestrain. It was noted that the fiber stiffened elastomer can effectively eliminate the cross section bulging problem that normally occurs for cylindrical actuators upon actuation [308]. However, with limited number of layers of the elastomer film, this cylindrical device
could only generate relatively small output force. More details about this actuator is included in section 4.2.2.

Figure 3.28 Fiber reinforced D-EAPs roll actuator [307]. (a) aligning nylon filaments to reinforce VHB; (b) laying the second layer of VHB on the surface of the first fiber reinforced one; (c) applying carbon grease on both surfaces of laminates and rolling; (d) complete actuator with leads

In another design of core-free silicone roll actuator, an external shell made of carbon fiber polymer composite is used for applying prestrain and magnifying the stroke in the longitudinal direction, see Figure 3.29(a)-(c) [309]. The weight of the shell (24.3% of total) is significantly smaller than the spring core (90% of total) in the roll actuator, as a result its work density is relatively higher than VHB spring roll actuators (about 10-20%). This design was successfully integrated into an insect-inspired thoracic mechanism to mimic exoskeleton musculature and drive a pair of flapping wings.
In summary, D-EAPs roll actuators are classified into two categories, one type has core such as spring and the other type is core-less. Table 3.1 summarizes reported properties of several roll actuators.
Table 3.1 Comparison among various rolled D-EAPs in literatures (reference [309])

<table>
<thead>
<tr>
<th></th>
<th>Pei et al. [12]</th>
<th>Zhang et al. [13]</th>
<th>Rajamani et al. [310]</th>
<th>Benslimane et al. [305]</th>
<th>Lau et al. [309]</th>
</tr>
</thead>
<tbody>
<tr>
<td>Dielectric electroactive polymers (D-EAPs)</td>
<td>Acrylic elastomer</td>
<td>Acrylic VHB 4910</td>
<td>Acrylic VHB 4905</td>
<td>Wacker silicone ELASTOSIL RT 625</td>
<td>Silicone BJB TC-5005</td>
</tr>
<tr>
<td>Electrode materials</td>
<td>Carbon black mixture</td>
<td>Graphite powder (TIMREX LB1300)</td>
<td>Carbon grease</td>
<td>Corrugated silver</td>
<td>Graphite powder (TIMREX KS6)</td>
</tr>
<tr>
<td>Number of layers</td>
<td>20-35</td>
<td>30-40</td>
<td>15</td>
<td>~64</td>
<td>4</td>
</tr>
<tr>
<td>Roll diameter</td>
<td>12 mm</td>
<td>12 mm</td>
<td>12 mm</td>
<td>--</td>
<td>11 mm</td>
</tr>
<tr>
<td>Roll length</td>
<td>70 mm</td>
<td>45 mm</td>
<td>16 mm</td>
<td>100 mm</td>
<td>61 mm</td>
</tr>
<tr>
<td>Pre-strain ratio</td>
<td>5×1</td>
<td>3(longitudinal)×6.5</td>
<td>2.3×3.5</td>
<td>1×1</td>
<td>1.15×1.15</td>
</tr>
</tbody>
</table>
### Table 3.1 Continued

<table>
<thead>
<tr>
<th>Pre-tensioner</th>
<th>Spring</th>
<th>Spring with telescopic structure</th>
<th>Spring Core-free</th>
<th>External carbon fiber reinforced composites shell</th>
</tr>
</thead>
<tbody>
<tr>
<td>Pre-tensioner stiffness</td>
<td>--</td>
<td>200 N/m</td>
<td>10.75 N/m</td>
<td>--</td>
</tr>
<tr>
<td>Roll stiffness</td>
<td>1250 N/m</td>
<td>1440 N/m</td>
<td>157 N/m</td>
<td>3052 N/m</td>
</tr>
<tr>
<td>Pre-tensioner weight</td>
<td>--</td>
<td>7.2 g</td>
<td>--</td>
<td>--</td>
</tr>
<tr>
<td>Total weight</td>
<td>9.6g</td>
<td>8 g</td>
<td>--</td>
<td>--</td>
</tr>
<tr>
<td>Maximum longitudinal strain</td>
<td>26%</td>
<td>31.25%</td>
<td>12.8%</td>
<td>6.5%</td>
</tr>
<tr>
<td>Maximum stoke</td>
<td>12mm</td>
<td>5mm</td>
<td>1.11mm</td>
<td>4mm</td>
</tr>
<tr>
<td>Maximum force output</td>
<td>15N</td>
<td>7.2N</td>
<td>--</td>
<td>7N</td>
</tr>
<tr>
<td>Electrical breakdown strength</td>
<td>109 MV/m</td>
<td>68.2 MV/m</td>
<td>36 MV/m</td>
<td>35-40 MV/m</td>
</tr>
<tr>
<td></td>
<td></td>
<td></td>
<td></td>
<td>33.5 MV/m</td>
</tr>
</tbody>
</table>
Spring and/or core-free roll actuators do have some critical limitations. The inner layers are subjected to the much higher compression due to hoop stresses in the outside layers, leading to uneven prestrain levels and thicknesses in layers (see Figure 3.30). This may cause early electrical breakdown of the inner layers [11]. Effects of compressive stress from outside layers on stroke and durability are reportedly negligible for total number of layers less than 35 [12]. Therefore, “thinner” D-EAPs spring roll actuators may be better than their larger counterpart. In an effort to address this issue, Hoffstadt et al. proposed a soft polymer core roll actuator [311]. The polymer core is soft and deforms as the pressure from outer layers of elastomer becomes large and thus the inner layers of elastomer can potentially avoid being under higher pressure. Needless to mention, the stiffness and poisson’s ratio of the polymer core is likely to affect the actuation performance of the actuator.
Figure 3.30 Theoretical distribution of mechanical compressive radial pressure on the layers in rolled D-EAPs actuators consists of N film layers [11].

Another problem with spring or core-free roll actuators is the difficulty in keeping the roll straight. Most often the free-standing actuator is bent, causing inaccurate displacement upon actuation (see Figure 3.31). In order to address this problem, telescopic guides have been used. Though effective, this telescopic shaft adds to device weight. Lastly, it is observed that spring rolls actually have some long-term hysteresis which can further affect the stroke magnitude and response accuracy [12].

Figure 3.31 Structural drawbacks of spring rolls actuator. (a) free-standing spring roll actuator shows obvious bent shape [12]; (b) spring roll actuator with telescopic core [13,118].
Fabrication rolled actuators is not simple, in particular, uniform pretensioning is a challenge. Danfoss A/S (company name) has automated the process for industrial roll-to-roll manufacture, see Figure 3.32. Additionally, a number of laboratory set up for efficient fabrication have been proposed [11].

![Progress diagram of automatic rolling up stage for assembling core-free D-EPAs roll actuators](image)

In conclusion, D-EAPs rolls are light-weight, compact, multifunctional, and mechanically robust with potential for a wide variety of applications. Many design principles of D-EAP based rolled actuators have been proposed and efforts continue toward a practical design.

### 3.2.9 Fiber-like Cylindrical Actuator

Cylindrical D-EAPs actuators were developed by SRI[114], and later modified and demonstrated by Ghosh [313], Carpi [314]and Cameron [315]. These class of actuators are particularly suitable for relatively compact and are particularly suitable for small force/displacement applications. Ghosh et al. proposed a fiber-like cylindrical actuator that is suitable for incorporating in textiles to fabricate smart and active products. As Figure 3.33 shows, a prestrained D-EAPs tube is filled with carbon black silicone composites or conductive...
silver grease as inner electrode and covered with the other carbon black silicone tube as outer electrode. Due to availability of the size of

In the proposed prototypes are fabricated from prestrained tubular silicone and polyurethane. D-EAPs with carbon black silicone composites or conductive silver grease as electrode. To control the radial and longitudinal prestrain a combination of inflation as well as axial tension was used. With optimized prestrain, this actuator is able to achieve up to 17% actuation strain in axial direction. Figure 3.33 demonstrates the typical morphologies of the actuator before and after action.

Figure 3.33 Fiber-like cylindrical actuator. (a) schematic of the fiber actuator; (b) axial actuation (about 10%) strain produced in uniaxially prestretched silicone based prototype [313].

Cameron et al. demonstrated a coextruded D-EAP fiber actuator made of thermoplastic polyurethane D-EAP with inner and outer electrodes made of graphite/liquid PDMS, and graphite respectively. The inner electrode and the polyurethane were simultaneously extruded out from a bi-component extrusion system, see Figure 3.34(a). The external electrode of graphite was applied in a post-painting process. The single fibers of diameter 1 mm produced maximum strain of 2%, same for assembled fiber in a rope form was about 0.3%.
Figure 3.34 Coextruded D-EAPs fiber actuators. (a) coextrusion mechanical schematic diagram; (b) coextruded fiber cross section; (c) an example of an assemble single fiber actuator; (d) an example of an assembled rope actuator with the ends capped with an epoxy plug. [315]

3.2.10 Cone Structure

In a basic cone shaped linear actuator, an annular-shape prestrained D-EAP film is constrained by a fixed frame along outer circumferential edge, and the inter circumference is attached to a moveable circular rigid part supported by a spring-loaded biasing mechanism. The biasing mechanism is in force equilibrium with the prestrained D-EAP. On application of an electrical stimulation, the D-EAPs film expands and the biasing mechanism deforms in order to achieve new load equilibrium. As a result, the top of the cone assembly moves upward in a linear
fashion. Cone shaped D-EAPs actuators have been examined in a wide variety of application, such as lens positioners, pumps, valves and tactile feedback actuators [316,317].

The biasing mechanism plays an important role in this actuator in terms of prestraining the film, and in guiding the movement in a preferential direction. Figure 3.36 illustrates effects of biasing mechanisms on displacement and force equilibrium of cone shaped actuators. The various biasing mechanisms results in correspondingly different equilibrium points and strokes. Understanding this relationship is crucial for designing effective cone actuator. In principle, the positive rate bias produces the least displacement and the negative rate bias provides the highest. The experimental results agree with this theoretical assumptions [319–322]. In comparison with positive rate and constant force biasing mechanisms, the bi-stable
negative rate bias provides the highest stroke. But this requires the specific level of prestrain in the D-EAP.

![Diagram showing passive and active stiffness curves with various bias mechanisms.][3]

Figure 3.36 Passive and active stiffness curves with various bias mechanism [316]

Figure 3.37 summarizes different designs of the biasing mechanisms. Among all of these designs, compression spring (Figure 3.37(a)), film (Figure 3.37(d)) and rigid strut (Figure 3.37(i)) have similar working principle. The compliant frame (Figure 3.37(e)) and parallel mechanisms provide positive rate bias and are considered similar. Dead weight (Figure 3.37(b)) as well as revolute joint (Figure 3.37(h)) are considered as constant force bias mechanisms. Diamond four-bar linkage mechanism (Figure 3.37(c)), half diamond double slider (Figure 3.37(f)) and bi-stable x-shaped post-pulled metal frame (Figure 3.37(j)) are examples of negative rate bias mechanisms.
Figure 3.37 Biasing mechanisms in cone shaped actuators. (a) spring [316,319]; (b) weight [316]; (c) diamond-shaped four-bar linkage mechanism [323]; (d) film [316,324]; (e) flexural pivot [318]; (f) double-slider [320]; (g) parallel mechanism [325]; (h) revolute [325]; (i) rigid strut [325]; (j) bi-stable negative rate mechanism [319].

Beside the biasing mechanisms, the dimensions of the device are also significant for actuation performances. Wang et al. investigated the importance of factors related to actuator performances by Taguchi method. These factors include D-EAPs film prestrain ratio, number of actuation units, inner and outside diameters. The results revealed that the outside diameter has the most significant effect on actuation, and by adding more actuation units the output force can be improved [322].
Among all of these designs, the film biased actuator has been commercialized. Artificial Muscle, Inc. (AMI) has commercialized Electroactive Polymer Artificial Muscle® (EPAM®) technologies. AMI has designed and tailored various EPAM® devices under the name SmartMOVE [316]. Universal Muscle Actuator® (UMA®) is one of their designs having double diaphragm configuration as shown in Figure 3.37(d) [324]. This device consists of two independent films attached to each other in the center and separated at the edges by spacer. In the free standing equilibrium, these two films are stretched by each other as biasing mechanism. As Figure 3.38 demonstrates, the UMA® film biased actuators have been explored to many applications on the basis of actuation force and stroke, such as haptic feedback and clutch actuator with high actuation force but smaller stroke, valve and pump with medium force and stroke, positioner with lower force but larger stroke.

![Load Response Plot](image)

Figure 3.38 Force and stroke operating profiles for various applications [316]
CHAPTER 4 Carbon Nanotube Sheet Electrodes for Anisotropic Actuation of Dielectric Elastomers

Abstract

The performance of dielectric electroactive polymer (D-EAP) based actuators depends critically on the electrode characteristics. Among the most challenging issues in the application of D-EAPs is the device-level complexity in producing sufficient directional actuation at acceptably low electric fields. In this work, a simple carbon nanotube (CNT) based electrode for D-EAP actuators is demonstrated that vastly improves directional strain response originating from the mechanical anisotropy of the electrode material. In this novel approach, highly aligned carbon nanotube (CNT) sheet electrodes are applied on acrylate adhesive films show high directed linear actuation strain of greater than 40% at a relatively low electric field (100 V μm⁻¹). The fiber-oriented CNT sheet applied around the D-EAP film, exhibits strong interaction between CNT fibers in the electrode and the D-EAP film to produce a robust conductive-nanolayer at the interface, on actuation cycling. The design paradigm provides a great potential for the fabrication of soft linear actuators.

4.1 Introduction

Conventional means of converting electrical energy to mechanical work are generally considered too noisy and bulky for many contemporary technologies. Electroactive polymers (EAP) constitute a class of macromolecules that undergo a dimensional change in response to an applied electric potential [326–332] and therefore offer a wide variety of alternative solutions to traditional means of electrical to mechanical energy transduction. As a growing
class of EAPs that shows great promise in biomimetic actuation, dielectric EAPs (D-EAP) [326,327,329,330] are considered well-suited for many applications including energy harvesters [331], and prosthetic devices [332]. D-EAPs are relatively inexpensive, lightweight, easily processable, and show large isotropic areal actuation strains (50-380%) with low to moderate stresses, large range of energy densities (0.01-3MJ m$^{-3}$), high electromechanical coupling efficiencies (~90%), over a broad range of electric fields (75-400 V $\mu$m$^{-1}$) [190,327,329,333]. D-EAPs are generally derived from chemically or physically cross-linked elastomers [331], and as a result they display low mechanical hysteresis [334]. Although D-EAP based actuators have been widely investigated, the most often used electrode materials have been percolative particle-polymer composites and metal films with a few notable exceptions that include efforts to develop transparent and self-repairing electrodes [335,336].

The real world application of D-EAPs has also been limited by a number of constraints; with the most important being the challenge of obtaining high directional electromechanical response at acceptable levels of electric field.

In the present study, we report a simple bifunctional carbon nanotube (CNT)-based electrode that serves both as an electronic conductor and a mechanically anisotropic constraint to produce directional actuation of the D-EAP. We also show that these CNT fibers penetrate the D-EAP on actuation cycling and thereby self-reconfigure towards a more stable nanolayer of composite structure on the D-EAP surface.
4.2 Dielectric Polymer Electrode: Background

The electromechanical response of D-EAP actuators is attributed to the electrostatic attraction between two electrodes in contact with the opposing surfaces of a D-EAP film that generates a compressive normal stress, $\sigma_z = \varepsilon_0 \varepsilon E^2$, along its thickness direction ($z$) [327]. Repulsive like charges that accumulate along both surfaces also act to stretch the film in the x-y plane, see Figure 4.1b. Here, $\varepsilon$ and $\varepsilon_0$ denote the relative dielectric constant of the elastomer and the permittivity of free space, respectively, and $E$ is the magnitude of the electric field.

Figure 4.1 Fabrication of actuators. (a) Drawing of CNT sheet from forest with SEM image of the sheet as inset. (b) Schematic illustration of VHB-CNT actuator fabrication (top), and the operational principle of the actuator before (middle) and after (bottom) activation, (c) Optical micrograph of the electrode area of an actuator.

The role of the electrodes in D-EAP actuation performance is of critical importance. Compliant
electrodes used in D-EAP actuators should maintain high electrical conductivity under large strain and allow unimpeded deformation of the D-EAP. Furthermore, the electrodes should be easily applicable and provide uniform charge distribution over the surface. Commonly used electrodes in D-EAP actuator fabrication include carbonaceous, metallic, or conducting polymer particles in sprayed-on powder form [226,330,337], or as electrically percolative particle-elastomer composites in cured and uncured forms [337,338]. Obvious problems associated with these approaches are increasing stiffness with higher levels of particle loading, in cured form, and instability in the uncured form [339]. Thermal evaporation or sputtering of thin metal films deposited on D-EAP surface produce electrodes of limited extensibility [226]. Although, patterning [340] as well as deposition of metals on prestrained elastomers to produce corrugated electrodes upon relaxation [226] have been shown to maintain conductivity at strains up to 80%, delamination remains a problem. Electrodes fabricated through low-energy gold ion implantation in elastomers survived strains up to 175% [335,341]. Lately, several authors have reported CNT based transparent compliant electrodes [336,342] including those with fault-tolerant capability through localized degradation of CNTs [336]. Silver nanowire based stretchable electrodes have been used to produce transparency, high conductivity as well as large strains (140%) [343].

Directional actuation, a key requirement in most applications of D-EAP actuators, is generally obtained through complicated design and mounting hardware [327]. A preferable route, however, is to design anisotropy in the D-EAP material and thereby minimize the overhead. Unidirectionally aligned nylon monofilaments embedded within silicone membranes as “flexible trusses,” has been reported to produce up to 35% linear strain [344]. D-EAPs
membranes (Acrylic, VHB4905) unidirectionally reinforced by carbon and nylon fibers in, have been used to produce 28 and 25% linear actuation strains, respectively, in the isostress direction [345]. Use of similar composite structure in cylindrical actuators resulted in 37% maximum actuation strain [346]. In each of these studies [344–346] the strain reported was measured at dielectric breakdown of the composites and under isotonic (constant load) conditions. Recently, we demonstrated effective use of unidirectional elastomeric fibers to yield composites with impressive electromechanical behavior including linear actuation strain of 55% at 90V μm⁻¹ [347].

Here, for the first time, we report a simple bifunctional CNT-based electrode that serves both as an electronic conductor and a mechanically anisotropic constraint to produce directional actuation. We also show that these CNT fibers penetrate the D-EAP on actuation cycling and thereby self-reconfigure toward a more stable nanolayer on the surface. We fabricated continuous sheets of entangled multiwall CNTs in a fibrillar network made up of mostly aligned bundles drawn out horizontally from vertically aligned CNT forests grown on quartz substrates, as shown in Figure 4.1a and c. The CNTs within the sheets were 25-40 nm in diameter and 1 mm long. In the as-drawn un-consolidated state the CNT sheets had a density of ~0.002 g cm⁻³ giving an average intertube spacing of 400 nm [348]. The CNT sheets were applied to uniformly prestrained (200%) acrylate adhesive films [329,334] (VHB-4905, 3M Corp.) to form actuators for electromechanical evaluation. This VHB line of commercial acrylic adhesives presently constitutes the often-investigated benchmark D-EAP.
4.3 Experimental

4.3.1 Sample Preparation

The D-EAP actuators were prepared by uniformly prestraining acrylate adhesive films (VHB-4905, 3M Corp., MN) to 200% linear strain and fixing it on a circular rigid frame. CNT sheets of appropriate width were drawn from the as-grown forests and applied on both sides of the prestrained elastomer films using a stencil mask (made of polyethylene film) with a circular active area of 10 mm in diameter to create the well-known circular test samples [327–330,333,334,347]. The CNT sheets were easily transferred to the sticky surfaces of the elastomer by gently pressing onto the sheet through circular masks. Another set of similar actuator samples were prepared using uniformly compliant carbon grease electrodes (Chemtronics Circuit Works, Kennesaw, GA) for comparative purposes.

4.3.2 Actuator Characterization

The uniaxial tensile tests were performed on a MTS-30G load frame operated at a crosshead speed of 4.4 mm min\(^{-1}\) (sample configuration is shown in Figure A1 of APPENDIX). Low magnification morphological features of the actuator specimens were investigated using an optical microscope (Nikon, Eclipse 50i). Higher magnification images were collected on a field emission scanning electron microscope (JEOL 6400F), and a transmission electron microscope (HITACHI HF2000) using an accelerating voltage of 200 kV. Images of specimens under various levels of strain were obtained using the optical microscope, immediately after application of strain on a specially designed extension stage.
Electrical properties of the electrodes on the VHB-CNT actuators, in both x- and y-directions were measured in steps of 10% uniaxial strain up to a maximum of 70%, using a customized four-point probe connected to a constant current source (model 6221, Keithley, Cleveland, OH) and a voltmeter (model 2182A, Keithley, Cleveland, OH). The samples were held under strain using a specially designed extension stage, see Figure A2 in APPENDIX.

Field-induced actuation response characteristics of the VHB-CNT test samples were evaluated by connecting the actuator electrodes to a Bertan 225-30R high-voltage power supply (Spellman High Voltage Electronics Corp., Hauppauge, NY), with a computer controlled automated voltage trigger. While the voltage was increased, real-time in-plane deformation of the active area was digitally recorded using a video camera. The resulting images were analyzed frame-by-frame with image analysis software (ImageJ, NIH) to calculate the actuation strains in different directions (see Figure A3 in APPENDIX). In the actuation cycling tests, the electric field was ramped up to the required voltage in 10s with no noticeable lag between increase in voltage and specimen actuation.

4.4 Actuator Performance

4.4.1 Mechanical Behavior

To explore the electrode induced mechanical anisotropy of the VHB-CNT actuators as well as their ability to withstand cyclic deformation, we performed multi-cycle uniaxial tensile tests along the fiber (y) and cross-fiber (x) directions of the actuator samples, see Figure 4.1b. The VHB-CNT actuator specimens were cycled 25 times between 0 and 25% strain in y- and x-directions respectively and the results are shown in Figure 4.2a and b. In both cases, the stress-
strain response becomes almost independent of cycle number after about 10 cycles; therefore, to facilitate visualization of the data, only the 1st, 2nd and 25th loading-unloading plots are shown. The highly lossy and distinctive stress-strain behavior of the VHB-CNT laminate in the y-direction (Figure 4.2a), during the first cycle, is dominated by the complex inter-fiber interactions within the CNT sheet. The initial low modulus at very low strains resulting from sliding and straightening of the CNT fibers along the loading direction quickly increases to about 28.0 MPa as the aligned CNT fibers contribute to the load before significant lowering of modulus is noted due to failure of CNT fibers beginning at ca. 4% strain. The failure strain measured here is consistent with that reported in the literature for CNT sheets [349] as well as pure and PVA infiltrated highly twisted yarns of multiwall CNTs [350,351]. The subsequent lower modulus at strains greater than ~ 4% (2 MPa) is closer to that of the neat VHB film. In the following cycles, the modulus in the y-direction for the large part remains relatively low (2.6 MPa) and shows limited hysteresis, as evidenced by a small shift in the data at zero stress to a higher unrecoverable strain. Curiously, a significant increase in the y-direction modulus after the 1st cycle is noted near the end of the strain cycle far beyond the initiation of apparent fiber failure at ca 4% strain. The same behavior, akin to strain hardening, was observed in VHB-CNT laminates at additional strain amplitudes of 6 and 40%, see Figure 4.2c. In all cases, the resistance to deformation at high strains is due to the significant number of CNT fibers that seem to have survived the first and subsequent loading cycles through relative sliding, plastic deformation, and consolidation. During recovery, these filaments most likely buckle and manifest itself in the next cycle of loading. We will discuss more on the morphological changes around the strain level corresponding to CNT sheet failure strain later. The stress-strain
behavior of the actuator assembly in the x-direction, seen in Figure 4.2b, is unremarkable and the modulus (2.3MPa) is slightly higher than that of the neat VHB film.

Figure 4.2 Cyclic tensile stress-strain curves of VHB-CNT actuators obtained by loading and unloading to 25% strain amplitude, (a) along the fiber (y) direction, (b) along the cross (x) direction. For clarity, data for 1st (●), 2nd (▲), and 25th (■) cycles are shown. (c) y-direction stress-strain diagram of the 25th cycle for 6 (●), 25 (▲), and 40% (■) strain amplitudes. The solid lines connect the data in all cases.

4.4.2 Morphological Characteristics

The change in morphology of VHB-CNT actuators upon extension cycling is presented in Figure. Low magnification optical microscopy images of the CNT sheet electrodes at various strain levels in the y-direction reveal lines of fracture, visible at 10% strain. Close-up view of the fracture boundaries shows evidence of bridging CNT fibers across the fracture (see inset of Figure 4.3a). On recovery, the CNT fibers form distinct kink bands in the direction normal to the applied strain, resulting from the elastic recovery of the elastomer and the buckling of the CNT fibers on its surface (Figure 4.3b). We also note that the highly drawn-out surviving CNT fibers (and broken fiber ends), near the fracture, that have undergone inter-tube sliding and plastic deformation [350] form distinct clusters of more tightly folded undulations and kink
bands, see Figure 4.3c. In general, the CNT sheets on the y-direction strained samples show parallel bundles of oriented fibers as shown in the inset of Figure 4.3c. The result of strain cycling in the x-direction produced somewhat similar changes in the electrode morphology, without any evidence of fiber failure (Figure 4.3d). However, at the micro-level, the CNT fibers that appear to have gathered together in bundles show a more open fiber network with no orientation.

Figure 4.3 Optical images of electrodes showing morphological changes of CNT sheet electrodes, (a) sample under 10% tensile strain in the y-direction showing fracture, the inset shows bridging fibers across lines of fracture, (b) sample after 25 cycles of 25% strain along the y-direction. Optical and SEM images (as insets) of, (c) CNT fiber clusters of tightly folded undulations and kink bands (circled area) in strain-cycled sample as in (b), (d) sample after 25 strain cycles of 25% strain in the x-direction.
4.4.3 Electrical Behavior of Electrodes

The electrical resistance of the CNT electrodes applied on the actuator samples measured as a function of uniaxial strain along both x- and y-directions shows a number of important features, see Figure 4.4. First, at zero strain, the resistance along the fiber direction (y) is significantly lower than that in the cross direction (x), indicating electrical anisotropy ratio of ca. 4 of the CNT sheet, which is lower than previously reported [352]. The change in resistance in the y-direction, as a function of applied strain in the y-direction (Figure 4.4a), is initially slower up to about 30% strain, beyond that the significant loss of conducting pathways through the CNT sheet is evident. Beyond the strain level associated with fiber failure, noted earlier at about 4%, the electrodes remain conducting, albeit at a lower level, due to the spanning fibers (the inset of Figure 4.3a) across the fracture lines in the CNT sheet. Beyond 70% strain in the y-direction the electrodes become almost insulating. In case of x-direction strain the change in electrical resistance in the x-direction is gradual and more modest, see Figure 4.4b. It is important to note that in potential practical application the VHB-CNT actuators the y-direction strain is likely to be severely limited compared to that in the x-direction due to the mechanical anisotropy discussed earlier.
Figure 4.4 Electrical resistance of the CNT sheet electrodes measured along y (■) and x (●) directions, plotted as a function of applied strain in the, (a) y-direction, and (b) x-direction. The solid lines connect the data, and the error bars represent one standard deviation around the mean.

4.4.4 Electromechanical Properties

Linear actuation strains of the VHB-CNT actuators, $S_x$ and $S_y$, in the cross-fiber (x) and fiber (y) directions, respectively, are presented as functions of nominal electric field in Figure 4.5a and reveal two important observations: $S_x$ for the VHB-CNT system is consistently greater than $S_y$, and the uniform linear strain produced by the same D-EAP, with carbon grease electrodes, at all electric fields. To put the actuation anisotropy in context of the electrode induced mechanical anisotropy, tensile moduli measured along the x- and y- directions ($E_x$ and $E_y$, respectively) are plotted as a function of cycle number as an inset in Figure 4.5a. Considering that the actuation strain in the y-direction is generally below the failure strain of the CNT sheet (microscopic evidence to be discussed later), the mechanical anisotropy of the actuator can be expressed as, $E_y(@strain<0.04)/E_x$. The resulting mechanical anisotropy of
about 10 compares well with the measured actuation anisotropy (defined as the strain ratio $S_x/S_y$) values of about 7.5.

Figure 4.5 Linear actuation strain plotted, (a) as a function of nominal electric field in the fiber (○) and cross-fiber (●) directions together with directionally uniform response of a similar actuator with carbon-grease electrode (■), (b) as a function of actuation cycle number for actuators at 50 and 80 V \( \mu \text{m}^{-1} \) electric fields as labeled. The inset of (a) shows the variation or tensile modulus of the VHB-CNT samples as a function of cycle number. The solid lines represent exponential fits to the data in (a) and the same in (b) is used to connect the data. Video capture images illustrate a test specimen, (c) before actuation, (d) at 80 V \( \mu \text{m}^{-1} \), and (e) at 100 V \( \mu \text{m}^{-1} \).

While the results presented in Figure 4.5a correspond to single cycle, we hasten to point out that actuation cycling up to 370 cycles shows hysteresis levels comparable to that reported earlier for VHB [353] and fairly consistent actuation strain as shown in Figure 4.6. Actuation cycling data for 25 cycles at 50 and 80 V \( \mu \text{m}^{-1} \) (about 40 and 70% of the breakdown electric
fields, respectively) is shown in Figure 4.5b. These electrical field magnitudes are considered representative of typical potential D-EAPs applications. The actuation strain increased after the first cycle and remained at approximately the same level, strongly suggesting no significant change in the electrode characteristics during actuation. A sequence of digital images acquired from the active area of the VHB-CNT actuator upon exposure to electric fields varying in strength is shown in Figure 4.5c-e illustrate the anisotropic deformation. Interestingly, the high electric fields near the bare nanometer-scale CNT tips at the boundary of the electrodes resulted in an inter-electrode “corona” discharge. The phenomenon forms basis of CNT based gas sensors suggested in the literature [354]. In our experiments, we used a thin layer of sprayed silicone oil to alleviate the problem. Needless to say that insulation of CNT electrodes will be necessary (e.g., multilayer stacked or cylindrical actuators) in practical use of CNT sheets in similar applications.

![Graph](image.png)

**Figure 4.6** Linear actuation strain in the cross direction (x) for the initial 220 cycles of the 370 cycles.

To assess the effects of actuation cycling on the CNT sheet electrodes, we also examined their morphology after 25 mechanical or actuation cycles. Low magnification optical microscopy
images of actuation cycled samples presented in Figure 4.7a revealed undulations and kinks, as noted earlier (Figure 4.3d), and no signs of CNT fiber failure. On further examination of the electrodes using scanning and transmission electron microscopy, we noticed superficial but significant penetration of the CNT fibers (Figure 4.7c and d) in the VHB film surface. To confirm and explore the level of penetration, we decided to clean the electroded areas of the actuators using silicone-oil soaked cotton swabs with 3-4 uniform and gentle strokes along the fiber direction with enough pressure to remove the CNT fibers from surface and measure the electrical resistance of the cleaned area. The data presented in Figure 4.7 confirm our morphological observation of significant CNT penetration in the VHB surface. The increase in electrical resistance of actuation cycled samples after cleaning is significantly lower than that of the similarly cleaned mechanically cycled samples. The CNT fiber penetration into the soft VHB film, may be due to the compressive electrostatic stress brought on by the applied electric field. Alternatively, this may be the result of the out of plane buckling of CNT fibers during recovery from repeated omnidirectional strain cycling in actuation. The penetration leads to a self-reconfigured composite layer that is stable after a number of actuation cycles and may result in longer lifetime and reliable performance of the actuators.
Figure 4.7 Self-reconfiguration of the CNT electrodes. The bar chart shows change in electrical resistance of CNT electrodes, along fiber (y) and cross-fiber directions, measured at various stages (as labelled) using a four-point probe set up. Insets (a), (b), and (e) are optical micrographs of CNT electrodes after 25 cycles of actuation, after 25 cycles of actuation and cleaning, and after 25 cycles of mechanical cycling at 25% strain followed by cleaning, respectively. Insets (c) and (d) are SEM (surface) and TEM (cross-sectional) images of CNT electrodes, respectively, showing penetration of CNT fibers in the elastomer after 25 cycles of actuation.

4.5 Conclusion

In conclusion, the electromechanical properties of the VHB-CNT actuators establish that the highly oriented CNT sheets provide an attractive alternative to conventional electrodes for D-EAPs applications. The synergistic property development results in high directional actuation strain. The strategy of exploiting anisotropic mechanical behavior of electrodes proposed for
the first time, represents a new pathway in the fabrication of effective and practical D-EAPs based devices requiring directional actuation and impedance matching. We have shown evidence of self-reconfiguration of CNT fibers along the surface of the elastomer actuators. Taken together, these results demonstrate that directional actuation strain of D-EAP actuators can be greatly and conveniently enhanced through electrode-induced anisotropy. Optimization of electrical and mechanical properties of the CNT sheet is expected to improve the actuation strain, and other performance.
CHAPTER 5 Enhanced Anisotropic Response of Dielectric Elastomer Actuators with Microcombed and Etched Carbon Nanotube Sheet Electrodes

Abstract

Dielectric electroactive polymers (D-EAP) offer tremendous potential in a wide-ranging application including microrobotics and wearable responsive systems. The real-world application of D-EAPs, however, has been limited by a number of factors, including facile means of producing directional stress/strain. As a critical component of the D-EAP actuator, the electrodes should have high electrical conductance under finite in-plane deformation, good electromechanical stability, and ease of shaping based on the design requirements. In this work, we investigate highly aligned carbon nanotube (CNT) sheets as electrodes in D-EAP actuators to yield anisotropic electromechanical response. The morphology of CNT sheets was altered by microcombing and selective laser etching to enhance mechanical anisotropy. The enhancement of CNT sheets alignment results in almost pure unidirectional strain of 33% at a relatively moderate electric field. The results demonstrate that the deformation anisotropy of D-EAP actuators can be significantly improved by directional laser etching of the electrodes rather than microcombing alone.

5.1 Introduction

Commonly used actuators, such as electromagnetic, pneumatic, and hydraulic types are bulky, heavy, noisy, and particularly not suitable for many of today’s applications such as microrobotics, and mobile devices that often require high power/torque to mass ratio, direct-drive, conformability, and small form factor. Materials based actuation technologies, such as
piezoelectric ceramics [355,356], shape memory alloys [357], carbon nanotubes[104] and
electroactive polymers (EAP) [2,32,190,358–361], seem to offer significantly better
alternatives. EAPs are lightweight, soft, and inexpensive, while they exhibit shape change
when subjected to an electric field. Unlike their inorganic counterparts (piezoelectric ceramics
and shape memory alloys, etc.), they are easy to process, shape, and tune to offer a broad range
of mechanical and electrical properties [8,14].

Dielectric electroactive polymers (D-EAP) constitute a class of electronic EAPs with great
potential [327]. Their unique advantages include large isotropic areal actuation strains (50%-
380%) with low to moderate stresses, large range of energy densities (0.01-3MJ m⁻³), and high
electromechanical coupling efficiencies (~90%) over a broad range of electric fields (75-400
V μm⁻¹) [14,190,327,333]. In a typical actuator configuration, a layer of soft D-EAP film is
sandwiched between two layers of compliant electrodes, see Figure 5.1(a). On application of
an electric potential across the electrodes, the attractive force between opposite charges on the
electrodes and the repulsive forces between the like charges in the same electrode together
result in in-plane expansive deformation of the dielectric elastomers. This electromechanical
response described as Maxwell stress (σ_m) is a function of the dielectric constant (ε), the
permittivity of the free space (ε₀), and the applied electric field (E) [327]:

\[ \sigma_m = \varepsilon_0 \varepsilon E^2. \]

D-EAPs consist of physically or chemically cross-linked macromolecular networks [14] and
are almost mechanically isotopic. Under an applied electric field, the in-plane expansions are
uniform in every direction. Therefore, in some actuator applications that require directional
electromechanical response [15], it is necessary to use other complex means to direct the
stress/strain in the preferred direction. Attempts to impart directional deformation of D-EAP actuators include use of rigid links, such as diamond-, bowtie-shaped rigid frames [8,260,257], and embedded aligned fibers to serve as constraints [307,362,363]. For example, Lu and coworkers employed carbon and nylon fibers to unidirectionally reinforce acrylic elastomers and observed up to 28% actuation strain solely in cross-fiber direction [307]. Also the same approach was used in cylindrical shaped actuators, with aligned nylon fiber reinforcement in hoop direction, axial strain was enhanced by ~10% [362]. Recently, we demonstrated effective use of unidirectional elastomeric fibers to yield composites with improved dielectric constant and impressive anisotropic electromechanical behavior including linear actuation strain of 55% at 90V μm⁻¹ with actuation strain anisotropy of ~2.3 [347]. The strain-dependence of elastic modulus of elastomers has been utilized to achieve anisotropic actuation of D-EAPs by application of nonuniform directional prestrain [4]. In addition, Benslimane et al. used a corrugated D-EAP film to achieve anisotropic actuation [227]. More recently, we reported a bifunctional CNT-based electrode that serves both as an electronic conductor and a mechanically anisotropic constraint to produce directional actuation [364]. The aligned CNT-film electrode applied on acrylic actuators produced directed linear actuation strain of up to 40% at a relatively low electric field (100 V μm⁻¹) with high mechanical anisotropy of 10 and actuation strain anisotropy of 7.5.

Ideal electrodes in D-EAP applications should be able to deform simultaneously with the elastomer to large strains without restraint, while maintaining high electrical conductivity. Often used compliant electrode materials are either cured or uncured percolative particle-polymer composites containing carbonaceous [222,223] or metallic particles [313].
Carbonaceous soft composites are not easy to apply and usually smudges as well as tends to dry out resulting in short working life in uncured or “wet” state. Additionally, these elastomeric composites when heavily loaded with fillers exhibit undesirable stiffness [221]. Graphite powders/sprays generally require additional adhesive binder that may stiffen the elastomers [220]. Rigid metal electrodes of any kind is likely to constrain the deformation of actuators upon actuation, and may crack under a relatively low strain [221]. Different from these materials, aligned continuous CNT sheet electrodes, are easy to apply and show great promise in tailoring the overall performance of the of D-EAP actuators [364].

CNT-based patterned thin film electrodes have been fabricated by growing CNT arrays on patterned catalyst or substrates using electron beam/ photo- lithography [365,366], photoresist [367], or printing techniques [368]. Shin and co-workers have reported contact transfer patterning that enables removal of CNT arrays via a stamp [369]. Inkjet printing has also been utilized to apply dispersed CNTs following a specific pattern [370,371]. Compared to these methods, laser ablation is a low-cost chemical-free method that is relatively easy to operate with high accuracy [372]. Laser techniques have been employed to directly ablate CNT arrays into a certain pattern [373,374] and to etch dispersed CNT particle thin films on the surface of substrates. For example, Lin and coworkers etched CNT film on the surface of polyethylene terephthalate film. Etching depth was in range of 15-60 nm depending on the wavelength of light employed, and the minimum width of the etched stripe was as small as 20 µm [375].

In-plane mechanical behavior of the CNT sheet electrode and the level of anisotropy could be important issues in the design of D-EAP actuators. Significant increase in Young’s modulus has been reported due to improved alignment in continuous CNT sheets through stress-pressing
[376], and stretch-dipping [377] methods. Zhang et al. reported a “microcombing” process [378] to enhance alignment in CNT sheets resulting in increase in elastic modulus (up to 172 GPa increased by 14%). In this method, CNT sheets were combed by passing the sheet over the edge of two surgical blades placed at an angle (80-85°) to the sheet-plane.

In this work, we explore the effect of alignment of CNT sheet through microcombing for application in D-EAP actuators to generate anisotropic electromechanical response. Also we report the effect of selective laser ablation of combed CNT electrodes to further enhance the anisotropic actuation performance, see Figure 5.1(b). The results demonstrate that the deformation anisotropy of D-EAP actuators can be significantly improved by directional laser etching of the electrodes rather than microcombing alone.
Figure 5.1 Schematics illustration of (a) operational principle of D-EAP actuators, (b) microcombing (on the left) and laser ablation (on the right) processes. The laser ablation process is used to selectively remove CNT fibers along 360 μm wide lines from the CNT sheet on the surface of VHB actuator, leaving stripes of 180 μm width.
5.2 Experimental

5.2.1 CNT Synthesis and Preparation
Sheets of mostly aligned continuous CNT network were drawn from vertically aligned multiwall carbon nanotube forests synthesized on quartz substrates in a tube furnace. The individual CNTs have diameters ranging from 30 to 50nm. The modified version of the chlorine mediated chemical vapor deposition route was used as described in detail elsewhere [379]. These CNT sheets were subsequently combed, and/or etched.

5.2.2 Microcombing Process
The microcombing process was performed, following Zhang et al. [378], by passing a layer of as-drawn CNT sheet over the sharp edge of one utility knife-blade (titanium, 3M Corp., MN) with equal contact angles of 75° while winding on a rotating mandrel at a slow speed of 0.96 m/min, see Figure 5.1(b). The method, reported earlier, demonstrably improves the fiber orientation in the CNT web.

5.2.3 Actuator Preparation
VHB-CNT actuators were prepared by uniformly stretching the acrylate adhesive films (VHB-4905, 3M Corp., MN) to 200% in-plane strain and fixing the stretched film on an annular rigid frame (inner dia. 45mm). Appropriately combed circular CNT sheet electrodes (dia. 10 mm) were applied in the center of the D-EAP film on both sides, see Figure 5.1(b). The CNT electrodes in opposite sides were positioned so as to completely overlap and have the same fiber orientation before applying to the tacky VHB film using light pressure and a stencil mask. Subsequently, leads were drawn on the D-EAP surface using carbon grease (Chemtronics
Circuit Works, Kennesaw, GA) in order to connect the electrodes to a computer controlled DC power supply synced with a video capture system. For comparison purpose, D-EAP actuators with as-drawn CNT sheet electrodes and carbon grease were also prepared and evaluated in the same manner.

5.2.4 Laser Ablation Process

In order to further enhance the mechanical anisotropy of aligned (already through microcombing) CNT sheets, CNT fibers were selectively removed from the D-EAP actuator surfaces, along lines parallel to fiber’s orientation, using an EPILOG Mini 8000 Laser System. The etched line width and separation, decided on the basis of optimal uniformity and resolution, were 360µm and 180µm, resulting in a total etched area of 67%. The depth of ablation was optimized by controlling the working power and linear speed of the laser engraver while avoiding severe damage to the D-EAP film.

5.2.5 Actuator Characterization

The morphological features of the CNT electrodes were determined using an optical microscope (Nikon, Eclipse 50i) and a field emission scanning electron microscope (Phenom-World B.V., Phenom-G1). Uniaxial cyclic tensile behavior of the fully built actuators along both fiber (y) and cross-fiber (x) directions were determined using a MTS-30G load frame (fitted with a 10N load cell) set to apply 40% strain for 25 cycles at a constant crosshead speed of 4.4mm min\(^{-1}\) with gauge length of 15mm. Electrical properties of the electrodes in fiber and cross-fiber directions were measured using a customized four-probe set-up connected to a
current resource (model 6221, Keithley, Cleveland, OH) and nanovoltmeter (model 2182A, Keithley, Cleveland, OH).

Electromechanical response was examined by a customized set-up consisting of a Bertan 225-30R high-voltage power supply (Spellman High Voltage Electronics Corp., Hauppauge, NY), a digital video camera and a computerized voltage-trigger. While the actuators were activated by an increasing voltage up to 5 kV in steps of 1 kV, the active area was recorded using the video camera in real-time to monitor the in-plane deformation. The acquired video images were analyzed frame by frame using image analysis software (ImageJ, NIH), to determine the actuation strain distribution in the sample, see Figure A3 in APPENDIX.

5.3 Result and Discussion

5.3.1 Morphological Characteristics

To explore the morphology of CNT sheet electrodes before and after microcombing and/or laser ablation, optical microscopic and SEM images were obtained for all the CNT films attached on the surface of VHB films. Although, optical microscopic images of as-drawn (Figure 5.2(a)) CNT sheets show irregular, entangled, and wavy bundles of CNT, after combing (Figure 5.2(b)), the wavy CNT bundles appear to be straightened and more aligned. However, some of the CNT fibers seem to have come together to form larger bundles and most of the apparent defects are mitigated. The microcombing process seems to have also straightened some of the spatial crimps present in the as-drawn samples. This is considered a desirable outcome since the nonuniform CNT bundles in film thickness direction can easily penetrate into the soft elastomer and thus induce electrical shorting of the actuator [364]. SEM
images of as-drawn (Figure 5.2(d)) and combed CNT electrodes (Figure 5.2(e)) confirm the earlier observations of more order in combed electrodes.

Microscopy of etched CNT sheets (refers to combed and subsequently etched CNT sheets) show that the etching process was effective in removing most of the CNTs in striped pattern (Figure 5.2(c)) and leaving relatively clear areas with many residual short CNT bundles (Figure 5.2(f)). These CNT fibers are likely to provide conducting paths throughout the electrode that is crucial for the functioning of the actuators.

Figure 5.2 Optical and scanning electron micrographs of CNT sheet electrodes. (a), (b), and (c) are optical microscopic images of as-drawn, combed, and etched samples, respectively. (f) is optical microscopic with higher magnification of etched area in the etched sample. (d) and (e) are the SEM images of as-drawn, combed samples, respectively.
5.3.2 Tensile Behavior

The cyclic stress-strain behavior of the actuators, under uniaxial tensile loading, were analyzed to investigate their mechanical anisotropy as well as their potential deterioration due to mechanical cycling. All VHB-CNT actuators show significant hysteresis during the 1st cycle (Figure 5.3(a)) along the fiber direction (y). At very small strain levels (< 1.5%) the crimps and curls in the CNT fibers manifest in relatively low moduli ($E_y$) (as-drawn: 2.7 MPa, combed: 2.9 MPa). However, both as-drawn and combed samples show rapid rise of modulus in the fiber direction beginning at about 2% strain. The increase is much higher for the combed sample (up to 29.1MPa) compared to that of the as-drawn (18.9 MPa). The high resistance to deformation of these actuator samples remains until the CNT fibers begin to fracture at about 8% strain for the combed and 12% strain for the as-drawn specimens. The relatively higher moduli as well as early onset of fiber fracture in case of the combed sample can be attributed to the straightening and improved alignment of the CNT fibers due to the microcombing process. The CNT fiber fracture strain (8-12%) reported here, albeit little higher, is well within the large variation in the synthesis of CNT forests and consistent with that reported earlier for CNT sheets [364]. Following the fiber fracture, the tensile moduli in both cases (combed and as-drawn) remain constant and similar to that of neat CNT at high strains, reflecting the “turtle shell-like” morphology with irregular shaped islands of harder pieces of VHB-CNT laminates connected by the softer VHB films, see microscopy inset of Figure 5.3(a). With increasing strain, the deformation is largely localized in the softer VHB films resulting in a low modulus. In contrast, the etched samples continue to show a much lower modulus of 2.9 MPa (this is calculated from overall specimen width and the value is 8.8 MPa by using the un-etched width)
during the first cycle, about 45% higher than that of the pristine VHB, throughout the applied strain regime of 40%. The lower modulus of etched samples is most likely due to the introduction of discontinuities in CNT fibers aligned close to the fiber direction (y) due to etching.

The tensile behavior of all of the actuator samples, along the fiber direction, beginning with the 2nd cycle show small but significant difference, see Figure 5.3(b). The elastic moduli for all samples at low strains (<15%) are the same but slightly higher for the combed sample. In case of as-drawn and combed samples, significant increase in the resistance to deformation near the end of the strain cycle is noteworthy. This behavior was reported earlier in our studies [364] and is due to the buckling and limited entanglement of the surviving fibers (from the 1st cycle) during recovery from the previous cycle. This phenomenon seems to be more pronounced in the combed sample, suggesting the effect of straightening and bundling of CNT fibers due to combing, noted earlier in the morphological results. Interestingly, the etched samples did not show any such hardening behavior. It is more likely that the removal some of the CNT fibers interrupted the integrity of the entire network in terms of continuity of fibers and entanglements, resulting in lower load transfer between CNT fibers and thereby buckling during recovery. After the 2nd cycle, the mechanical behavior in all case remained mostly unaltered.

Unidirectional tensile behavior along the cross-fiber direction (x) for all the samples during the 25 cycles were remarkably similar and comparable to that of pristine VHB films. For clarity, the data for only the 1st cycle along is included, see Figure 5.3(c). The modulus values along
the cross-fiber direction ($E_x$) of the as-drawn, combed and etched samples were 2.5 MPa, 2.4 MPa and 2.3 MPa, respectively.

The effect of combing and etching is readily manifested in the mechanical anisotropy of the VHB-CNT actuators. Considering that the actuation strain (discussed later) in the fiber ($y$)-direction is generally below the fracture strain of the CNT fibers (~10%), the mechanical anisotropy of the actuator is defined as, $E_y(@\text{strain} < \text{fiber fracture strain}) / E_x$. The as-drawn, combed and etched CNT electrodes produced mechanical anisotropy of 7.9, 11.6 and 3.8, respectively, showing that combing process improves the alignment within the CNT sheet. In case of etched samples, the lower anisotropy is due to the significantly lower fiber direction modulus discussed earlier.
Figure 5.3 Stress-strain curves of cyclic uniaxial tensile test of actuators obtained under 40% strain amplitude. For clarity, the data for only the 1st cycle (a) along fiber (y), 2nd cycle (b) along y-directions, and 1st cycle (c) along cross-fiber (x) direction of as-drawn (●), combed (▲) as well as etched sample (■), are shown. Additionally, the 1st cycle stress-strain plot (- - -) of the pristine VHB film is shown in (a). Optical micrograph of the as-drawn sample under 20% tensile strain in y-direction shows the “turtle shell-like” morphology.

5.3.3 Electrical Behavior

Electrical resistance of all electrode types measured along the fiber (y) and cross-fiber (x) directions shows the lowest resistance for as-drawn samples, and in all cases fiber direction resistance is lower than that of cross-fiber direction, see Figure 5.4(a). To characterize the effect of etching on the electrode electrical resistance a completely etched combed sample was evaluated. The microcombing and etching processes, were found to increase the electrical
resistance in both directions, with greater degree of increase in completely etched samples. Electrical anisotropy, defined as the ratio between fiber and cross-fiber direction resistance values, for all types of electrodes remains in the range of 3.4-3.8. Taken together these observations point to the significant role of the fiber alignment in contributing to the conductive pathways and thereby determining the overall electrical behavior of the electrodes. Also important to note that the improved alignment of the CNT fibers due to the microcombing process results in marginal increase in electrical anisotropy to 3.86.

The electrical resistance value of combed CNT sheets in both directions reported in this study is slightly higher than those of as-drawn CNT sheets and is contrary to what has been reported by Zhang et al. [378] in a recent study. The difference is likely due to the measurement techniques utilized. In our four-probe set-up the probes measuring 560 µm in diameter are separated by equal intervals of 3 mm. Within this small measurement region, a single layer of combed CNT sheet containing bundles (see Figure 5.2(b)) is likely to have fewer electrical pathways along the cross-fiber direction due to fewer spanning CNT fibers. In contrast, the resistivity data reported by Zhang et al. [378] was collected using 200 layers of combed aligned CNT sheets [378]. The layered assembly of CNT sheets are likely to contain more bridging CNT fibers between bundles both within and between layers through the thickness.

The effect of laser ablation on the electrical conduction of the electrodes is also worthy of discussion. After complete etching, the electrical resistance in both fiber and cross-fiber directions are higher. It is likely that the relatively rigid CNT bundles have penetrated sufficiently into the soft VHB film surface during sample preparation and survived the laser
ablation. In either case, the data provides evidence of potential to retain electrical conductivity of CNT electrodes through optimum use of laser power, treatment time, and pitch of laser-head movement, while tailoring their mechanical properties as well as the optical transparency (see Figure 5.4(b) and (c)).

![Figure 5.4](image)

Figure 5.4 (a) Electrical resistance of the as-drawn, combed and completely etched CNT sheet electrodes, measured along the fiber (y) and cross-fiber (x) directions. Images of (b) as-drawn, and (c) completely etched CNT sheet electrodes mounted on a printed paper to demonstrate optical transparency.

### 5.3.4 Field-induced Deformation

The CNT sheet electrodes are expected to act as mechanical constraints and thereby produce anisotropic electromechanical response. Accordingly, the measured actuation strain values in the cross-fiber direction (x) is significantly larger than that in fiber direction (y) for all types of CNT sheets electrodes, see Figure 5.5(a) and (b).
Actuators with as-drawn CNT sheet electrodes produced highly anisotropic strain distribution with 6% and 26% actuation strains in the fiber and cross-fiber directions, respectively, at an electric field of 110 V µm⁻¹. With combed CNT electrodes, at the same electric field the actuation strain in the fiber direction was insignificant while the same in the cross-fiber direction was 24%. This implies that the enhanced alignment of combed CNT sheets results in the higher mechanical constraint in fiber direction and an almost pure unidirectional actuation deformation. Actuators with directionally etched CNT electrodes produced strains of 0.3% and 33% in the fiber and cross-fiber directions, respectively, at 110 V µm⁻¹. The considerably larger deformation in the cross-fiber direction for etched electrodes compared to those with as-drawn and combed CNT electrodes reflects the altered mechanical anisotropy due to etching. After etching, the non-etched areas were sufficient to constrain the deformation along the fiber direction, while in the cross-fiber direction the modulus reduced significantly. Also, with help of enough residual CNT fibers in the etched areas to provide conductive pathways in the etched electrodes, consequently, the actuation strain in cross-fiber direction was significantly greater. The video-capture images in Figure 5(c-h) present clear visual evidence of the effects of microcombing and etching of CNT sheet electrodes.
Figure 5.5 (a) Linear actuation strain plotted as a function of electric field of actuators with as-drawn (●), combed (▲) as well as etched (■) CNT sheet electrodes. For comparison, actuator with carbon grease (♦) electrodes is included. (b) Magnified view of (a) in electrical range of 80 V µm$^{-1}$ to 120 V µm$^{-1}$. Video capture images of actuators before actuation with (c) as-drawn, (d) combed, (e) etched CNT electrode, and actuators at 110 V µm$^{-1}$ with (f) as-drawn, (g) combed, (h) etched CNT electrodes.

It is important to note that all electric field values mentioned here represent ratio of applied electric potential and true instantaneous thickens of the D-EAP films calculated from the measured areal strain under isochoric deformation.
To summarize, the mechanical, electrical, and actuation anisotropy values of the actuators with various CNT sheet electrodes evaluated in this study and previously reported in the literature are presented in Table 5.1. Overall, the actuators with aligned CNT sheet electrodes show higher mechanical and actuation anisotropy than those fabricated with directionally placed polyurethane filaments [347]. Obviously, mechanical anisotropy has strong influence on actuation anisotropy.

Table 5.1 Comparison of mechanical, electrical and actuation anisotropy of actuator with aligned CNT sheet electrodes and actuators with fiber reinforcement. In all but one case (as noted) the D-EAP material is VHB-4905 acrylic.

<table>
<thead>
<tr>
<th>Electrode material</th>
<th>Mechanical (Electrode)</th>
<th>Electrical (Electrode)</th>
<th>Actuation</th>
<th>Reference</th>
</tr>
</thead>
<tbody>
<tr>
<td>carbon grease (D-EAP is the unidirectional polyurethane fibers reinforced VHB, 5 v%)</td>
<td>~1.4</td>
<td>~2.3</td>
<td>[347]</td>
<td></td>
</tr>
<tr>
<td>as-drawn CNT sheet</td>
<td>10</td>
<td>4</td>
<td>7.5</td>
<td>[364]</td>
</tr>
<tr>
<td>as-drawn CNT sheet</td>
<td>7.87</td>
<td>3.55</td>
<td>4.2</td>
<td></td>
</tr>
<tr>
<td>combed CNT sheet</td>
<td>11.64</td>
<td>3.86</td>
<td>infinite anisotropy</td>
<td>this work</td>
</tr>
<tr>
<td>etched CNT completely etched</td>
<td>-</td>
<td>3.45</td>
<td>-</td>
<td></td>
</tr>
<tr>
<td>etched CNT striped pattern</td>
<td>3.78</td>
<td>-</td>
<td>48.2</td>
<td></td>
</tr>
</tbody>
</table>
5.4 Conclusion

We have demonstrated a number of ways to alter and potentially improve the electrical and mechanical properties of CNT sheet electrodes in the context of D-EAP actuators. The results illustrate that the utility of microcombing and/or laser ablation processes on orientation of CNT fibers as well as mechanical anisotropy of CNT sheet electrodes resulting in substantial enhancement in the directional actuation performance of D-EAP actuators. The laser ablation process, was effective in selectively and precisely removing CNT fibers form the D-EAP surface while maintaining electrical conductivity. Hence the process can be used to apply transparent electrodes on film surfaces. On the basis of these results, aligned CNT sheets show great promise in D-EAP actuators with customizable anisotropic electromechanical performance.
CHAPTER 6 Anisotropic D-EAP and Application in Spring Roll Actuator

Abstract

D-EAP linear actuators are promising in many applications, such as microrobots, haptic devices and smart wearable devices etc. Spring roll actuator consists of a compressed spring in the core and wound elastomer roll as shell, which is one type of D-EAP linear actuator configurations. Rolled structure imparts compact nature to the actuators able them to generate high stroke. Compressed spring’s supporting allows to apply prestrain to elastomers that is well known process to improve dielectric strength of D-EAP. Also, it is possible to tuning the whole actuator’s stiffness by changing the spring rate. Rolled elastomer structural shell is very compact as comparing to thin film flat actuator and thus able to generate high force and stoke. This chapter analytically builds model to investigate the influence of mechanical and configurational parameters of spring roll on the final actuation performance. In addition, both neat D-EAP and fiber reinforced D-EAP composites are used to fabricate actuators. Improved actuation strokes are observed in fiber reinforced samples due to the fiber supporting effect. In the end of chapter, aligned CNT sheets are used as electrodes in neat D-EAP spring roll actuator and similar or lower actuation stroke of these actuators are observed while compares to that of neat elastomer samples. The results of this chapter demonstrate the route to improve actuation performance of D-EAP spring roll actuator, moreover, the principles behind this improvement are well explained by analytic model and will be useful for further design similar structured linear D-EAP actuators.
6.1 Introduction

Spring roll actuator has the very unique configuration among D-EAP actuators that is combined a compressed spring in the core and a roll of elastomer shell outside. Many advantages stem from this structure, including directly transmission of thrust stroke, compact structure, free-stranding, potential of possessing high electroelastomer-to-structure weight ratio, high stroke, versatile deformation styles such as one degree of freedom (DOF), 2 DOF and 3-DOF, relatively longer shelf lifetime than that of flat formed actuators and potential to be developed for capacitive strain sensor [12,380]. Hence, there are a wide range of applications of rolled structural D-EAP actuators, such as robotic inchworm-like propulsion system[8], human-like face with endowing expressivity [381], legged robots (known as FLEX 2, and Skitter Robot) [247], arm wrestling robot [11], and haptic feedback devices [13,118].

D-EAPs’ flexible nature makes it is possible to form rolled structure and high possibility to tailor the structure [115]. There are many diverse designs of rolled actuator. Spring as pretensioner can be replaced by elastomers cylinder [311] or external frame [309]. EAP materials employed in roll actuator includes the most used acrylic elastomers [12,13], silicone [363] and PVDF [382]. Beside the spring roll, various configurations have been developed on basis of rolled structure, including core-free rolled D-EAP actuators [115,250], tube spring actuator [301], and tube core-free actuator [307].

Although, many unique advantages make spring roll actuator attractive, some drawbacks are still as obstacles for its practical applications. One of the major one is the radial press from outside layers of elastomers toward the inner layers. Because inner layers are pressed by the
high pressure, thickness of individual layer as a function of radius position decreases while radius decreases (closed to core) and in the extreme case there is an upper limitation of number of layers able to be wound around spring [12]. Pei and coworkers reported spring roll actuator with 20 layers elastomer had the maximum strain of 26% whereas the 35 layers sample only shown 23%. This reduction might is not obvious, but it at least indicates that extra 15 layers did not further contribute to improve the stroke of actuator[12]. This might because the high radial pressure significantly reduced the cross-section area of elastomers. Also, Zhang and coworkers analytically calculated the standardized film thickness as a function of layer number position, which indicates thickness decreases from most outside to the core of the actuators [13]. On the basis of Zhang’s work, Kovacs calculated distribution of radial pressure on different layers of spring roll actuation. It shows from core to most outside, the radial pressure exponentially reduce and as number of layers increases the radial pressure in the most inside increases linearly [11].

To address this problem, many researches related to the solution have been done. Firstly, a method to remain the prestrain of elastomers is to impart the guest polymeric interwork to interpenetrate into prestrain elastomers networks and after curing to fix the shape, which is called interpenetrating polymer network (IPN). Kovacs and coworkers employed IPN post treated VHB materials to fabricate spring roll actuator which shows a longer lifetime and better specific volume efficiency of the actuator. But the major drawback is the limited elongation [304]. Moreover, the corrugated electrode structure silicone D-EAP with silver electrodes was explored in application of spring roll actuator due to no external prestraining mechanism required [305,306]. Moreover, a practically effective method is embedding parallel aligned
stiff fibers into elastomers to support the overall structure. For example, Bolzmacher and coworkers employed Nylon monofilament to hold silicone in a prestrained state [363], and similar observations were reported by other reference [307]. Additionally, Huang and coworkers used aligned stiff fibers to reinforce VHB elastomers and fabricated tube actuators. This work observed 1) inactive tubes with fibers did not show necking while it is stretched by a constant weight in axial direction, but sample without fibers showed; 2) upon the same electric potential, axial actuation strain increases of fiber reinforced tube actuator; 3) large actuation strain can be achieved by fiber reinforced elastomers even without prestrain, which cannot be observed in neat VHB samples [362]. However, this work only described the observations and did not theoretically explain the reason of enhanced actuation strain of fiber reinforced VHB composites and the functions of the fibers. There are many other works have conducted analytic modeling of spring roll actuator [13,383,384], which are helpful to understand the working mechanisms of spring roll actuator.

This chapter experimentally and analytically investigates using three types of monofilaments made of polyamide (PA), polyurethane (PU) to sandwich with VHB films and further employing these composites to fabricate spring roll actuators. The results show fiber reinforced spring roll actuators have larger stroke as comparing to net VHB samples, which majorly because that less supporting stress in hoop direction of elastomeric roll is required with fibers’ supporting. Spring roll actuator, made of VHB composites reinforced by PA fibers with the highest stiffness, has the largest stroke than other samples. In the end, this chapter employed aligned CNT sheet electrodes in neat VHB spring roll actuator to examine the influence of CNT rigid mechanical nature and induced anisotropic actuation property on performance of
spring roll actuators. A less stoke was observed in CNT electroded spring roll actuators that may attributes to the interlayer constraint between adjacent tacky elastomers films attaching together with dry CNT sheets electrodes.

6.2 Materials and sample preparation

6.2.1 Spring

The dimensions and rate of compression spring (Small Parts, Inc.) are listed in Table 6.1. In order to check the force to deform it by unit length (spring rate), compression test of spring was conducted using MTS-30G load frame operated at a crosshead speed of 10 mm min$^{-1}$, see load-displacement curve in Figure 6.1 (a).

<table>
<thead>
<tr>
<th>Material type</th>
<th>Stainless steel</th>
</tr>
</thead>
<tbody>
<tr>
<td>Free length (mm)</td>
<td>80.5</td>
</tr>
<tr>
<td>Compressed length (mm)</td>
<td>20.6</td>
</tr>
<tr>
<td>Outside diameter (OD$_{spring}$) (mm)</td>
<td>8.63</td>
</tr>
<tr>
<td>Wire diameter (mm)</td>
<td>0.63</td>
</tr>
<tr>
<td>Spring rate (N/mm)</td>
<td>0.14</td>
</tr>
<tr>
<td>Load capacity (N)</td>
<td>8.32</td>
</tr>
</tbody>
</table>

Table 6.1 Spring specifications
Figure 6.1 (a) Load-displacement curve of compression spring; (b) stress-strain curves of unidirectional tensile tests of monofilaments used to reinforce VHB composites. Inset is tensile young’s modulus results.

### 6.2.2 Fiber

Fibers used to parallel reinforce VHB (4905, 3M Corp., MN) composites include 3 types, including PU (Invista\textsuperscript{TM}), PA-I (PA 6, 4lb, South Bend\textsuperscript{®}) and PA-II (plasticized PA 6/66 copolymer) (Jarden Applied Materials Corp.). Fiber’s specifications are listed in Table 6.2, which show highly different dimensional and mechanical properties. Uniaxial tensile test results of all fibers are included in Figure 6.1 (b).
Table 6.2 Specifications of monofilaments used to reinforce VHB composites

<table>
<thead>
<tr>
<th>Material type</th>
<th>Diameter (μm)</th>
<th>Young’s modulus (MPa)</th>
<th>Fiber volume fraction in composites (%)</th>
</tr>
</thead>
<tbody>
<tr>
<td>PU</td>
<td>145</td>
<td>10.1</td>
<td>5.17</td>
</tr>
<tr>
<td>PA-I</td>
<td>215</td>
<td>1887.4</td>
<td>11.55</td>
</tr>
<tr>
<td>PA-II</td>
<td>76</td>
<td>2307.5</td>
<td>1.47</td>
</tr>
</tbody>
</table>

6.2.3 Fiber/VHB composites

In the composites, parallel aligned monofilaments are in equal space of 3mm and sandwiched between two layers of uniformly prestrained VHB films (200% linear strain) bonding together by tacky surface of VHB, see Figure 6.2. The thickness, the specific section of composites where fibers are located, was tested by thickness meter. Assuming fibers remain dimension in composites, VHB film surrounding the fibers are deformed. According to the fiber numbers in the unit width of VHB film and their diameters, fiber volume fractions are calculated shown in Table 6.2.

![Figure 6.2 Schematic illustration of fiber reinforced VHB composites cross-section within 3mm length.](image)

160
6.2.4 Spring roll actuator preparation

The process of building spring roll actuator is shown in Figure 6.3. Two layer of VHB film were firstly stretched alone x- and y- direction uniformly by 200% linear strain. In the bottom layer, 11 fibers were parallel added with equal space of 3mm and pressed slightly to stick onto VHB film tacky surface. A very thin layer of carbon grease electrode (rectangular with length of 84mm and width of 30mm) was brushed on the surface of bottom VHB layer. Then, the top layer was carefully laminating on the prepared bottom one (limiting bubble forming and aligning two electrodes). In each layer, one electrically conductive tape (9712, 3M Corp., MN) lead was added for later connecting electrodes to power supply. Until here, uniaxial fiber reinforced (in y-direction) VHB composites were prepared.

To fix the compressed spring, it was kept between two nuts fastening on a thread bolt inserting into spring’s core. There were two shoulder washers inserting into spring ends before putting nuts that are used as supporting places for later trussing VHB film on spring. Then slowly rolling this compressed spring assembly on the top surface of fiber/VHB composites alone fiber length (y-) direction (to avoid electric shorting failure, laminates without electrodes at two ends of rectangular active area were left in the same length of 6mm), a spring roll actuator was finally built. Before unscrew the two nuts, zip ties were used to fasten at the two ends firmly to keep VHB integrated with spring. For comparison purpose, control sample without fibers reinforcement was prepared too.
6.3 Characterizations of fiber/VHB composites flat film actuator and spring roll actuator

6.3.1 Mechanical tests of fiber/VHB composites

To test mechanical properties of fiber reinforced VHB composites, MTS-30G load frame was used to conduct unidirectional tensile test at cross-head speed of 10mm min\(^{-1}\) alone both fiber (y-) and cross-fiber (x-) direction. Composites samples are prepared flowing almost the same route as that same section in processing to prepare spring roll actuator. The difference is that composites was not wound on spring but fixed on an annular rigid frame (inner dia. 45mm). Only 6 fibers are sandwiched in the middle of the two prestrained VHB films and no electrode was added. Clamp width is 15mm, see Figure A1 in APPENDIX for more details.
6.3.2 Blocking force tests of flat film actuator made of fiber/VHB composites

The force required to return a fully activated actuator to its original state is defined as blocking force. In a planar actuator, it is the in-plane force exerted toward the inactive area at the boundary. The fiber/VHB composites flat circular actuators used for blocking force test are similar as the one of mechanical test. Differences is that square electrodes (15mm × 15mm) were painted on both sides of two layers of VHB (in total 3 layer of electrodes). Two clamps of MTS-30G load frame held the composite with isometric space of 21mm, and rectangular active area was located in the middle of two clamps (tensile direction along the length direction of the rectangular electrode). Before applying electric field, the inactive area above top clamp needs to be cut off for the purpose of avoiding force against clamp while stretching in the test. Then an electric field was applied, meanwhile a real-time video of the actuator deformation was record by video camera and force was record by MTS frame (actuation set-up and parameters same as those in the following section), see Figure A4 in APPENDIX.

6.3.3 Field-induce deformation characterization of flat film actuator and spring roll actuator made of fiber/VHB composites

Electromechanical responses of both flat film actuators and spring roll actuators were examined by a customized set-up consisting of a Bertan 225-30R high-voltage power supply (Spellman High Voltage Electronics Corp., Hauppauge, NY), a digital video camera and a computerized voltage-trigger. While the actuators were activated by an increasing voltage up to 5 kV in steps of 1 kV, the active area was recorded using the video camera in real-time to monitor the deformation. The acquired video images were analyzed frame by frame using image analysis software (ImageJ, NIH), to determine the actuation strain distribution in the
sample as shown in Figure A3 of APPENDIX. Flat actuators used here have similar configuration as those used in blocking force tests but the active area is circular with diameter of 12mm.

6.4  Result and discussion

6.4.1  Mechanical test results of fiber/VHB composites

Fiber reinforced VHB composites are both stretched in fiber(y-) and cross-fiber(x-) directions. Firstly, in y-direction, stress magnitude at the same strain from high to low in turn is the PA-I reinforced, PA-II reinforced, PU reinforced composites and pristine VHB matrix. Although PA-II fiber itself has the highest modulus, its composites has lower fiber volume fraction than that of composites reinforced by PA-I fibers. So PA-I composites mechanically perform superiorly. In order to express the mechanical anisotropy, a ratio ($A_m$) between composites modulus in fiber ($E_y$) and cross-fiber direction ($E_x$) is calculated. PA-I and PA-II composites has $A_m$ value of 64.3 and 21.2, respectively. By normalized the modulus by fiber volume fraction the resultant relative composites moduli are following the same rank as that of fibers. Highly distinguished from PA-I and PA-II composites, PU reinforced sample has very low modulus in both fiber and cross-fiber direction (1.88MPa and 1.65MPa respectively) due to the soft nature of PU fiber. It results in an insignificant anisotropic mechanical property ($A_m=1.1$).
Figure 6.4 Unidirectional tensile test results of VHB and its composites. Stress-strain curves of strain alone (a) fiber direction (y) and (b) cross-fiber direction(x). (c) Initial modulus of composites straining in both fiber(y) and cross-fiber(x) directions.

It is worth to mention one observation of self-supporting phenomenon of composites in tensile testing. Before clamp starts to stretch, the area of composites above the top clamp was cut off as mentioned in experimental section. Different cutting collapsing structures are observed, see Figure 6.5. The cut area of PA-I composites show the self-supporting structure in which fibers well hold the prestrained VHB films (Figure 6.5 (a)), PA-II composites shown the less supported structure and PU composites show almost collapsed configuration. All of these imply that reinforcing fibers are able to play a role of “skeleton” in this composites to support prestrain VHB “muscle”.

Figure 6.5 Photographs of cutting off edge of composites located above the top clamp before tensile testing. (a) PA-I reinforced composites, (b) PA-II reinforced composites and (c) PU reinforced composites.
Qualitatively, stress loaded to the top clamp after cutting also was recorded, see Figure 6.6. Stress presented here are normalized to divide force by cross-sectional area of composites (including fibers cross-sectional area). It is notable that in fiber direction, significant less stress is required to supporting the PA-I and PA-II fiber reinforced composites as comparing that of pristine VHB. This agrees well with those observations of self-supporting structures of these two composites as mentioned before. PU fiber reinforced composites perform differently that needs larger stress to support the prestrained elastomers. In x-direction, the differences between all composites samples and neat VHB are negligible.

Figure 6.6 Stress supporting the prestrained VHB. It is recorded in tensile testing after mounting the sample, clamping samples and zeroing load cell, then the force was recorded after cutting off the film above the top clamp.
6.4.2 Field-induced electromechanical response tests

Actuation performance of flat circular actuators made of fiber reinforced VHB composites are examined, see Figure 6.7(a). Firstly, it is notable all fiber reinforced actuators deform in anisotropic manner that actuation strain in fiber (y-) direction is less than that in cross-fiber (x-) direction. Differently, pristine VHB deform isotopically upon applying the electric field. At the same electric field, actuation strains in y-direction, the largest of PU reinforced composites and the least of PA-I reinforced ones are observed. For example, at 110 V µm⁻¹, strain in y-direction is 3.0%, 4.7% and 13.9% of PA-I, PA-II and PU fiber reinforced composites, respectively. This can be explained by the different mechanical supports from fibers. As discussed in mechanical testing results, PU reinforced composites has the lowest young’s modulus in y-direction among all three types of fiber reinforced composites. Hence, under the same electric field, PU composites may deform more than other two types. In x-direction, the strains are in the reversed ranking as that in y-direction but the difference between samples are insignificant.
Figure 6.7 (a) Linear actuation strain as a function of true electric field of flat circular actuators. (b)-(e) is actuation video capture of VHB, PA-I/VHB composites, PA-II/VHB composites and PU/VHB composites actuator before actuation, respectively; (f)-(i) is capture of them at about 110 V/μm.
6.4.3 Blocking force test results of flat film actuator made of fiber/VHB composites

Upon actuation actuators deform between two isometric separated clamps that attaching to load cell to record the force against them. Because in spring roll actuators, fibers aligned in the circumferential (or hoop) direction, only the force in cross-fiber direction was tested here that directly related to extending of spring roll in longitudinal direction.

Figure 6.8 (a) show a typical force-time curve in blocking force testing. After mounting sample (force was zeroing), area above top clamp was trimmed and the force increases to a certain value, for example it is ~0.65N here. Until the force is stable after cutting, electric field is applied and thus a compressive force is generated in thickness direction of actuator. The compressive force is transferred to in-plane force and against to clamp. While actuation finishes, the load comes back to a value close to that of before actuation. At the same time, a real-time actuation video is recorded. By matching the load result with video through the actuation process by time records, force as a function of true electric field is achieved. And blocking stress is further calculated via dividing force by initial cross-sectional area of actuator in clamp edge, see Figure 6.9.
Figure 6.8 Blocking force testing results of PU fiber reinforced composites. (a) force-time curve; (b-c) photographs of actuator before and after actuation, respectively.

Figure 6.9 Blocking stress as a function of true electric field of fiber reinforced composites
While electric field less than 80, difference in blocking stress between all samples is insignificant at the same electric field. The similar trend is also observed in results of actuation strain in cross-fiber(x-) direction of flat film actuators (Figure 6.7). And if the moduli in x-direction are similar among all samples, their output stresses are expected to be close to each other. As electric field higher than 80 V µm\(^{-1}\), PA-I and PA-II composites behave similarly and show the higher blocking stress at the same electric field. This may mainly because of their high actuation anisotropy. On the contrary, PU reinforced composites show the lowest stress due to less mechanical and actuation anisotropy. In summary, fiber reinforced composites actuators have similar output blocking stress at lower electric field, and only PA-I and PA-II reinforced samples generate higher stress than VHB while electric field is higher than 80 V µm\(^{-1}\).

6.5 Working principle of spring roll actuator

Spring roll actuator’s working principle has been explored in previous work [385], see Figure 6.10. There are four status are important for understanding their working mechanism.

(1) Passive equilibrium statue. At this stage, in spring roll actuators, there is a force equilibrium in cross-section between compressed spring and prestrained VHB or its composites. In the load-length curve, it corresponds to the intersection between curves of compressing spring and that of tensioning inactive elastomer roll. (electric potential U=0)

(2) Initial moment of activation. This is the moment when just starts to apply electric field cross electrodes of the actuators and a compressive force is generated in the thickness direction of elastomer film at the moment. (U>0)

(3) Active equilibrium. In this state, a force balance is again existing in the cross-section of actuator between the actuated elastomer VHB roll and spring. Actuation
compressive force in the thickness of elastomers, it also is transfer towards in-plane directions and helps supporting the prestrained elastomer roll. So this state corresponds to the intersection between curve of compressing spring and that of tensioning active elastomer roll in the load-length plot. \((U>0)\)

(4) Initial moment of deactivation. This is the moment when begins to stop applying electric potential. So there is no actuation compressive force in thickness of roll and spring tends to recover back to passive equilibrium state. \((U=0)\)

Figure 6.10 Working principle of spring roll actuator (redrawn from reference [385])
6.6 Fiber reinforced VHB composite spring roll actuator

6.6.1 Passive equilibrium statue

As described before, in passive equilibrium state, extending force of the compressed spring and contractive force of prestrained elastomer roll are balanced in the cross-section of spring roll actuator, see Figure 6.11.

Before winding elastomer film on spring, it is prestrained biaxially in both x- and y-direction. The stress required is $\sigma_{px}$ and $\sigma_{py}$ in x- and y-direction, respectively. They can be calculated from strain and modulus of elastomer ($E_{elst}$) assuming the elastomer materials behave isotopically and linearly following Hook’s law as stretching.

$$\sigma_{px} = E_{elst} \cdot \varepsilon_{px}$$

$$\sigma_{py} = E_{elst} \cdot \varepsilon_{py}$$

Equation set 6.1
After winding elastomer film surrounding spring, x-y-z coordinates are transferred to L-r-h coordinates. A unit cell of elastomer roll located in circumference with radius of \( r \) is subjecting three directional stress: \( \sigma_L \), \( \sigma_r \), and \( \sigma_h \). \( \sigma_L \) is the stress alone longitudinal direction of spring roll and is sum of spring extensive force and elastomer contractive force. \( \sigma_h \) is the stress alone circumferential direction of roll and is elastomer prestraining stress in y-direction. \( \sigma_r \) is the stress alone spring radius direction and is sum of spring supporting forcing and collapsing force of VHB roll toward the core of spring.

\[
\begin{align*}
\sigma_L &= \sigma_{px} - \sigma_{sprL} \\
\sigma_h &= \sigma_{py} \\
\sigma_r &= \sigma_{elstr} - \sigma_{sprr}
\end{align*}
\]

Equation set 6.2

Where, \( \sigma_{sprL} \) is the extensive stress of spring in L direction, \( \sigma_{elstr} \) is the contractive stress of elastomer in r direction and \( \sigma_{sprr} \) is the supporting stress of spring in r direction.

In equilibrium state, all three stresses in Figure 6.11 should be zero. Because of symmetry in circumferential direction, \( \sigma_h \) is canceled and balanced.

\[
\begin{align*}
\sigma_{sprL} &= \sigma_{px} \\
\sigma_{sprr} &= \sigma_{elstr}
\end{align*}
\]

Equation set 6.3

If considering the tube with radius of \( r \) in roll as the thin-walled rube (generally defined as ratio between tube’s thickness to its radius is less than 0.1), then its radial stress (\( \sigma_{tube-r} \)) towards the core of the spring could be expressed as a function of radius \( r \), see Equation 6.4.
\[ \sigma_{\text{tube-}r}(r) = \frac{\sigma_h \cdot dr}{r} \]  

Equation 6.4

Where \( dr \) is the wall thickness of this tube, \( r \) is its radius, then \( \sigma_{\text{elst}} \) is the integration of \( \sigma_{\text{tube-}r} \) from radius \( r \) to \( r_o \),

\[ \sigma_{\text{elst}}(r) = \int_r^{r_o} \frac{\sigma_h \cdot dr}{r} = \sigma_h \ln \frac{r_o}{r} \]  

Equation 6.5

So the constitutive relationship of the spring roll in passive equilibrium states can be summarized as:

\[ \sigma_{\text{sprL}} - E_{\text{elst}} \cdot \varepsilon_{px} = 0 \]

\[ \sigma_{\text{spr}} - \sigma_h \ln \frac{r_s}{r_i} = 0 \]  

Equation set 6.6

Assume the spring has infinite modulus, so the boundary condition should always be most inner layer radius equals to \( r_i \) (the outside diameter of the spring).

Above is the algebraic derivations, the following is the numerical analysis on the basis of experiment results.

Firstly, in L-direction, the force corresponds to this stress is given as,

\[ F_{\text{sprL}} = \sigma_{\text{sprL}} \cdot \frac{t_0}{(1 + \varepsilon_x)(1 + \varepsilon_y)} \cdot l_{\text{roll}} = 0.387 \text{MPa} \times 2 \text{layer} \times 0.055 \text{mm} \times 96 \text{mm} / \text{layer} = 4.09 \text{N} \]

Then compressed length of spring under \( F_{\text{sprL}} \) is \[ d_{\text{sprL}} = \frac{F_{\text{sprL}}}{\text{spring rate}} = \frac{4.09N}{0.14} = 29.2 \text{mm} \]

and resultant length of actuator should be 51.3 mm, see Figure 6.12.
However, according to experiments results, see Table 6.3, in general 61mm-62mm is the free-standing length of actuator to supporting 200% biaxial prestrain roll in length of 96mm (84mm is active length and 12 are the two ends of inactive areas). It implies the calculated force $F_{sprL} = 4.09N$ is larger than actually required value. It is maybe because of two main reasons. The first is that there is about 3mm pitch between the neighboring wires of spring, so after winding VHB film on rigid spring, the area of elastomers between two spirals are concavely hanging and there is an angle (about 25° for neat VHB actuator) between hanging VHB film and the longitudinal axis of spring roll, see Figure 6.13 (a-b). It may result in components (F1 and F2) of prestraining VHB force along film length direction (F). F2 is about $\cos(25°)F \approx 0.9F$. 

![Figure 6.12 Load-length curve of spring and inactive VHB roll](image)
Table 6.3 Dimensions of the spring roll actuator

<table>
<thead>
<tr>
<th>sample</th>
<th>length (mm)</th>
<th>outside diameter ((2r_0)) (mm)</th>
<th>t(^7/) OD(_{spr})</th>
<th>mean thickness per layer of composites (mm)</th>
</tr>
</thead>
<tbody>
<tr>
<td>VHB</td>
<td>62.00</td>
<td>9.21±0.02</td>
<td>0.07</td>
<td>0.097</td>
</tr>
<tr>
<td>PA-I spring roll</td>
<td>61.33±0.58</td>
<td>9.81±0.10</td>
<td>0.14</td>
<td>0.197</td>
</tr>
<tr>
<td>PA-II spring roll</td>
<td>61.50±0.71</td>
<td>9.42±0.04</td>
<td>0.09</td>
<td>0.132</td>
</tr>
<tr>
<td>PU spring roll</td>
<td>62.00±1.41</td>
<td>9.47±0.07</td>
<td>0.10</td>
<td>0.140</td>
</tr>
</tbody>
</table>

* t is overall thickness D-EAP tube surrounding the spring \(t=2r_0 - OD_{spr}\)

The second reason is radial stress (see, Figure 6.13 (c)) from the outside layers of elastomers to the inner layers. This presses inner layers and also helps to extend VHB elongating in longitudinal direction. As shown in Figure 6.13 (c), inner layers of VHB elastomer is subjected higher radial pressure than those located in outside layers. In addition, as number of layers increase this stress increases. In the work, 6 layers neat VHB spring wall was built and the most inner layer was compressed by stress of about 0.035MPa.
Figure 6.13 Photographs of (a) compressed spring and (b) spring roll actuator made of VHB elastomers. (c) Radial pressure ($\sigma_{elstr}$) as a function of radius in the spring roll actuator (assuming each layer thickness $t$ is 0.055mm with $\sigma_h=0.468$MPa and spring outside diameter $r_i$ is 8.63mm)

For fiber reinforced composites, assuming fibers are forming flat spirals inside actuators, space between the $k^{th}$ and the $(k+1)^{th}$ layer (equal to $r_{k+1} - r_k$) is larger than thickness of composites where no fiber is added, see Figure 6.14 (a-b). All actuators are made with same length of elastomer film, so before comparing radial pressure of different actuators, the number of rolling layers in all actuators needs to be calculated. Figure 11(c) shows the cross-sectional view of flat spiral with inner radius of $r_i$, outside radius of $r_o$, thickness of $t$ and distance between neighboring layers is zero.
Figure 6.14 Cross-sectional images of spring roll actuator made of (a) neat VHB and (b) fiber reinforced composites. (c) Cross-sectional view of flat spiral with inner radius of $r_i$, outside radius of $r_o$, thickness of $t$ and distance between neighboring layers is zero.

By assuming every layer is a full circular, total length of wire in a flat spiral is $L$ expressed as Equation 6.7. Because $L$ is known for all case of 96 mm, $N$ can be calculated as shown in Table 6.4. All actuators have number of layers in range of 3.2-3.5 close to 3. So in the following radial pressure calculations, $N=3$ will be used in all cases.

$$L \approx \sum_{0}^{N} 2pi[r_i + (N-1)t]$$

Equation 6.7
Table 6.4 Number of layers in spring roll actuators with constant length

<table>
<thead>
<tr>
<th>Sample</th>
<th>thickness of composites with fibers $t_f$ (mm)</th>
<th>number of layer $N$</th>
</tr>
</thead>
<tbody>
<tr>
<td>VHB</td>
<td>0.113</td>
<td>3.5</td>
</tr>
<tr>
<td>PA-I</td>
<td>0.276</td>
<td>3.2</td>
</tr>
<tr>
<td>PA-II</td>
<td>0.129</td>
<td>3.4</td>
</tr>
<tr>
<td>PU</td>
<td>0.174</td>
<td>3.3</td>
</tr>
</tbody>
</table>

As mentioned before spring roll actuator made of fiber reinforced composites have spaces between layers where no fibers are added, thereby in calculation of radial pressure, assuming the pressure still is completely transferred between layers, Riemann sums is used instead of integration used in pure VHB fabricated actuator as expressed in Equation 6.8.

$$\sigma_{spre} - \sum_{i=1}^{N} \frac{\sigma_h \cdot t_{VHB}}{r_i + (N-1)t_f} = 0$$

Equation 6.8

Where, $t_{VHB}$ is the thickness of composite where only has two layers of VHB film attached, $t_f$ is thickness of composites where is with fibers.

Radial pressure of elastomer as a function of radius position for all case are shown in Figure 6.15. Spring roll actuator made of neat VHB has the highest radius pressure in the core due to the highest hoop stress $\sigma_h$. PA-I and PA-II reinforced composites spring roll in sequence shows the less pressures due to the lower $\sigma_h$ and larger radius. PU reinforced sample shows
similar radial pressure of the most inner layer as that of neat VHB sample because they have similar $\sigma_n$ and insignificant difference in radius.

Radial pressure is a highly important parameter here. With the same number of layers, the higher the radial pressure in the spring roll actuator may induces 1) the higher compressive deformation of inner layers, 2) the larger constraint between layers while actuation.

![Figure 6.15 Radial pressure of elastomers as a function of elastomer radius position of spring roll actuators made of neat VHB and fibers (PA-I, PA-II and PU) reinforced composites.](image)

6.6.2 Initial moment of activation

When electric potential is applied across the electrodes of spring roll actuators, a compressive stress is generated in thickness direction of elastomer film. Maxwell stress ($\sigma_M$) is normally used to express this compressive stress:
\[ \sigma_M = \varepsilon_0 \varepsilon \left( \frac{U}{t} \right)^2 \]  

Equation 6.9

Where \( \varepsilon_0 \) and \( \varepsilon \) is the free space permittivity and material relative permittivity, respectively.

U is the electric potential applied and t is the thickness between two electrodes.

If elastomers are considered as fluid, stresses should be uniform in every direction [116]. This assumption makes it is possible to relate the compressive stress to in-plane stresses.

\[
\begin{align*}
\Delta \sigma_L &= \sigma_M \\
\Delta \sigma_R &= \sigma_M \\
\Delta \sigma_r &= -\sigma_M
\end{align*}
\]

Equation set 6.10

6.6.3 Active equilibrium

This status is force equilibrium between compressed spring and active elastomer roll in cross-section of actuator. This new force equilibrium could be summarized as Equation set 6.11.
\[ \sigma_L' = \sigma_{px}' - \sigma_{spr}' = 0 \]
\[ \sigma_h' = \sigma_{py} - \sigma_M = 0 \]
\[ \sigma_r' = \sigma_{elstr}' - \sigma_{spr}' = 0 \]

Equation set 6.11

Where \( \sigma_{px}' \), \( \sigma_{spr}' \), \( \sigma_{elstr}' \) and \( \sigma_{spr}' \) remains same meaning as expressions in passive equilibrium but they are presenting the corresponding values in active state here.

According to blocking stress testing results of flat film actuators, Maxwell stress \( \sigma_M \) is smaller than prestraining stress in hoop direction, so \( \sigma_h \) stress is larger than zero and thereby radial pressure for activated rolls is still exiting. On the basis of this, the boundary condition is that the most inner layer is still supported by spring and neighboring layers are attached. The resultant radial pressure upon actuation can be expressed as,

\[ \sigma_{elstr}'(N) = \frac{(\sigma_{py} - \sigma_M )[t_{VHB}'(1 - \frac{\sigma_M}{E_z})]}{(r_0 + N \cdot t_f')} \]

Equation 6.12

\[ \sigma_{elstr}' = \int_1^N \frac{(\sigma_{py} - \sigma_M )[t_{VHB}'(1 - \frac{\sigma_M}{E_z})]}{[r_0 + (N - 1) \cdot t_f']} \]

In longitudinal direction, \( \sigma_{px}' \) is not simply equaling to \( \sigma_{px} - \sigma_M \), because in this active equilibrium strain of VHB roll increases, the stress needs to strain it is no longer the same as that of prestraining. It will be easier to understand this equilibrium by reviewing the load-length curve of spring roll actuator working principle, see Figure 6.17. If to assume both spring and inactive/active elastomer roll mechanically response linearly, a triangle can be built through points (1), (2) and (3). Distance between point (1) and (2) is considered consisting of
two components, F1 and F2. F1 is load required to extend spring from state (1) to (3), and F2 is load used to extend the activated elastomer rolls from state (2) to (3). And the sum of F1 and F2 is blocking force generated by elastomer roll upon actuation. There are two angles in this diagram are important, $\alpha$ and $\beta$. This because tangent ($\alpha$) and tangent ($\beta$) is slope of spring and elastomer roll (representing spring rate $\kappa$ and elastomer roll elastic constant $E_{elst}$ which is the slope of the load-length curve if assuming it is a linear fitting), respectively. And relations between F1, F2 and $\alpha$, $\beta$ as well as the displacement of actuator $\Delta l$ are list in Equation set 6.13. Be solving Equation set 6.13, spring roll actuation displacement $\Delta l$ can be expressed as Equation set 6.15 (similar relation of actuator stiffness can be found in reference [12], equals to sum of stiffness of VHB roll and spring).

Figure 6.17 Force equilibrium in spring roll actuator at (1) passive equilibrium, (2) initial moment of activation, (3) active equilibrium, and (4) initial moment of deactivation.
\[ \Delta l = \frac{F_1}{\tan \alpha} = \frac{F_2}{\tan \beta} \quad (1) \]

\[ F_1 + F_2 = F_{blc} \quad (2) \quad \text{Equation set 6.13} \]

\[ \Delta l = \frac{F_{blc}}{\tan \alpha + \tan \beta} \quad \text{Equation set 6.14} \]

\[ \tan \alpha = \kappa \]

\[ \tan \beta = E_{elst} \]

\[ \Delta l = \frac{F_{blc}}{\kappa + E_{elst}} \quad \text{Equation set 6.15} \]

\[ F_{blc} = \sigma_M \cdot A_{cr} \]

Based on Equation set 6.15, it is possible to tune displacement of spring roll actuator. For example, in order to enlarge the displacement, to increase blocking force and to reduce spring constant as well as elastomer modulus could be optional methods. Blocking force improvement could be realized by increasing the elastomer dielectric constant, properly increasing the voltage applied and decreasing the thickness of elastomers. In addition, to add more layers in spring roll actuator will increase the cross-sectional area and blocking force with the same Maxwell stress. However, this needs to be optimized because that the more layers added the larger radial pressure will compress inner layers and decrease their thickness.

Flowing is the active state stress calculation of spring roll actuators based on algebraic derivation. Firstly, in radial direction, in active state, Maxwell stress is generated cross each layer’s thickness direction and elastomer films are compressed to be thinner. Meanwhile the
stress in hoop direction ($\sigma_h$) decreases too. Hence, radial pressure decreases with boundary condition of radius remains same of most inner layer. Figure 6.18 shows the comparison of radial pressure before and upon actuation of all types of spring actuators. Assumptions are made here, including 1) fibers are not compressible and thus the outside diameter of spring roll remains; 2) VHB thickness used to calculation of Maxwell stress is thickness of unidirectional deformable VHB flat actuator under 5 kV ($\approx 0.044\text{mm}$).

![Graph of radial pressure vs radius position](image)

Figure 6.18 Radial pressure of elastomers as a function of elastomer radius position of spring actuators in passive and active status (U=5kV)

In longitudinal direction, length of spring roll actuator as a function of electric potential is shown in Figure 6.18. Neat VHB spring roll actuator shows the least length under the same voltage actuation as comparing to all of the composites samples. Among composite samples,
the rank of actuator mean length under same electric potential from highest to lowest is: PA-I reinforced sample, PA-II fiber reinforced one and the PU reinforced. The difference of longitudinal strain between the longest and shortest is about 2.5%.

Figure 6.19 (a) Field-induced actuation strain in longitudinal direction of spring actuator made of VHB and its fiber reinforced composites; (b) video captures of PA-II monofilament reinforced VHB spring roll actuator before actuation and upon 4kV electric potential.
Because all actuators have the same spring rate and inactive VHB elasticity, the displacement difference is mainly induced by the different blocking force under the same electric potential. Figure 6.20 shows the load-length curve of spring, inactive/active VHB roll and active composites roll. Curve of inactive VHB roll here is modified from Figure 6.12 through multiplying load value by a constant (0.57) to fit experimental actuator length in passive equilibrium. The reason to using this constant less than 1 is because of concaved roll surface on spring and radial pressure reducing cross-section area as discussed before. So after this modification, length of actuator matches with experiment data and slope of this curve in length range of 40mm-70mm decreases from 0.073 N/mm to 0.041 N/mm.

To plot active VHB/composites roll curve, it is firstly assuming elastomers/composites in this length range maintain linear mechanical property and subtracting the load value of inactive sample by the blocking force at the same length. Blocking force is used product between Maxwell stress and the radial cross-section area. Here in all case, uniform Maxwell stress is used that is the value generated by neat VHB under 4 kV electric potential. Actually, due to the radial pressure, thickness of each layer decreases from outside to inside of roll. So each layer has the different true electric field.

Cross-sectional area in radial direction of all spring roll actuators are different due to different radial pressure and with/without fiber separation between layers. First of all, for neat VHB sample, cross-section area appropriately equal to annual area between most outside of actuator and outside wall of spring. For composites sample, they all have different actuator outside diameters are both larger than the value of pure VHB roll without radial pressure (all remains the thickness between wrapping). Hence, it is assumed that flat spiral sections made of the
section of composites where fibers located are working as spacer to periodically separate neighboring layers and all VHB maintain thickness before being wound on spring, so all composites samples have the same radial cross-section area. The resultant load-length relationships for all actuators are calculated and plotted in Figure 6.20. Pure VHB actuator is expected to extend by about 5.5mm corresponding to 9.1% strain, and composites samples, differently, are expected to elongate by 7.5mm corresponding to 12.4%. These theoretical expectations agree well with the experimental results, excepting accuracy of the difference between three different types of composites.

![Load-length curve of compressing spring, tensioning inactive and activated neat VHB, VHB/composites roll](image)

Figure 6.20 Load-length curve of compressing spring, tensioning inactive and activated neat VHB, VHB/composites roll

Taken together of stress required to supporting prestrained VHB in flat film actuator (Figure 6.6) and radial pressure in spring roll actuator(Figure 6.15), it is clear that PA-I reinforced composites require the least stress to support stretched VHB elastomer and thus the least radial
pressure from outside layers toward the inside ones in roll form. Differently, PU fiber with significantly smaller modulus almost did not play a role of skeleton in the composites to supporting elastomer and thereby have the highest radial pressure, and PA-II composites have the medium supporting stress and the radial pressure. These practical conditions are different from assumption that all composites maintain same elastomer thickness.

As comparing to PA-I (1887.4MPa, 215 $\mu m$), PA-II fiber has the similar modulus (2307.5MPa) but much smaller diameter (76 $\mu m$), oppositely, PU fiber has significantly lower modulus (10.1MPa) but relatively smaller diameter (145$\mu m$). Also, PA-II composites have much less fiber volume (1.47%) than that of PU composites (5.17%). However, PA-II reinforced spring roll actuators extend longer than PU reinforced samples by ~12% under the same electric potential. This indicates rigidity of reinforcement fiber plays more important role in improvement of supporting affecting on elastomer and extensive performance of spring roll actuator. Although PU fibers have the lowest modulus, they still effectively improved the deformation ability of their spring roll actuator by ~20% while compares those neat VHB samples due to the fiber spirals fixed by surrounded elastomers still show profound anti-compressive effect. PA-I reinforced spring roll actuators have larger displacement than that of PA-II reinforced ones but has very large variations. This demonstrates, with similar rigidity of reinforcing fibers, increasing fiber volume fraction is effective to enhance fibers’ skeleton supporting effect to elastomers and thus to reduce radial pressure from outside layers towards to core of actuator. And if fiber diameter is too large as comparing to elastomer thickness, its composites cannot promise stable spring roll actuation extension.
6.7 CNT sheet electroded spring roll actuator

From the above section, it is known that employing fibers to reinforce elastomer in hoop direction of spring actuators are able to extend longer under the same electric potential. Hence it will be interesting to investigate using aligned CNT sheets to replace fibers and examining its influence on performance of spring roll actuators. Also, CNT sheets electrodes in “dry” state are different from carbon grease with “wet” nature. Carbon grease in wet form laminating with tacky VHB films is mechanically working as lubricates, whereas CNT sheets can transfer force between neighboring layers of elastomer without lubricating. Thus, it will be interesting to test properties of samples with different layers. In this section, aligned CNT sheets, works as not only the electrodes but also the reinforcement in spring roll actuators have been examined.

6.7.1 Spring roll actuator preparation and characterization

A new batch of CNT forest was grown especially for this work. Aligned CNT sheets are prepared following similar route as described in reference [379]. Instead of using carbon grease electrodes, CNT sheets were gently applied in processing spring roll actuator following the method described in section 4.3.1. Similar as all characterization methods in fiber reinforced composites spring roll actuators, CNT sheets electroded actuators also were examined.

6.7.2 Result and discussion

6.7.2.1 Mechanical test results of CNT-VHB flat film actuator

To characterize the mechanical properties, unidirectional tensile tests were conduct on CNT-VHB flat actuator in both fiber (y) and cross-fiber (x) direction. There are two types of CNT
electroded flat film actuators were prepared, 2 layers sample and 6 layers ones, see Figure 6.21 (b) and (c). Because VHB laminates used in spring roll before winding are two layer structure, same structured flat film was prepared and tested (two layers of elastomer films and 3 layers of electrodes). Here one more test of 6 layers sample was added that is laminates of 6 layers VHB films and 6 layers of CNT sheets electrodes.

Figure 6.21 Schematic illustration of structure of one layer, 2 layers and 6 layers flat film actuators

Similar as the described in Chapter 4 and 5, CNT electroded VHB flat actuators show anisotropic mechanical property, see Figure 6.22 (a-b). There is a slight difference between 2 layers sample and 6 layers one in both fiber (y-) and cross-fiber (x-) direction. 6 layers sample has the lower stress while comparing to that of 2 layer sample at the same strain level. This might be due to lack of alignment of CNT in the laminates. Under stretching, not well aligned CNTs firstly rotate which causes less contribution on force bearing and less tensile stress. Also,
6 layers sample has the lower modulus (in strain range of 8% to 9%) than that of 2 layers one, see Figure 6.22 (c).

![Unidirectional tensile test results of CNT sheet electroded VHB alone (a) fiber direction(y) and (b) cross-fiber direction(x).](image)

Figure 6.22 Unidirectional tensile test results of CNT sheet electroded VHB alone (a) fiber direction(y) and (b) cross-fiber direction(x).

Before tensile testing, stress required to support prestrained elastomer also was examined by cutting area of elastomers above top clamp and recording the force increase (testing details can be found in section 5.2.5). As Figure 6.23 (a) shows, CNT electroded VHB actuator have similar supporting stress as comparing to that of pristine VHB sample in both fiber and cross-fiber direction. This indicates although have high modulus, very thin CNT sheets cannot support the prestrained elastomers. It is also observed of collapsed structure at the cutting area as shown in Figure 6.23 (b). While number of layers increases, supporting stress shows slightly decrease. It might because the constraint effect between neighboring layers due to dry CNT electrodes attaching adjacent VHB films.
6.7.2.2 Field-induced electromechanical response test results

Aligned CNT sheets with anisotropic mechanical property constrain elastomer deformation differently along fiber (y-) direction and cross-fiber (x-) direction. As described in Chapter 4, linear actuation strain in x-direction is higher than that in y-direction of CNT-VHB flat actuator. In Figure 6.24, actuation strains of different CNT-VHB flat actuators with 12mm diameter active area are summarized, including single layer (VHB: 1 layers; CNT electrodes: 2 layers), 2 layers (VHB: 2 layers; CNT electrodes: 3 layers) and 6 layers samples (VHB: 6 layers; CNT electrodes: 6 layers), see Figure 6.21. Although single layer actuator has anisotropic actuation strain, in fiber direction it still deforms by about 20% strain at ~140 V/μm due to limited constraint from CNTs. On contrary to single layer actuator, 2 and 6 layers show almost absolute unidirectional deformation in cross-fiber direction because of interlayers constraints.
For comparison purpose, carbon grease electroded VHB actuators of 1 layer, 2 layers and 6 layers are also tested, see Figure 6.24 (b). The influence of adding more layers of VHB and electrodes on actuation strain is insignificant. As discussed before, this is because lubrication function of carbon grease electrodes.

![Figure 6.24](image)

Figure 6.24 Linear actuation strain as a function of true electric field of flat film actuator of 1 layer, 2 layers and 6 layers structure with (a) aligned CNT sheets electrodes and (b) carbon grease electrodes.

### 6.7.2.3 Blocking force test results

Blocking forces generated by flat film VHB actuators with CNT sheets and carbon grease electrodes are summarized as Figure 6.25 shows. Upon same electric field, they generate similar blocking stress output and that of CNT electroded sample is slightly lower.
Figure 6.25 Blocking stress as a function of true electric field of VHB actuators with CNT sheets electrodes and carbon grease electrodes.

6.7.2.4 Spring roll actuator performance characterization

Dimensions of the spring roll actuators made of CNT electroded VHB actuators are listed in Table 6.5. Their outside diameter is slightly larger than those with carbon grease electrodes. Because the thickness of single layer CNT sheet is very hard to test, which highly is affected by the pressure applied during testing. There is no significant difference between thickness of neat VHB part and that with CNT sheets on. In multiple layered structure, increase of thickness by adding CNT sheets may influences test results.
Table 6.5 Dimensions of the spring roll actuator with CNT sheet electrodes

<table>
<thead>
<tr>
<th>sample</th>
<th>length (mm)</th>
<th>outside diameter ((2r_o)) (mm)</th>
<th>(t_O/OD_{spr})</th>
<th>mean thickness per layer of composites (mm)</th>
</tr>
</thead>
<tbody>
<tr>
<td>VHB</td>
<td>62.00</td>
<td>9.21±0.02</td>
<td>0.07</td>
<td>0.097</td>
</tr>
<tr>
<td>CNT</td>
<td>61.50±0.71</td>
<td>9.31±0.02</td>
<td>0.08</td>
<td>0.113</td>
</tr>
</tbody>
</table>

On the basis of dimension of spring roll actuator and the mechanical, electromechanical results of flat circular actuator, radial pressure of spring roll actuation was calculated. As shown in Figure 6.26, electrodes type seems almost does not affect the radial pressure of spring roll actuator in both inactive and active status.

![Graph showing radial pressure of elastomers as a function of radius position.](image)

Figure 6.26 Radial pressure of elastomers as a function of radius position of spring actuators with CNT sheets and carbon grease electrodes in inactive and active status \((U=5\text{kV})\)

In the end, neat VHB spring roll actuator both with CNT sheets electrodes and carbon grease electrodes are examined, see Figure 6.27. While voltage beyond 2kV, actuation strain of actuators with CNT sheets electrode is lower than that of carbon grease electrodes.
As discussed before, these two types of sample have similar radial pressure at inactive and active status, which implies their cross-sectional area of VHB film are similar. The reason only can be used to explain reduction of actuation strain in CNT sheets electroded actuator might be interlayer constraints of VHB films in roll. And this effect is almost eliminated in carbon grease electroded actuator. Taken together of mechanical and actuation test results of two materials electroded flat actuators, there is large constraint between neighboring layers of films in actuator with CNT sheets electrodes. As an integrity, multiple layers of VHB are pasted to each other with CNT sheets in the middle. The lack of alignment in CNT sheets and non-uniform prestrain might against deformation of spring roll actuator.

![Graph](image.png)

**Figure 6.27** Field-induced actuation strain in longitudinal direction of neat VHB spring actuator with CNT sheets electrodes and carbon grease electrodes.

### 6.8 Conclusions

In summary, spring roll actuators, with simple working principle by maintaining force equilibrium between compressed spring and pre-stretched VHB elastomer roll, is a special
compact structured D-EAP linear actuator. With rigid spring as core, it is possible to apply prestrain to elastomer film that is known as effective method to improve elastomers’ dielectric breaking strength. The major drawback of spring roll actuator roots in radial pressure from outside layers of elastomer film towards inner layers. When this press is as large as the compressive strength of elastomer, it may cause mechanical failure of elastomer and thereby there is an upper limitation for number of layers of roll in spring roll actuator. Although in practical design radial pressure is less than the critical value, it still induces reduced overall area of elastomers in cross-section of actuator and thus less extensive force can be generated. This work proposed by employing fibers as skeleton to support prestrain elastomers and further to reduce the radial pressure of spring roll actuator. The results show improvement in extensive displacement of spring roll actuators made of composites upon actuation while compare to those made of neat VHB. Three different types of fiber (PA-I, PA-II, and PU) have been employed here and it is found the most preferred property of reinforcing fiber used in spring roll actuator is high rigidity. PA-I and PA-II monofilaments with higher modulus, their composites fabricated actuators show the larger displacements. This attributes to the better supporting effect of the fibers. Interestingly, PU monofilaments, with the lowest modulus among these fibers, improved displacement of VHB spring roll actuator to some extent. This indicates relatively soft fibers as PU still help to against radial pressure that properly due to high anti-compression stress of fiber spirals fixed by elastomer.

In the end, aligned CNT sheets are also employed as electrodes in neat VHB spring roll actuator. Although CNTs with very high stiffness, thin layer of aligned CNT sheet is not strong enough to support contractive elastomers. Moreover, strong interlayer mechanical constraint
of elastomer film is observed in CNT sheets electrodes actuators since dry nature of CNT without lubricating function. Hence, there is no enhancement of actuation stroke observed in spring roll actuators with CNT sheets electrodes.

Both of analytic modeling and experiments results of these work demonstrate the approach to improve actuation performance of D-EAP spring roll actuators. The principle behind the improvements is significant useful for future design and tailoring spring toll or similar structured rolled actuators.
CHAPTER 7 Conclusion and Future Perspective

Aligned carbon nanotube (CNT) sheet electrodes for D-EAP actuators have been shown to vastly improve directional strain response originating from the mechanical anisotropy of the electrode material. In this novel approach, mechanical anisotropy, ratio of initial modulus in fiber direction to that in cross-fiber direction of CNT electroded VHB, ranged from 7.9 to 11.2. CNT sheets electroded VHB flat films actuator shown high directed linear actuation strain in cross-fiber direction of greater than 25% meanwhile almost zero strain in fiber direction at a relatively low electric field (120 V μm⁻¹). This demonstrates an easy route to generate anisotropic electrotechnical response without complex add-ons.

The morphology of CNT sheets has crucial influence on their mechanical properties and resultant actuator performance. Using microcombing and selective laser etching of aligned CNT sheets, the CNT fiber alignment was improved and the mechanical anisotropy increased significantly. This improvement resulted in almost pure unidirectional strain of 33% at a relatively moderate electric field. It demonstrates that deformation anisotropy of D-EAP actuators can be significantly improved by directional laser etching of the electrodes rather than microcombing alone. Laser ablation process, was effective in selectively and precisely removing CNT fibers form the D-EAP surface while maintaining electrical conductivity. Hence the process can be used to apply transparent electrodes on film surface.

Anisotropic D-EAP was then employed in spring roll actuator to investigated their directional mechanical and electromechanical properties. CNT electroded D-EAP spring roll actuators did not perform better than the same with carbon grease electrodes in terms of actuation strain.
However, spring roll actuators made of fiber reinforced VHB composites with carbon grease electrodes shown improvement in displacement (9.9%-11% strain in longitudinal direction at 5kV). Analytical model of the spring roll actuator demonstrate the role of hoop stress on the radial pressure from outside layers of elastomer towards inner layers. Fibers in the D-EAP laminate support prestrained elastomers films in the hoop direction and therefore reduce the radial compressive stresses. PA and PU fibers were used to demonstrates this function but they performed differently with higher actuator stroke of PA reinforced sample and lower stroke of PU reinforced ones. Among fiber’s properties, high stiffness is major desired parameter. There were two type of PA fibers with different diameters and slightly different moduli. The stiffer fiber (PA-I) with much smaller diameter (same for fiber volume fraction) reinforced spring roll actuator shown stable but slightly smaller strain than that of sample with lower modulus and larger diameter (PA-II). Both of analytical modeling and experimental results of this work demonstrate the approach to improve actuation performance of D-EAP spring roll actuators. The principle behind these improvements is significant useful for future design and tailoring spring roll or similar structured rolled actuators.

During this work, there are several drawbacks or limitations have been noticed. One of the major ones is poor stability of CNT sheets property that cannot be neglected. For example, with same synthesis parameters, although CNTs in some extent maintained the anisotropic structure, their areal density and forest height are not identical. These changes further influenced their mechanical property and resultant actuator’s property.

Based on this works, potential areas of research could be:
1. Study the electrical charge distribution on CNT sheet electrodes to further understand influence of CNT sheet spatial structures on their property as electrodes in D-EAPs.

2. Explore other means to improve CNT sheets’ alignment or uniformity for improved electrode performance. For example, using solvent to condense CNT sheets.

3. Develop optically transparent tunable electrical conductive surface based on laser etched aligned CNT sheets on a certain substrate.

4. Extend the application of aligned CNT sheet electrodes to other type of EAP actuator for anisotropic mechanical and actuation properties.

5. Further improve the analytic model in this work of spring roll actuator based on unidirectional fiber reinforced composites. For example, to integrate fiber’s properties in to the model.

6. Replace normal spring in core of spring roll actuator with a system to support the compressive radial stresses between layers.

In summary, this dissertation thoroughly investigated novel aligned CNT sheets as electrode used in D-EAP actuators in terms of morphological, mechanical, electrical and electrotechnical aspects. Also, this work demonstrated methods to flexibly tailor D-EAP actuators’ anisotropic deformation by altering alignments and entanglements of CNT sheets, including microcombing process and laser etching. Additionally, anisotropic D-EAPs realized by CNT sheet electrodes or unidirectional fibers reinforcement were explored in spring roll actuator. Results of this work will be very helpful for understanding and further designing spring roll actuator with improved performance.
REFERENCES


[165] S.-H. Park, K.-Y. Cho, S.S. Hwang, K.-Y. Baek, Selective Dispersion of Porphyrin Metal Complex into Styrene-(Ethylene-co-butylene)-Styrene Triblock Copolymer,


[282] X. Niu, H. Stoyanov, W. Hu, R. Leo, P. Brochu, Q. Pei, Synthesizing a new dielectric elastomer exhibiting large actuation strain and suppressed electromechanical instability


stretchable conductors and piezocapacitive strain gauges based on simple contact-

[370] A. Denneulin, J. Bras, F. Carcone, C. Neuman, A. Blayo, Impact of ink formulation on
carbon nanotube network organization within inkjet printed conductive films, Carbon.

[371] M. Ha, Y. Xia, A.A. Green, W. Zhang, M.J. Renn, C.H. Kim, M.C. Hersam, C.D. Frisbie,
Printed, Sub-3V Digital Circuits on Plastic from Aqueous Carbon Nanotube Inks, ACS

[372] D.S. Hecht, D. Thomas, L. Hu, C. Ladous, T. Lam, Y. Park, G. Irvin, P. Drzaic, Carbon-
nanotube film on plastic as transparent electrode for resistive touch screens, Journal of

[373] J.W. Elmer, O. Yaglioglu, R.D. Schaeffer, G. Kardos, O. Derkach, Direct patterning of
vertically aligned carbon nanotube arrays to 20 μm pitch using focused laser beam

doi:10.1088/0957-4484/14/4/305.

254


APPENDIX

Figure A1 Uniaxial tensile test sample configuration of flat film actuator

Figure A2 Schematic illustration of set-up for electrode electrical resistance test
Figure A3 Schematic illustration of set-up for actuation test

Figure A4 Blocking for test sample configuration of flat film actuator connecting to actuation set-up