DESIGN AND TESTING OF FIBER-CONSTRAINED ELECTROACTIVE POLYMER

BY

KUN HYUCK LEE

THESIS

Submitted in partial fulfillment of the requirements
for the degree of Master of Science in Mechanical Engineering
in the Graduate College of the
University of Illinois at Urbana-Champaign, 2015

Urbana, Illinois

Adviser:

Professor Sameh Tawfick
Abstract

Soft robotics take advantage of active materials to regenerate many complex motions, including low-force functions, such as camouflage, micro-grabbing, or agile locomotion of micro robot. For these applications, traditional actuators, like electric motors and combustion engines, are unnecessarily large and heavy. In contrast, electro-active polymers (EAP) such as electrostatically actuated dielectric elastomers (DE) are considered very suitable technologies for these applications. In particular, owing to their simplicity, high strain and energy densities, and low noise, DE’s are currently the most promising candidates for small scale biomimetic applications. In this study, new fiber-constrained DE designs are developed, in order to obtain controlled uniaxial actuation that suits soft robotics and small scale actuation. Also, micro-wrinkle instabilities that occur in fiber-constrained membranes are examined. The property of wrinkle defers by controlling the geometrical parameters and the amount of strain in a membrane. Therefore controlling the shape of wrinkle is applicable for micro-actuation. In addition, image analysis methodology is discussed and creative ways of analyzing the image data is established.
Acknowledgments

First and foremost, I would like to express my sincere gratitude to my advisor, as well as my life mentor, Prof. Sameh Tawfick for his immeasurable support and encouragement, for his insightful guidance and immense knowledge, and for his genuine concerns for my success in my career throughout my Master’s degree. Without Sam’s supervision, I would not have been able to develop my creativity, perseverance, and many skillsets that are necessary for an engineer.

Secondly, I also would like to thank my peers in Kinetic Material Group: Jonathan Bunyan, PingJu Chen, and Kaihao Zhang for helping me to figure out the solutions to many of problems that I interfere and stuck on.

Last but definitely not least, I would like to thank my beloved family for being supportive of my decisions and teaching me integrity.
# TABLE OF CONTENTS

## CHAPTER 1 INTRODUCTION

1.1 Motivation ...................................................................................................................... 1

1.2 Dielectric Elastomer ........................................................................................................ 2

1.3 Figures and Tables .......................................................................................................... 3

## CHAPTER 2 LITERATURE REVIEW

2.1 Background ...................................................................................................................... 5

2.2 Applications of DE .......................................................................................................... 6

2.3 Problem Statement ......................................................................................................... 7

2.4 Fiber-constrained DE actuators ....................................................................................... 7

2.4.1 Analytical modeling of DE actuation ......................................................................... 8

2.5 Wrinkling instabilities in thin membranes ...................................................................... 9

2.5.1 Analytical modeling of wrinkling instability in DE .................................................... 10

2.6 Figures .......................................................................................................................... 16

## CHAPTER 3 ACTUATION of FIBER-CONSTRAINED DE

3.1 Introduction .................................................................................................................... 19

3.2 Experimental Methods .................................................................................................. 19

3.2.1 DE Membrane without Constraints .......................................................................... 19

3.2.2 DE Membrane with Fiber Constraints ...................................................................... 21

3.3 Results and Analysis ..................................................................................................... 25

3.3.1 DE Membrane without Constraints .......................................................................... 25

3.3.2 DE Membrane with Fiber Constraints ...................................................................... 26

3.4 FEA Modeling and Analysis ......................................................................................... 31

3.5 Figures and Tables ......................................................................................................... 34

## CHAPTER 4 ELECTROMECHANICAL WRINKLING INSTABILITY IN DE

4.1 Introduction .................................................................................................................... 55

4.2 Experimental Methods .................................................................................................. 55

4.3 Results and Analysis ..................................................................................................... 56

4.4 Figures .......................................................................................................................... 58

## CHAPTER 5 CONCLUSION AND OUTLOOK

5.1 Actuation of Fiber-Constrained DE ............................................................................... 60
CHAPTER 1 INTRODUCTION

1.1 Motivation

For the past decades, there has been much development in new active materials to create effective polymers that can mimic the actuation mechanisms in nature. Polymer, especially elastomer suits application like biomimetic, due to its flexibility and high energy storage capacity. Elastomer, also known as rubber, is composed of highly cross-linked chains that allow the material to store energy and make materials reversible.

There are many different polymers and other materials that can actuate and generate deformation, mainly using electronic and ionic behavior of the materials. In electronic actuation, the materials employ electrostatic interaction between flexible electrodes, causing mechanical pressure on a sandwiched thin membrane to create an actuation. This kind of actuator is known as Dielectric Elastomer (DE). Other kinds of electrically actuated muscles include Electrostrictive Relaxor Ferroelectric Polymer, Electrostrictive Graft Elastomers, Electrolytes and Electrostatic Repulsion, and Liquid Crystal Elastomers. Similarly, ionic actuation drives mobile ions in the polymer using electric field. They include CNT actuator, Conductive polymer, and Ionic Polymer-Metal Composites (IPMCs). Another material that is also commonly used for actuation is Shape Memory Alloy (SMA), actuate by thermal energy.

Each actuator has its own advantages and disadvantages (Figure 1). For example, Electrostrictive Relaxor Ferroelectric Polymer can actuate with high rate and stress, but can only obtain small strain value. Conductive polymer gives high stress and strain, but has low life cycle. SMA gives good strain value, but there is huge energy loss. Evidently, there are many options to select from, but one needs to know what aspect of the actuator system one is willing to sacrifice.
Likewise, DE also has both pros and cons. However, for biomimetic application, even though DE requires relatively large amount of voltage to operate, it is determined to be the most suitable material (Table 1) owing to its high strain and energy density, short response time and high efficiency that are necessary to mimic the natural muscle, it is determined to be an ideal material for mimicking many dynamical systems from nature.

1.2 Dielectric Elastomer

DE, like other electronic actuators, utilizes electric field. DE is usually a thin membrane sandwiched between two oppositely charged compliant electrodes. When the voltage is applied, these electrodes generate electrostatic pressure to make compressional force that results the sheet area to expand as its thickness reduces (Figure 2). Most commonly used compliant electrodes are ionic solution and carbon grease. There has been much work on developing the compliant electrodes. For example, Whiteside’s group has developed ionic hydrogel. It means that DE can actuate with any type of electrodes as long as they do not resist its expansion with their stiffness. The choice of electrodes is important, since DE performs better (i.e. high strain with sufficient blocked stress) when the electrodes have good conductivity and enough compliance.

Since DE is a polymeric material, creating composite is an option to mechanically modify its actuation pattern. DE composite can utilize other materials as reinforcements, and this allows it to exploit large range of material properties. For example, by having aligned CNTs as reinforcing materials and depositing them systematically, we can take advantage of high stiffness of CNT and elastic property of DE elastomer (Figure 3).
1.3 Figures and Tables

Figure 1. Comparisons between each actuator technology$^2$

Figure 2. DE actuation and its mechanics$^4$
Figure 3. Display DE-CNT composite’s performance relative to other materials

Table 1. Comparison of actuators and natural muscle

<table>
<thead>
<tr>
<th>Actuator</th>
<th>Strain</th>
<th>Actuation Pressure</th>
<th>Density</th>
<th>Efficiency</th>
<th>Response Rate</th>
</tr>
</thead>
<tbody>
<tr>
<td>Natural Muscle</td>
<td>O</td>
<td>O</td>
<td>O</td>
<td>O</td>
<td>O</td>
</tr>
<tr>
<td>Electromagnetic</td>
<td>O</td>
<td>O</td>
<td>X</td>
<td>O</td>
<td>X</td>
</tr>
<tr>
<td>Piezoelectric</td>
<td>X</td>
<td>O</td>
<td>Δ</td>
<td>X</td>
<td>X</td>
</tr>
<tr>
<td>Shape Memory Alloy</td>
<td>Δ</td>
<td>O</td>
<td>Δ</td>
<td>X</td>
<td>X</td>
</tr>
<tr>
<td>Magnetostrictive</td>
<td>X</td>
<td>O</td>
<td>X</td>
<td>O</td>
<td>X</td>
</tr>
<tr>
<td>Electrostatic</td>
<td>O</td>
<td>X</td>
<td>O</td>
<td>O</td>
<td>O</td>
</tr>
<tr>
<td>Dielectric Elastomer</td>
<td>O</td>
<td>O</td>
<td>O</td>
<td>O</td>
<td>O</td>
</tr>
<tr>
<td><strong>Good</strong></td>
<td>O</td>
<td><strong>Fair</strong></td>
<td>Δ</td>
<td><strong>Poor</strong></td>
<td>X</td>
</tr>
</tbody>
</table>
CHAPTER 2  LITERATURE REVIEW

2.1  Background

The general equation describing the behavior of a DE related the polymer stretching and polarization to Helmholtz free energy as noted below \(^7\).

\[
\sigma_i + \epsilon E^2 = \frac{\mu^\alpha (\lambda_i^2 - \lambda_i^{-2} \lambda_j^{-2})}{1 - (\lambda_i^2 + \lambda_j^2 + \lambda_i^{-2} \lambda_j^{-2} - 3)/J_{lim}^\alpha} + \frac{\mu^\beta (\lambda_i^2 \xi_i^2 - \xi_i^{-2} \lambda_i^{-2} \lambda_j^{-2})}{1 - (\lambda_i^2 \xi_i^{-2} + \lambda_j^2 \xi_j^{-2} + \xi_i^{-2} \lambda_i^{-2} \lambda_j^{-2} - 3)/J_{lim}^\beta} \quad (1)
\]

\forall \text{ integers } i, j \in [1, 2]

Each subscript, \(i\) and \(j\) represent sheet plane coordinate system, \(x\) and \(y\) (Figure 2). First term on the left hand side of the equation, \(\sigma\), represents the amount of stress in the sheet plane, \(x\) and \(y\). The stress depends on the amount of force applied on the sides of membrane that have \(x\) and \(y\) as their normal vectors. Second term on the left hand side of equation represents Maxwell stress, where \(\epsilon\) is permittivity of dielectric elastomer and \(E\) is electric field. The field can be calculated using the amount of applied voltage and the thickness of membrane.

On the right hand side of the equation, it represents the behavior of viscoelastic relaxation, using rheological model that integrates Voigt model (a spring and a damper in parallel) and Maxwell model (a spring and a damper in series) (Figure 4) \(^7\).

Each superscript, \(\alpha\) and \(\beta\) represent different springs in the model. \(\mu\) represents the shear modulus of a spring, and \(J_{lim}\) represents spring extension limit. \(\lambda\) represents the total amount of stretch of the system (i.e. final length over initial length). Lastly, \(\xi\) represents the amount of stretch in the dashpot. When the model goes under constant strain, coming from tensile load, spring will react immediately, and then the dashpots will respond to both internal and external stresses in delayed manner.
2.2 Applications of DE

As defined from the ideal DE model (eq. 1), voltage, stretching force, and permittivity of the material play important role in determining the amount of actuation. Many studies have demonstrated the effect of high permittivity in large deformation. For example, one integrates colloids with high permittivity with DE. As the DE composite contains more of these colloids, the amount of actuation increases until when the stiffness of colloids becomes dominant.\(^8\)

Another parameter, voltage, or electric field, is more intuitive contribution to DE actuation, since the actuation is literally driven by the compressional stress generated from electrostatic pressure. However, the force may appear to be redundant component, and is not independently contributing to the amount of deformation. The intuition is that the force depends on both mechanical property of the material and the amount of applied voltage. However, contradicting with the argument, force, voltage and material properties, like stiffness, are appeared as a separate and independent terms in eq. 1. This can be explained with DE’s unique property.

DE can generate larger amount of actuation with faster response time relative to same amount of voltage, when the material is prestretched: mechanically stretched in biaxial direction before the actuation. This is due to the reduction in its thickness that results much larger electrostatic pressure with the same amount of voltage. Also, the materials are in favor of expansion in sheet plane direction since it is under tension. This characteristic of DE proves that the stress, or the force, is needed to be an independent component.\(^5\)

There are many applications already those benefit from DE’s unique nature. There have been many robotic implementations including FLEX and Skitter. For both robots, they use rolled
DE and utilize its uniaxial actuations to generate the movement of their robots\textsuperscript{9,10}. As it is observed, by applying DE in creative way, one can obtain simple and efficient actuation.

\textbf{2.3 Problem Statement}

These studies show many unique characteristics of DE that makes it convenient to handle, in both theory and experiment. Yet, there are challenges still remaining to be solved. One of the main issues is controlling aspect of the actuator. Problem with the control is unavoidable due to resilient nature of the material. Its nonlinear electromechanical behavior ultimately results instability of the system.

As shown, for the membranes with 1.5 and 2 prestretching values, once after they reach their local maxima, the curves descend (Figure 5)\textsuperscript{4}. This indicates the instability of material. In this phase, the material expands in much faster rate, resulting buckles in the membrane. It is also shown that, as the amount of prestretching increases, instability may not occur. However, in this case, the sample breaks down with relatively smaller amount of strain. The study also shows that the state of instability can change as the stiffness of electrode differs. That is, as the electrodes get stiffer, the instability becomes more extreme\textsuperscript{4}. This results the development of instability, even for the samples with high prestretching values.

In this thesis, many different methods are developed to augment DE, in order to reduce the amount of effort that it takes to control the actuation. Then, the instability in DE is examined by inspecting the periodic wrinkles in the material.

\textbf{2.4 Fiber-constrained DE actuators}

As shown in the eq. 1, the actuator system exhibits in plane symmetry, and therefore, when DE operates, it shows strong isotropy. This is not ideal for an actuator. Actuation with
controlled directionality is required for many applications, especially in robotics \(^9,10\). To provide the directionality in DE actuation (i.e. anisotropic deformation along one axis), many novel approaches are taken, which include unevenly prestretching the material \(^11\), rolling the material to infinitely constrain the radial expansion \(^10,12\), and adding the aligned fibers to the film with prestretch in biaxial direction \(^8,13\).

Unevenly prestretched actuator is expected to give anisotropic actuation by expanding larger in more prestretched direction. However, from the experiment, it is shown that the amount of deformation is significantly smaller than when the actuator is prestretched biaxially. In addition, it does not even generate significant anisotropy \(^11\). There are two types of rolled tubular actuator, one with spring core and one without. The one with the spring gives significant anisotropic actuation. However, due to the high shear stress in the interface in between the core and the material, it fails after short amount of time. On the other hand, coreless tubular actuator gives good durability, but it only generates small amount of actuation \(^10,12\).

Recently, many have implemented fiber reinforcement DE actuator. This anisotropic DE (ADE) allows large anisotropic actuation, and with the additional magnitude of permittivity coming from the fiber, the magnitude for its allowable deformation increases. The fiber is usually applied to ADE in two different ways: aligned fibers on both surfaces of the membrane, or aligned fibers sandwiched between two DE membranes \(^8,13\). Despite the constraint of fiber, bounding the direction of actuation, still large amount of actuations are observed in both directions when a high voltage is applied.

### 2.4.1 Analytical modeling of DE actuation

Using eq. (1), ideal DE uniaxial actuation in relation to stretching and polarization of the elastomer is derived as
\[ \sigma_1 + \varepsilon E^2 = \frac{\mu^\alpha (\lambda^2 - \lambda^{-1})}{1 - \frac{\lambda^2}{j_{\text{lim}}} + 2 \frac{1}{\lambda^{-1} - 3}} + \frac{\mu^\beta (\lambda^2 \xi^{-2} - \lambda^{-1} \xi^1)}{1 - \frac{\lambda^2 \xi^{-2}}{j_{\text{lim}}} + 2 \frac{1}{\lambda^{-1} \xi^1 - 3}} \] (2)

Here, only one directional component is considered since, in ideal condition, the material is expected to be quasi-static in the direction it is constrained (Figure 6). Both stretching and polarization of the material only affect the actuation in unconstrained direction. Figure 7 compares the amount of actuation in constrained direction and unconstrained direction when DE composite has anisotropic stiffness.

As it is viscoelastic material, the hysteresis plays big role in the actuation as much as polarization, prestretching, and constrains. Therefore, we also need to understand the past condition of the material as much as the current condition. The past conditions include the way that membrane is manufactured and packaged, and how much of actuation cycles that the membrane has went through\(^{14}\). This is important to know to generate accurate control over the actuation.

### 2.5 Wrinkling instabilities in thin membranes

As discussed, for a dielectric elastomer (DE), polymer chain expands when there is electrostatic field applied across the DE film. This enhances the compressional force on the surface of membrane and dielectric polarization with in the film. Thus, the film expands in sheet plane direction. However, in my experiments, I observed as will be discussed later, that when the film is constrained with aligned fibers, it restricts the film to expand in uniaxial direction. This replicates the uniaxial tension of a thin membrane as mentioned above. When the electroactive elastomer reaches certain amount of expansion, it, indeed, generates wrinkles in the membrane.

In mechanical system, buckling is identified as a deformation state that comes from instability of materials. The mode varies according to boundary conditions, and also the rate that
stress is applied to specimen. Euler buckling mode differentiates each mode by the specimen’s boundary condition. There are three different boundary conditions. They include no degree of freedom, one degree of freedom with a rotational motion (or a translational motion) perpendicular to the compressive stress, and all degrees of freedom. Each mode generates different characteristics in buckling.

Buckling also depends on the rate that compressional stress is applied. By changing the rate that the stress is applied, the number of nodes in buckling changes. If the rate is fast enough, we can obtain buckling profile with higher harmonic.

We can interpret this behavior using thermodynamics. Buckling can be classified in two types of thermodynamic system, and they are isothermal and adiabatic buckling. In case of high thermal conductivity of the material, or when the material is isothermal system, the buckling may happen slowly. On the other hand, when the system is adiabatic, the buckling would happen soon after the stress is applied. Basically, the state of energy diffusivity plays important role in buckling.

2.5.1 Analytical modeling of wrinkling instability in DE

There have been many researches related to controlling the wrinkles, or instability in deformation. In other words, we allow instability to happen, but in organized way. One of the studies explores a thin membrane by simply pulling it. This induces interesting behavior, periodic wrinkles. In this study, parameters that influence the characteristics of wrinkles, such as geometry and strain is discovered.

Of the membrane with thickness $t$, width $W$, length $L$, Young’s modulus $E$ and Poisson’s ratio $\nu$ where $t << W < L$, periodic wrinkle appears in the membrane when the strain value, $\gamma$ exceeds $\gamma_c$. This membrane is fixed by two rigid parallel clamps that pull the membrane in
tension and resulting compression in the midway of membrane. Due to the rigidity in the clamps, there are no wrinkles near the clamps (Figure 8) \(^{16}\). The relation between characteristics of wrinkle, such as the wavelength, \(\lambda\) and amplitude, \(A\), and condition of membrane is evaluated using energy balance \(^{16}\).

There are two dominant energy components when it comes to wrinkling. One is bending energy and another is stretching energy. They are defined as

\[
U_B = Et^3 \left(\frac{A}{\lambda^2}\right)^2 LW, \quad \left(\frac{A}{\lambda}\right)^2 \approx \nu \gamma
\]

which makes the energy equal to

\[
U = \left(\frac{Et^3}{\lambda^3} + \frac{E\nu \gamma \lambda^2}{L^2}\right) \nu \gamma LW
\]

According to thermodynamic, Free energy always converges to minimum. As results we find

\[
\lambda = \frac{(tL)^{\frac{1}{2}}}{\gamma^{\frac{1}{4}}}
\]

\[
A = (\nu tL)^{\frac{1}{2}} \gamma^{\frac{1}{4}}
\]
Another study shows control of wrinkle formation and pattern using thermal energy instead of mechanical energy \(^{17}\). On a glass substrate, 1cm thick PDMS is deposited. Using electron beam evaporator (E-beam), thin layer (~5nm) of titanium or chromium is deposited as an adhesion interlayer. Then, 50nm thick gold layer is deposited. Due to the high temperature of deposited metals, PDMS expands during the deposition process. When the specimen is taken out of E-beam chamber, the periodic waves are formed on the top surface of the specimen as the specimen cools down. Whenever PDMS goes under thermal expansion, the wave form disappears, and then regains when it shrinks back when it cools down. The amount of compressive stress, or compressive energy per volume, before the film goes under buckling during the deposition can be written as,

\[
\sigma_o = \frac{E_m(\alpha_p - \alpha_m)(T_D - T)}{1 - \nu_m}
\]

where subscripts p and m stand for PDMS and metal. \(\nu\) stands for Poisson’s ratio, \(\alpha\) stands for coefficient of thermal expansion, and \(E\) stands for Young’s modulus. Here, the energy gradient between each layer is in equilibrium, and no wrinkles are formed.

When the specimen is cooled down, compressive stress starts to increase, and starts to form buckling at \(\sigma_{\text{crit}}\).

\[
\sigma_{\text{crit}} \approx 0.52 \left(\frac{E_m}{1 - \nu_m^2}\right)^{\frac{1}{3}} \left(\frac{E_p}{1 - \nu_p^2}\right)^{\frac{2}{3}}
\]

Also the wave that it produces will have wavelength of \(L\),

\[
L \approx 4.36t \left(\frac{E_m(1 - \nu_p^2)}{E_p(1 - \nu_m^2)}\right)^{\frac{1}{3}} \approx 4.4t \left(\frac{E_m}{E_p}\right)^{\frac{1}{3}} \approx 61t
\]

where \(t\) is the film thickness.
These representations are in the case of uniform or equi-biaxial condition, and therefore, the waves are periodic locally, but when the scope gets larger, the waves are in unorganized form.

More organized periodic waves are formed by casting PDMS layer on a substrate with patterns, made out of photoresist. When we have the patterned PDMS, the condition won’t be uniform throughout the film. Instead, due to difference in the boundary condition, owing to variation of the thickness, we will observe different trend of deformation.

For instance, when we have periodic square wave patterns (steps) on the PDMS surface with variation of its thickness, we will observe more organized periodic wrinkles. In the cooling process, the compressional stress needs to be relieved by deformation. However, due to higher stiffness in the step region, the stress builds up in the direction that the steps are oriented. Resisting the deformation, unrelieved stress forms the wrinkles, angled in perpendicular to the orientation of steps, or unrelieved compressional stress (Figure 9).

As mentioned, the amount of compressive stress is not uniform. In fact, the stress is different depending on the orientation:

\[
\sigma_x = -\sigma_o \left[ 1 - \frac{\cosh \left( \frac{x}{L} \right)}{\cosh \left( \frac{d}{T} \right)} \right] \tag{11}
\]

\[
\sigma_y = -\sigma_o \left[ 1 - \nu_m \frac{\cosh \left( \frac{x}{T} \right)}{\cosh \left( \frac{d}{T} \right)} \right] \tag{12}
\]

where \( L \) is transition length, characterizing the distribution of stress from the step to the remote smooth film, and \( d \) is the half of the width of a step.

Here, we could interpret that metal film as adiabatic system that the conformation of intermolecular structure is not submissive to the external stress. Also, we can look at PDMS as
isothermal system that balances the energy difference by changing its number of allowable conformation states. For the region where PDMS thickness is relatively large, also known as steps, we can also interpret this local condition as isothermal dominant system. In the region where isothermal system is more pronounced, or PDMS is thicker, the external energy may dissipate more stably.

This study also talks about changing in stiffness of PDMS caused by thermal effect, and how it is related to the formation of wrinkles. As the modified PDMS (stiffer) layer gets thicker, the wavelength of buckling is found to be increasing, and this finding leads to the following study. In this study, it demonstrates the interaction between soft and stiff polymer layer and its effect in the buckling.

To generate the variation of stiffness in a single PDMS, photolithography technique is employed. A mask with a pattern is used to regulate the exposure of PDMS to UV. The area that is exposed to UV light stiffens. Once the stiffening process is finished, it goes through the same process as described in the previous experiment. During the process of cooling, the section with lower stiffness buckles first. In this area, the wrinkles align parallel to the boundary between different stiffness. This is because the softer area is more submissive to the deformation, and therefore the unexposed area goes under unstable deformation first. Alignment of the buckles is in such way since the stiffer parts of the PDMS resist compressive stress along the direction that strips is extended. When the stress in perpendicular direction to the boundary is relieved by forming the wrinkles in unexposed area, the remained stress in parallel direction to the boundary is working its way to relieve its stress by generating wrinkles in the exposed area, aligned in perpendicular direction to the boundary (Figure 10).
This behavior can be understood more easily by identifying the differences in buckling stiffness of the exposed and unexposed area as expressed in the eq. 13.

\[ \bar{B}_i = \left( \frac{E_m t_m^3}{12(1 - \nu_m^2)} \right) \left[ 4 \left( E_R t_R^3 + ((1 + t_R) - t_R^3) \right) \right. \]
\[ \left. - \frac{3 \left( E_R t_R^2 + ((1 + t_R) - t_R^3) \right)^2}{E_R t_R + 1} \right] \]

where \( i \) represents the specific area of PDMS (exposed or unexposed), and subscript R represents the ratio of the parameter (\( i \) to \( m \)).

Again this can be explained using thermodynamic. In Figure 11, of the UV exposed area, the compressive stress in y direction is higher than x. The energy in y direction is more dominant. It makes the conformation change in y direction to also dominate, since the change in enthalpy has to be balanced by change in entropy, and vice versa to the unexposed area.

These studies form the basic theoretical framework that I will use to interpret the wrinkling instability in fiber constrained DE discussed in Chapter 4.
2.6 Figures

Figure 4 Rheological model of DE

Figure 5. Actuation of DE with compliant electrodes (blue) and breakdown voltage (red)

Figure 6. DE with well aligned equally spaced fiber-constraints
Figure 7 DE composite with hydrogel composite electrode (HG) integrated with carbon nanotubes (CNT) (20% CNT/ 80% HG). DE is 300μm with 50kPa shear modulus and HG composite is 100μm with 1200kPa shear modulus of CNT and 300kPa shear modulus of HG. Both actuation in axial direction of the aligned CNT (solid) and actuation in transversal direction of the aligned CNT (dashed) are shown in the graph.

Figure 8 Film under tensile load using two rigid clamps

Figure 9 Organized wrinkles and PDMS with steps
Figure 10 UV Patterned PDMS and its wrinkles

Figure 11. Energy state of PDMS in each area
CHAPTER 3 ACTUATION of FIBER-CONSTRAINED DE

3.1 Introduction

We took many different approaches to generate uniaxial actuation, exploring from the most basic DE membranes to fiber reinforced DE composites. Different parameters, including dimension and orientation of the constraints, and number of the actuation cycle, are tested to understand their relationship with different actuation systems. Most importantly, for the first time, we demonstrate self-directed micro scale actuation of DE that contracts perpendicular to the direction it expands, resulting actuation with large anisotropy.

3.2 Experimental Methods

3.2.1 DE Membrane without Constraints

Effect of Prestretching in Anisotropic Actuation

An attempt is made to generate uniaxial actuation by prestretching the material in a single direction. For this experiment, an actuator prepping stage is made (Figure 12). Two translational stages (Figure 12c) that translate in opposite direction are used for stretching the membrane by pulling on the edges of the membrane (Figure 13). First, the maximum strain value for 1x1 inch DE membrane (3M VHB 4946) in a single direction is measured by stretching the membrane until it fails (Figure 12c). Once the maximum strain value is determined which is found to be 500\%, DE is prestretched in single direction, near to its maximum strain value, but slightly under. Then, carbon grease is applied on both sides of the membrane (Figure 13). 10kV High Voltage Supplier (SRS PS365) is employed to generate electrostatic pressure (Figure 12d), and conductive thread (Digi-Key Conductive Thread) is used to makes connection between the high voltage cables and the compliant electrodes. The voltage is applied until it fails, and as the
voltage is applied, its behavior is recorded using the handheld digital microscope (Celestron 44302-B Digital Microscope Deluxe) (Figure 12a).

Due to its viscoelasticity, the polymer is influenced by its stress, or strain history. This phenomenon is also known as hysteresis. To examine its nature of hysteresis, various combinations of prestretching are tested. This experiment uses the new prepping stage that enables biaxial prestretching of the membrane in both x and y direction (Figure 14). Similar to the previous stage, each edges of the square DE membrane (same as previous experiment) are clamped onto two fixed stages and two translational stages (Figure 14a, d). Translational stages stretch the membrane in biaxial direction. Carbon grease is applied to each side of the membrane and conductive thread is used to transfer electricity from the high voltage supplier to the compliant electrodes. This set up is utilized for the next few experiments.

Since the VHB tapes that are used in the experiments come as roll, an assumption is made that there will be intrinsic prestretch that may have occurred during the process of rolling. First experiment is done on unevenly prestretched membranes to learn the relationship between the actuation directionality and the amount of prestretch, in parallel with the effect of intrinsic prestretch value. In addition, tapes with different thicknesses are used to study the effect of field intensity to their actuations.

Second experiment is done by actuating an actuator multiple times and examining how an actuation of a membrane influences its next actuation.

5kV and 500V are applied to 1.016mm thick membranes (3M VHB 4946) and 260μm thin membrane (3M VHB F9460PC), accordingly. For the second experiment, simply, the same voltage 5kV is applied to same membrane. Each time the voltage is applied, it is held for a minute, and then brought back down to zero.
3.2.2 DE Membrane with Fiber Constraints

I developed a novel technique to deposit aligns microfibers with uniform micron-level spacing between each fiber. This process also allows these microfibers to be printed on both sides of a pre-stretched and suspended 3M VHB DE membrane (Figure 15 and Figure 16). This technique is used to generate DE composites with aligned Fine Conductive Wire (FCW) and High Stiffness Micro Fibers (HSMF) as reinforcement.

a) DE Membrane with Fiber Constraints: FCW

*Top: FCW/ Bottom: Carbon Grease*

A set of equally spaced (150 µm) and well aligned FCW (25.4 µm diameter copper wire) is applied onto the three times isotropically prestretched DE membrane (3M VHB F9460PC). Here, FCW is used as both constraining media to restrict DE actuation in its axial direction, and conductive electrodes to generate the electrostatic pressure. To prevent from the misalignment issue from having the conductive wires on both surfaces of the membrane, FCW is only applied on a surface and compliant electrode (Digi-Key carbon grease) is applied on the opposite surface (Figure 17).

*Top: FCW with Ionic Liquid/ Bottom: FCW with Ionic Liquid*

Two sets of FCW (18µm diameter stainless steel strand from the conductive thread: Digi-Key) are applied onto the three times isotropically prestretched DE membrane (3M VHB F9460PC). FCW are not well aligned as the previous experiment with copper wire, since the experiment was done prior to the development of the deposition technique. Here, FCW is used as constraining media to restrict DE actuation in its axial direction, and ionic solution (Sigma
Aldrich 1-Decyl-3-methylimidazolium chloride) is used as conductive electrodes that generate the electrostatic pressure (Figure 18).

Here, both directionality of the actuation, and the effects of an actuation to next actuation is examined. 500V is applied for each cycle. Each time the voltage is applied, it is held for a minute, and then brought back down to zero.

**Top: FCW with Ionic Liquid/ Bottom: Ionic Liquid**

Same experiment as the “Top: FCW with Ionic Liquid/ Bottom: FCW with Ionic Liquid” is practiced. However, for the sample used in this experiment, only one surface of the membrane has a set of FCW with the ionic solution (Figure 19).

**b) DE Membrane with Fiber Constraints: HSMF**

There have been many issues with different types of FCWs. Very fine copper wire has very low stiffness, and therefore it cannot withstand the amount of tension that is applied when it is placed on the troughs of aligning mold. On the other hand, stainless steel conductive threads are stiff enough, but due to their elasticity and twisted conformation of the single strand, it is hard for them to stay aligned when they are deposited on to the membrane. Plus, the conductivity of the wire is not utilized well. Therefore, replacement for the FCW was needed and micro-polyethylene fiber, or the HSMF (Berkley NanoFil), is chosen.

For the HSMF composite experiments, to minimize unbiased actuation condition caused by uneven prestretch (Figure 14), new technique of prestretching is introduced.

A two inch diameter circle with backing tape is used to trace the circular pattern on the DE membrane. Once the circular pattern is traced, the two inch diameter circle is remained to adhere
to the DE membrane temporarily. Then, they are fixed to the table using very thin square frame. Four corners of the membrane are overhanging (not adhered to anywhere). Each of the corners is mechanically pulled and adhered to the six inch ring (inner diameter). When the corners are adhered to the ring, the traced line is matched to the inner diameter of ring while the tape is still adhered to the thin fixture and the circle. Once all four corners are firmly adhered to the ring, DE membranes are pulled carefully and detached from the thin fixture. Lastly, the location of electrode is marked by red ink pen (Figure 20). A demonstration of this procedure can be found from ‘soft robotic toolkit’ webpage (http://softroboticstoolkit.com/book/dea-step-1).

Top: HSMF with Carbon Grease / Bottom: Carbon Grease

Since the stored energy of capacitor is related to its capacitance which is proportional to the area of electrode, assumption is made that the area, or spacing between each fiber, may have an effect to DE actuation. To observe the effect of fiber spacing, three different values of the uniform spacing between a set of HSMFs are deposited on three different samples accordingly. Window sizes of the electrodes for each sample are kept constant.

Different spacing values (75µm, 150µm, 300µm) are chosen. 150µm spacing is arranged by winding HSMF on the mold that has aligned struts with 150µm distance apart from each other (Figure 15b). 300µm spacing is arranged by winding HSMF every other strut of the same mold. 75µm spacing is created by using the wire deposition technology presented above (Figure 15, and Figure 16). One set of HSMF with 150µm spacing is deposited, then the microscope module is used to deposit the second set in between the first set of HSMF.

A set of well-aligned HSMF is deposited on one side of the three times isotropically prestretched membrane (3M VHB F9460PC), and then carbon grease is applied on both sides
within the window drawn at the end of prestretching process (Figure 20). Carbon grease sandwiches a HSMF set and a membrane. 1.5kV is applied for each sample.

**Assembly**

Two sets of HSMF are placed next to each other with their preferred actuation direction to be perpendicular to each other. Here, the interactions between two devices are observed as the distance between the devices are changed.

**Micro-architecture Electro- Elasto- Kinematic [MEEK]**

A compositied with two sets of well-aligned, equally spaced HSMFs are deposited, and sandwiched the four times prestretched DE membrane (3M Corporation’s VHB 4910), with a bias angle between each set, using the techniques that are explained in Figure 15 and Figure 20.

The geometry of fiber sets is similar to what is seen in pneumatic McKibben muscles. McKibben actuator uses braided sleeve with stiff fibers in helices in opposite directions with a bias angle. McKibben actuator creates a coupled motion where it expends in one direction and compress in the transversal direction. The expansion lies on the minor axis (obtuse angle) and contraction lies on the major axis (acute angle) of a rhombus, which will eventually make the difference of two angles smaller. Ionic liquid (Sigma Aldrich’s 1-Decyl-3-methylimidazolium chloride) is used as a transparent conductive electrode (Figure 22). This actuator is hence partially transparent except for the parameters of rhombus of the HSMF (Figure 23).

This actuator is designated as Micro-architecture Electro- Elasto- Kinematic [MEEK] muscles. The effect of bias angle on the actuation of an individual rhombus is examined (Figure 23). The characteristics of actuation for each sample with different bias angles at every 1kV
interval, and up to ~7kV are evaluated. The trend of actuation and efficiency of the actuation is studied as the bias angle changes.

3.3 Results and Analysis

3.3.1 DE Membrane without Constraints

No significant observation is made from the membrane with extreme uniaxial prestretching, even after raising the voltage until it failed. This can be explained by the limits of thickness reduction for the cases of uniaxial prestretch and biaxial prestretch. In order for uniaxially prestretched membrane to achieve the same amount of thickness reduction of biaxially stretched membrane, the membrane has to exceed its strain limit of a uniaxial tension. In other words, uniaxial prestretch has lower limit of thickness reduction than it of biaxial prestretch, since Poisson effect is applied to both height and width when the membrane is stretched in single direction which also enhances a compressive stress that works against the actuation of the membrane in transverse direction of the uniaxial tensile load. No measurements are made from the experiments since the observation did not show any significance.

Interesting correlations are observed from the experiment with the biaxial prestretching. The assumption was correct. As it is shown from Table 2 and Figure 24, the material prefers to expend more in the “rolled direction” (tangent to the tape role), even when the amount of prestretching is much larger in the “unrolled direction.” This is due to intrinsic strain coming from the packaging process. In order to make a roll of tape, the tape goes under small tension is applied in tangential direction to its roll. This enhances the polymer chain to orient in the direction.
Also, the effect of an actuation to the next actuation cycle is observed. The amount of the actuation is consistent for each cycle (Table 3). However, we see that initial area and actuated area gets larger as the number of actuation cycle increases (Figure 25). One of the factors that cause change in the area is from the migration of compliant electrodes. They provide small resistance to the membrane when it is retracted from the actuation. Another factor is the relaxation of polymer chains after each actuation. The chains find new conformation that gives new minimum free energy state each time it is under stress, or strain.

3.3.2 DE Membrane with Fiber Constraints

a) DE Membrane with Fiber Constraints: FCW

Top: FCW/ Bottom: Carbon Grease

Due to discontinuity in the electrodes, only negligible amount of local deformations around the FCW were observed. Therefore, not much of overall actuation was unobserved, and no measurements are made from the experiments since the observation did not show any significance (Figure 26).

Top: FCW with Ionic Liquid/ Bottom: FCW with Ionic Liquid

Visual deformations occurred for each actuation cycle, but not much of anisotropy in the actuations was observed. It is due to poor alignment of FCW which resists the actuation in multiple directions. This resistance can withstand the tension coming from the membrane expansion until the adhesion between FCW and the membrane fails. From then, the membrane will act as any other unconstrained DE membrane. When the fibers are not well-aligned as well as the prior experiment: Top: FCW/ Bottom: Carbon Grease and distribution of the fibers is low, these fibers will be detached from the membrane very easily in early phase of the actuation, since
the resistance is not strong enough to oppose the expansion of membrane. This experiment was done prior to the development of fiber deposition technique, and therefore, the control of fiber deposition was challenging.

However, significant effect of an actuation on the following actuation cycle is observed. It is observed that the initial area and actuated area get larger as the cycle number increases (Figure 27, and Figure 28). One of the factors that cause change in the area is, again, from the migration of compliant electrodes. The compliant electrodes and FCW provide small resistance to the membrane when it is retracted from the actuation. Relaxation of polymer chains after each actuation is also speculated as a factor to the behavior. The chains find new conformation that gives new minimum free energy state each time it is under stress, or strain.

As it is observed from Figure 27, and Figure 28, here, uniform increments and decrements for each actuation and retraction are observed for each cycle in both x and y direction. Figure 28 shows that every time when the voltage is withdrawn, the area of actuation increases, which make starting area of the actuation larger than it of the previous actuation cycle. This is same as what was observed from the previous experiment without any constraints. This also proves that the actuator had isotropic expansion.

**Top: FCW and Ionic Liquid/ Bottom: Ionic liquid**

Here, significant anisotropic deformation is observed. The discrepancy on the directionality of actuation with the result from **Top: FCW with Ionic Liquid/ Bottom: FCW with Ionic Liquid** may come from the amount of poorly-aligned wires that are deposited on the membrane. With more wires that are poorly-aligned, there is higher chance that the fibers are detached from DE in earlier phase of the actuation as explained above. In other words, the
anisotropy here is mainly due to lower number of poorly-aligned fibers. Again, this experiment was also done prior to developing the fiber deposition technique, and therefore, the control of fiber deposition was challenging.

Significant effect of an actuation on the following actuation cycle is also observed. It is observed that the initial area and actuated area get larger as the cycle number increases (Figure 29 and Figure 30). The reasoning is same as the one from Top: FCW with Ionic Liquid/ Bottom: FCW with Ionic Liquid.

As it is observed from Figure 29, and Figure 30, uniform increments and decrements for each actuation and retraction are observed for each cycle, but only in the y direction. Unlike the previous experiment, there are not much of deformations in x direction because of better alignment of the constraints. This means, by utilizing the fiber constraints, we can gain not only directionality, but also higher persistence in multiple actuation cycle.

b) DE Membrane with Fiber Constraints: HSMF

Top: HSMF with Carbon Grease / Bottom: Carbon Grease

The experiment data show that the spacing and actuation have nonlinear relationship and the amount of actuation decreases as the spacing between fibers increases (Figure 31, and Figure 32). Such trend is as expected. If each area between fibers is considered as a small “actuator,” larger actuation is anticipated when there are smaller, but more “actuators” operating in much closer distance. Simply, there are more “actuators” with less room for each of them to dissipate the compressive stress in smaller spacing.

Assembly
The interaction between each HSMF constrained area is expected to generate out of plane motion. However, it is proven to be a false prediction. There was no out of plane motion. This is due to submissive nature of the film around the active area that is compressive even with a small amount of force. Though, other interesting interaction is observed between each set of actuators. As shown in Figure 33, and Figure 34. The active area with horizontally oriented fibers is pushed by the actuation of device that has fibers vertically oriented. Also the horizontally oriented fibers start to make an angle and fan out in the opposite side of where the actuator with vertically oriented fibers is located. The device with vertical fiber alignment resists expansion of the device with horizontal fiber alignment, especially near the spacing between two devices.

*Micro-architecture Electro- Elasto- Kinematic [MEEK]*

For the MEEK muscle, as voltage increases, the acute angle increases in a scissor-like mechanism, and the dimension contracts and expands in the major axis and minor axis respectively.

Figure 35a and b shows a MEEK muscle with 44 degree bias angle. It shows that, in the low voltage range, the actuation shows linear behavior but the deformation is minimal. When it enters high voltage range (3kV and above), nonlinear, and significant deformations in both directions are observed. Resembling the McKibben actuator, contraction is in the major axis direction (x) and expansion is in minor axis direction (y) of the rhombus. Each rhombus shifts its location as it is shown from the Figure 35a. This is due to different boundary conditions that each rhombus is situated in which results rhombuses to push their adjacent rhombuses.

Samples of MEEK muscle with different initial bias angles are tested. It shows that the actuation in function of initial bias angle has nonlinear relationship (Figure 36a). The expansion of the rhombus in minor axis direction reaches its maximum when the initial bias angle is around
45 Degree and contraction of the rhombus in major axis direction reaches max when the initial bias angle is around 62 Degree. The change in bias angle is found to be most sensitive when the angles are around 15, or 70 degree, for expansion, and around 40, or 85 degree, for contraction. Of course, in order for the fiber to rotate, or the membrane to generate shear motion along its thickness, the angle cannot be too small. Otherwise, there will not be much of shear in the membrane, but pure compression. It cannot be too big, or close to 90 degree also since the motion will be interlocked by the fiber. Therefore, somewhere in the median of these two points should have been the extrema, and in fact, it is true.

Based on the general trend of the data points, it is speculated that there are saturation point in the amount of expansion and contraction, which are 15% for expansion and -4% for contraction (Figure 36a). The actuation amount becomes steady once after it reaches these points. Therefore, even though the data shows that the difference in the bias angles between maximum contraction and expansion is 20 degrees apart, there may be theoretical maximum somewhere in the range where the amount of strain is saturated. The saturation is caused by the limitation of manipulation coming from the material properties of the membrane and the constraints.

Here, the values of expansion and contraction of each experiment are normalized with each of their angles to eliminate discrepancy factor related to the size of an area. The maximum expansions, coming from maximum voltage that each sample can withhold, are measured for each experiment. They are 70kV +/- 0.5kV. During these experiments, the breakdown has high correlation with the thickness of electrodes as much as the permittivity, which we did not explore much into it yet.
3.4 FEA Modeling and Analysis

I qualitatively explain our results in light of an analytical model based on Generalized Maxwell Model (GMM) and Neo-Hookean model. The experimental values are compared with the theoretical values.

To get the general idea of what to expect from the experiments, Finite Element Analysis Methods is employed. First, simple models of the specimens that I used for each experiment are designed. Commercial software, Creo 3.0 Simulate, is used. Here are two experiments that the models are designed for: Top: HSMF with Carbon Grease / Bottom: Carbon Grease, and MEEK. For the both experiments, 3M VHB tapes with same materials properties, and Berkley NanoFil fibers are used, and therefore the assignment of the material properties for each models are identical.

3M VHB tape model has density of 9.8E-7 kg/mm^3, Neo-Hookean properties of 25kPa and 0.0008/kPa as C10 and D1, coefficient of thermal expansion of 7.7E6/C, tensile ultimate stress of 690kPa, and thermal conductivity of 16000 mmKg/sec^3C. Fiber model has the material property of nylon that comes with the software, since the nylon had closest material property as the fiber used in the experiment out of all the materials that the simulation tool already has. Linear approximation is used to reduce the amount of computational cost.

*Top: HSMF with Carbon Grease / Bottom: Carbon Grease*

The model has same thickness as the membrane used for the actual experiment (3M VHB F9473PC with 260µm original thickness, prestretched three times).
Models with different boundary condition are made to demonstrate how the spacing between HSMF affects the actuation (Figure 37). These different boundary conditions are applied to the surface perpendicular to the fiber orientation to represent the resistance of the neighboring expansions in the experiment. For each model, three different samples with different spacing between HSMF (75µm, 150µm, 300µm) are simulated. Each model contains one fiber laying on the surface of the membrane, dividing a square area into two identical rectangles. The dimensions of square edge are equal to those different spacing values.

In general, the spacing and actuation has nonlinear relationship. That is, as the spacing increases, the amount of actuation decreases but the rate that it decreases slows down and reaches plateau Figure 39.

The simulation of single fiber with different spacing shows consistent trend. As the spacing increases, the area of actuation also increases. However, each model gave slightly different values. In general, the model with strict bound on the surface normal to the fiber orientation shows the highest actuation values, then the model with constrain just with the fiber, and the model with constrain just with the spring constrain gives lowest actuation values. The trend is as expected and same as the experimental results. The explanation for this trend explained in the experimental result of Top: HSMF with Carbon Grease / Bottom: Carbon Grease.

The discrepancy of simulated actuation values within same spacing comes from, of course, the difference in boundary condition. Stronger the constraint is in a direction, more deformation there is in perpendicular to the motion that is confined Figure 39.

*Micro-architecture Electro- Elasto- Kinematic [MEEK]*
The model has same thickness as the membrane used for the actual experiment (3M VHB 4910 with 1mm original thickness, prestretched 4 times).

Total eight trusses are used to represent the HSMFs with different bias angles. 150kPa is applied on both top and bottom surface of the model. It is constrained in the center in all direction. For all the rhombi with different bias angles, the major axis is fixed to be 1mm. That means, only the minor axis of the rhombi increase.

The simulation shows that the actuation in function of initial bias angle has nonlinear relationship (Figure 41b). Also, expansion of the rhombus in minor axis direction reaches max when the initial bias angle is around 25 Degree and contraction of the rhombus in major axis direction reaches max when the initial bias angle is around 65 Degree. The simulation predicts that large expansion takes place when the degree is closer to 0, and large contraction takes place when the degree is closer to 90.

The extrema of simulated data is different from the extrema of the experimental data. The extrema of contraction and expansion from the experimental data are closer to each other and observed near 50 degree. The discrepancy of these two trends may come from the linear model approximation of MEEK muscle. This would allow it to reach the maximum expansion and maximum contraction in much earlier and later phase due to the convexity of the viscoelastic nature of elastomer and the interpolation in the linear approximation process. This allows us to establish design rules for the self-directed actuation of MEEKs, which can be tailored to a specific actuation.
3.5 Figures and Tables

Figure 12. a) Handheld microscope with light source, b) high voltage translation stage that can adjust the altitude of the voltage prong, c) stretching translational stages that DE is clamped down and stretched, d) high voltage supplier

Figure 13. Uneven prestretching. Prestretching in one direction to DE's allowable maximum strain before it fails
Figure 14. a) Fixed stages holding on to two adjacent edge of the membrane, b) clamps that holds down the membrane edges, c) high voltage translation stage that can adjust the altitude of the voltage prong, d) stretching translational stages that DE is clamped down and stretched.
Figure 15. a) Aligning mold that has b) uniformly spaced identical micro-struts. Once the mold has aligned wires wrapped around it and situated at each trough, the mold is placed on the c) the mold mount. Using d), four legs that are attached to the e), microscope ring, the mold with the mount is attached to the microscope. These four legs from the microscope module is dimensioned so that they allow the aligned wires to be positioned right at the focus of objective lens (Zeiss 5x). The image of aligned wires on the microscope module and the procedure of them being transferred to DE are displayed in Figure 16.
Figure 16. Microscope images of deposition and alignment of parallel fibers. In a), it shows the well aligned wires in focus. In b), it shows out of focus, deposited wire sets on the DE film. In c), it shows when the wires are deposited onto the DE with another set of wire that already has been deposited previously. In d), it shows that the wires are transferred from the stamp to the membrane.
Figure 17. The membrane with fiber-constraints on one side and carbon grease on the other side

Figure 18. The membrane with fiber-constraints with ionic solutions on both sides

Figure 19. The membrane with fiber-constraints with ionic solutions on one side and just the ionic solution on the other side
Figure 20. Membrane prestretching procedure.

Figure 21. Schematic shows the exploded view of MEEK muscle assembly and the set-up of the experiments
Figure 22. MEEK muscle with 30 Degree Bias Angle using clear DE membrane

Figure 23. Partially transparent except for the parameters of rhombus of the HSMF
Figure 24. These pictures depict increment of the actuated area. The image subtracts the unactuated area from the actuated area.

Figure 25. These pictures depict increment of the actuated area and also change in the area as the cycle of actuation increases. The image subtracts the area before the actuation from area after actuated.

Figure 26. The membrane with fiber-constraints on one side and carbon grease on the other side after breakdown. a) Carbon grease, b) fine copper wire.
Figure 27. The membrane with fiber-constraints with ionic solutions on both sides. From the pictures, we can see isotropic actuation and increase of both active and inactive actuation area as the actuation cycle increases.
Figure 28. Measurement of pixel number of the actuation area in x and y direction for each picture from Figure 27. It clearly shows the actuation in both direction and also gradual increase in actuation area in both directions.
Figure 29. The membrane with fiber-constraints with ionic solutions on one side and just the ionic solution on the other side. We can see anisotropic actuation and increase of both active and inactive actuation area in just y direction as the actuation cycle increases.
Figure 30. Measures pixel number of the actuation area in x and y direction for each picture from Figure 29. It clearly shows the actuation in just y direction and also gradual increase in actuation area in y direction.

<table>
<thead>
<tr>
<th>Spacing</th>
<th>Prestretched</th>
<th>Actuated</th>
</tr>
</thead>
<tbody>
<tr>
<td>a)</td>
<td><img src="image1.png" alt="Image" /></td>
<td><img src="image2.png" alt="Image" /></td>
</tr>
<tr>
<td>b)</td>
<td><img src="image3.png" alt="Image" /></td>
<td><img src="image4.png" alt="Image" /></td>
</tr>
<tr>
<td>c)</td>
<td><img src="image5.png" alt="Image" /></td>
<td><img src="image6.png" alt="Image" /></td>
</tr>
</tbody>
</table>

Figure 31. It displaces actuation values of the actuators with different fiber spacing. In a), it displays an actuator with 300µm spacing. In b), it displays an actuator with 150µm spacing. In c), it displays an actuator with 75µm spacing.
Figure 32. The plot shows the values of actuation amount obtained from the experiments as the fiber-constraints’ spacing gets larger.
Table 3. It shows how devices interact when they are placed adjacent to each other. Distance between the devices and its effect in the actuation is observed. Each device has 150µm fiber-constraint spacing. In a), the distance between devices is ~4mm. In b), the distance between devices is ~2mm. In a), the distance between devices is ~1mm.

Figure 34. This graph shows the actuation values for each device. ‘x’ indicates the actuation values of the actuators expending in vertical direction, and ‘o’ indicates the actuation values of the actuators expending in horizontal direction. Red curves indicate the assembly with ~4mm distance between devices. Blue curves indicate the assembly with ~2mm distance between devices. Green curves indicate the assembly with ~1mm distance between devices.
Figure 35. In (a), a snapshot from video displays the test specimen (44 degree bias angle) before the actuation. Dashed lines trace a rhombus shape of the aligned fibers, dotted line indicates the length of minor axis (y) (red), and doubled line indicates the length of major axis (double line). In (b), the snapshot displays the same specimen after 7kV has been applied. Solid lines trace a rhombus shape of the aligned fiber after the actuation. The comparison of the unactuated and actuated rhombi depicts the obvious deformation of MEEK muscle. In c), it illustrates the amount of strain in both minor axis, and y (●), and major axis, or x ( ■ ). In d), experimental strain values of MEEK muscles with different biased angles in the range from 0 to 90 degree are measured. It exhibits the measured strain values of each specimen in both minor axis, or y (●), and major axis, or x ( ■ ) after 7kV is applied.
Figure 36. In (a), the FEA representation of MEEK muscle with 44 degree bias angle simulates both before (dashed lines) and after (solid line) actuation. In (b), it plots the simulated strain values for bias angles from 0 to 90 degrees when 7kV is applied. It shows both minor axis, or y (●), and major axis, or x (■) simulated strain values.
Figure 37. The stress is only applied on top surface of the model. A bottom surface of the model is constrained so that the material can only expand without having out of plane deformation. a) Model with just fiber constraint. b) Model with bound on the surfaces that are perpendicular to the fiber orientation (red and orange) along with the fiber constraint. c) Model with the spring constrain attached to the clamps applied on the surfaces perpendicular to the fiber orientation along with the fiber constraint.
Figure 38. This picture depicts the difference in the spacing of the constraining fibers relative to the normalized square area. a) Model with only fiber constraint. b) Model with strict bound on the surface normal to the fiber orientation along with the fiber constraint. c) Model with constrain with the spring constrain along with the fiber constraint.
Figure 39. The graphs show the normalized values of actuation amount as the fiber-constraints’ spacing gets larger using the FEA models. In a), it displays the values of maximum displacement magnitude of the top edges of models. In b), it displays the values of maximum displacement magnitude of the bottom edges of models. In c), it displays the value of maximum displacement of the top edges of models in y direction. In d), it displays the value of maximum displacement of the bottom edges of models in y direction.

Figure 40. MEEK muscle model with 44 degree bias angle. a) DE model, b) Fibers in truss form
Figure 41. In (a), the FEA representation of MEEK muscle with 44 degree bias angle simulates both before (dashed lines) and after (solid line) actuation. In (b), it plots the simulated strain values for bias angles from 0 to 90 degrees when 7kV is applied. It shows both minor axis, or $y$ (●), and major axis, or $x$ (■) simulated strain values.
Table 2. Amount of stretching and direction of tape roll

<table>
<thead>
<tr>
<th>Date</th>
<th>pre$\lambda_x$</th>
<th>pre$\lambda_y$</th>
<th>Voltage (kV)</th>
<th>act$\lambda_x$</th>
<th>act$\lambda_Y$</th>
<th>Orientation</th>
</tr>
</thead>
<tbody>
<tr>
<td>22714</td>
<td>4.67</td>
<td>2.25</td>
<td>5.00</td>
<td>1.01</td>
<td>1.06</td>
<td>Roll Y</td>
</tr>
<tr>
<td>22814 (Thin)</td>
<td>3.45</td>
<td>0 (Uni)</td>
<td>.4~.6</td>
<td>1.01</td>
<td>1.01</td>
<td>Roll X</td>
</tr>
<tr>
<td>30314</td>
<td>2.67</td>
<td>2.00</td>
<td>5.00</td>
<td>1.02</td>
<td>1.03</td>
<td>Roll X</td>
</tr>
<tr>
<td>30314-1</td>
<td>5.36</td>
<td>2.86</td>
<td>5.00</td>
<td>1.05</td>
<td>1.17</td>
<td>Roll Y</td>
</tr>
</tbody>
</table>

Table 3. Amount of stretching of an actuator going through multiple actuation cycles

<table>
<thead>
<tr>
<th>Date</th>
<th>pre$\lambda_x$</th>
<th>pre$\lambda_y$</th>
<th>Voltage (kV)</th>
<th>act$\lambda_x$</th>
<th>act$\lambda_Y$</th>
<th>Orientation</th>
</tr>
</thead>
<tbody>
<tr>
<td>30314-1</td>
<td>5.36</td>
<td>2.86</td>
<td>5.00</td>
<td>1.05</td>
<td>1.17</td>
<td>Roll Y</td>
</tr>
<tr>
<td>2$^{rd}$</td>
<td></td>
<td></td>
<td>5.00</td>
<td>1.05</td>
<td>1.17</td>
<td></td>
</tr>
<tr>
<td>3$^{rd}$</td>
<td></td>
<td></td>
<td>5.00</td>
<td>1.05</td>
<td>1.17</td>
<td></td>
</tr>
</tbody>
</table>
CHAPTER 4 ELECTROMECHANICAL WRINKLING INSTABILITY IN DE

4.1 Introduction

Thin membranes are ubiquitous in applications spanning from filtration and packaging, to thin displays and structures. In particular, thin membranes are prone to interesting modes of instabilities under uniaxial tension and strain value, due to its geometry and Poisson’s ratio. It goes under severe shrinkage in the transverse direction to the tensile load. This is because thin membrane elastomer is structured to submit to compressive stress, caused by Poisson’s effect. As result, membrane yields to generate periodic parallel wrinkles.

This behavior can also be explained by thermodynamic energy balance. This can be modeled as a minimization of bending and stretching stresses. Thus, with different amount of strain in the membrane, the characteristics of the wrinkle are subject to change. It is determined that, as the amount of stretching increases, the wavelength decreases and amplitude increases accordingly. The details in the mechanism are explained in more detail in Chapter 2.

So far, all of these studies related to wrinkles have some similarity. There are coexistence of stiff and soft material property within a single system, existence of the compressive stress, and relatively and significantly smaller thickness.

Similar condition is observed in the fiber reinforced DE membrane. In this study, I will integrate the behavior of constrained DE expansion with the buckling of thin membrane.

4.2 Experimental Methods

Here, I modify the uniform spacing between aligned fibers, and observe how it affects the characteristics of wrinkles. Each sample implements a sets of high stiffness micro fibers (Berkley NanoFil), with 150μm, or 250μm uniform spacing, and deposit the set on a surface of DE
membrane (3M VHB F9473PC). Carbon Grease (Digi-Key) is used as compliant conductive layer to create electrostatic pressure. Carbon Grease covers the exact area of where fibers are applied. The technique of applying these fibers is elaborated in the method of manufacturing MEEK muscle (Figure 15, Figure 16, Figure 20).

Once the actuator composite is made, stainless steel conductive thread (Digi-Key) is used to connect voltage outlet from high voltage supplier (SRS PS365) to the compliant electrodes (Carbon Grease). The Thor Lab stage is used as a platform for the actuator to adjust its altitude easily to have the sample in focus of the camera (Figure 42). This is important because the observed wrinkle magnitude and wavelength (period) is on the order of tens of microns.

The pictures are taken until the material breaks down. The breakdown voltage of sample with 250µm spacing was 1.35kV, and the breakdown voltage of sample with 150µm spacing was 1.5kV. The small discrepancy in strain value coming from the difference in applied voltage can be ignored since its effect in the calculation of wavelength is minimal as proven from the previous work 15.

4.3 Results and Analysis

Figure 43 shows the gradual occurrence of wrinkles in the membrane. As it is observed, wrinkling does not happen until it reaches certain voltage or strain value. The instability of the membrane happens after when the membrane reaches its maximum voltage (Figure 5) 4. To analyze the data, the image is converted into greyscale, than binary after setting certain threshold. Of the binary image, Fast Fourier Transform (FFT) is used to identify the frequency (or wavelength) of wrinkles and p-norm is used to identify the most reasonable frequency values that are measured. Eq. 6 from Chapter 2 is used to model the electric-induced wrinkling. According to this model, I expected to see the sample with 150µm spacing to have smaller wavelength than
the one with 250µm spacing, by factor of 0.7746. With our image processing tool, the factor is found to be 0.8068 (+/- 0.0568), which is quite close to the theoretical values.

Again, we can interpret this behavior with thermodynamic. The polymer chains in DE membrane already have certain number of conformation state for each case of deformation, coming from polarization and compression. However, with the constraints from the fiber, the number of allowable and stable conformation decreases. Especially when the membrane goes under large degree of polarization and compression, there are not many conformation states to begin with. Therefore relative number of the stable conformation states that the membrane loses due to the constraints is much higher. Therefore, to balance the loss, unstable conformation states are developed and complement the lost stable conformation states by creating wrinkles in the membrane, establishing out of plane deformation.

Since the boundary is where the least constrains are, and therefore where the stress, or energy, is concentrated, as it is observed from Figure 43 and Figure 44, wrinkles form only partial area of the membrane around its edges (conductive layer boundary). Around the center of device, according to our analogy made from Chapter 3, cancelation of the stresses coming from the expansions of each “actuators” takes place in this local area. As result, even before the wrinkle propagates throughout the entire area, uneven distribution of energy causes the material to break down around the edges. This causes discrepancy between theoretical and experimental values, however, in the future, can be resolved by using control system.

With the model (Figure 45), I can obtain wrinkles with wavelength, ranging from 30 to 90µm with membranes have thicknesses ranging from 10 to 50µm.
4.4 Figures

Figure 42. Experimental setup: a) fiber optic light source, b) high voltage supply, c) objective platform, d) experiment stage

Figure 43. Pictures of DE membrane with a set of aligned fiber spaced 250μm apart (before actuation) display gradual occurrence of wrinkling deformation
Figure 44. Example of electro-induced VHB membranes wrinkling. In (a), picture of DE membrane with a set of aligned fibers, 250μm spacing apart, (before actuation) displays the wrinkling deformation after 1.35kV is applied. In (b), picture of MEEK membrane with a set of aligned fiber, 150μm spacing apart, displays the wrinkling deformation after 1.5kV is applied.

Figure 45. Theoretical model of wavelength of wrinkles in relation to the membrane's thickness and fiber spacing
5.1 Actuation of Fiber-Constrained DE

From the experiments, it is evident that the stress and strain history of DE, resulting from multiple actuation cycles, and intrinsic stress/strain coming from manufacturing process, can be controlled by adding fiber-constraints to a thin DE membrane actuator. For perfectly parallel HSMF actuators, the actuation is higher perpendicular to the fibers than it in parallel to the fibers. The amount of actuation increase as the spacing between fibers decreases. This is observed from both experiment and FEA method. However, its effect in actuation is determined to be not significant. It is shown that, by increasing the spacing from 75μm to 150μm, the strain value change from 1.74 to 1.26, and by increasing the spacing from 150μm to 300μm, the strain value change from 1.26 to 1.24. The difference in the amount of actuation that the membrane makes is very small, especially when the spacing values are relatively large.

Secondly, it is demonstrated for the first time that, by designing the fiber constraints in a mesh like pattern, contraction can also be obtained in a thin membrane DE. This new type of actuator is introduced in this thesis for the first time, and it was called Micro-architectured Electro- Elasto- Kinematic muscle (MEEK). By choosing the initial bias angle smartly, MEEK muscle can achieve a large range of actuation including expansion and contraction in the two orthogonal in-plane directions.

In the future, MEEKs can be used as versatile building blocks for future micro scale robotics and active functional surfaces due to their ability to actuate at the ~ 100μm range and flexibility. For example, in the future, the fiber angles can be locally varied among the neighboring cells to generate much more complex, and also simple actuation in plane sheet direction. Also, even though each individual rhombus has relatively large contraction in one
direction and expansion in another, collection of these rhombuses adjacent to each other’s gives nice anisotropy of the actuation in the area that each set of the fibers intersect (Figure 22). More specifically, MEEK muscle can be used as a micro manipulation tool to transport a small particle along the direction of actuation, or by scaling them further, as optical polarizers to generate much fine movement of diaphragm, which filters lights differently.

The fabrication of fiber-constrained DE can be improved. The current process of depositing is effective and controllable, however very time consuming. To accelerate testing, especially when a membrane fails after electro-mechanical instability, I suggest developing a process for reusing the fibers on a new membrane. I designed a special tool to achieve this goal but didn’t get a chance to test it. The operation mechanism of this tool is described in Figure 46.

5.2 Electromechanical wrinkling instability in DE

Fiber-constrained DE membranes can also be used to generate texture change by harnessing their electrically-induced wrinkling instability. The wrinkle pattern can be tuned by changing specific geometrical constraints, including the fiber spacing, membrane thickness, strain magnitude, and Poisson’s ratio. This behavior can be captured by a simple analytical model based on the energy minimization which shows that the wrinkling wavelength is proportion to square root of the membrane’s thickness and initial length, and inversely proportional to fourth root of the strain value. The wrinkling from the experiments in this thesis is measured to have a wavelength, ranging from 75 to 86μm.

Many future applications can utilize this micro wrinkle formation. The wrinkle generation can be used to make tunable surface that can manipulate its surface texture and hence hydrophobicity (or -philicity), as well as in generating optical patterns at very small scales.
Overall, there are several promising directions to be taken with these two new phenomena observed in micro scale DE actuators (local expansion/contraction and wrinkling). In this thesis, the membranes were fabricated by accurate but manual processes of alignment and assembly. For some of the new speculated applications, scaling down the sizes by using MEMS fabrication technologies will enable optical and chemically active surfaces at ~ 1 μm scale instead of the ~ 100μm scale.
5.3 Figures

Figure 46. a), the exploded view of fiber aligning modules. a)-1 shows the outer aligner and a)-2 shows the inner aligner. a)-3 is aligning connector that holds each halve of aligners together. Inner aligner is assembled first and fibers are aligned. Outer aligner is brought together afterward and has another set of fibers aligned. a)-4 is indicating the struts on each aligner that are used to align the fibers with uniform spacing. The relation of each aligner is clarified in more detail in c)-3. b), the assembled view of fiber aligning module. c) the assembled view of fiber depositing module. c)-2 shows the interlocking arrangement of the struts from outer and inner aligners. Aligning connector {a)-3} is replaced with depositing connector {c)-3}, which is slightly shorter so that the aligned wires are under slack, which is further explained in pictures, d) and c). d) shows difference in length between aligning modules (left) and depositing modules (right). In e), it shows the
relationship between fibers and the aligners. Aligning modules with fibers (left) are used until the fibers are adhered to DE membrane. Once the fibers are adhered to the DE membrane, by replacing the aligning connector with depositing connecter, the fibers are in slack and can move as the actuation area expands.
References


APPENDIX: IMAGE PROCESSING

A.1 Introduction

Image processing is important for experimentalists, since it can reduce the amount of complication and long process of analyzing the data. Here I demonstrate the methods that are used to analyze our experimental data that are discussed from the previous chapters.

A.2 Established Method

![Image Analysis]

Figure 47 It compares various methods of image processing and see its capability of identifying most ideal method of finding the orientation of the lines. In a), when the lines in the picture are very well aligned, all of the methods accurately represent the orientation of lines. In b) and c), as the orientation of the lines from the pictures has low directionality, only FTM has capability of representing the orientation of lines accurately. 20

Figure 47 compares the orientation of collection of lines that are artificially generated, represented as O, with the measured orientation information using three commonly used image processing methods.
Fourier Transformation Method (FTM) is self-explanatory. It uses Fourier transform (FT) to find the orientation of the fiber. This method measures how frequently, and in which direction the color changes from black to white, or white to black.

Mean Intercept Length (MIL) uses a mask with fine parallel lines that are equally spaced. This method measures the number of intersecting points of the lines from mask with the lines in the sample drawings. The mask rotates and measures the number intersecting points for each angle of rotation. Probabilistically, it is reasonable to assume that the angle with less number of the intersections is also the orientation of lines in the sample drawings. This method is only effective when the lines have strong polarization in the orientation.

Line Fraction Deviation (LFD) is similar to the MIL. It uses mask that divides up a picture into many parallel strips. This method measures the pixel brightness fraction of each strip and the standard deviation of the measurement is evaluated. The mask rotates and measures the standard deviation for each angle of rotation. Statistically, the orientation of the mask with higher standard deviation is also the orientation of the lines in the sample drawings. However, it still has similar problem as MIL. This method is only effective when the lines have strong polarization in the orientation.

As we can see from the Figure 47, Fourier Transformation Method (FTM) resembles the distribution of actual orientation of the lines, O, the most for any orientation of the lines.
A.3 Developed Methods

1. Threshold and Filter

![Figure 48](image-url) First picture shows the raw image of the sample. Second picture displays the image after the red channel filtering is applied. Third picture displays the conversion of the second picture into greyscale. Third picture displays the conversion of the greyscale image into binary image

I developed a method to harvest only the essential information from image data. It is to prevent any possible confusion that may occur during the manual measurement. In addition, the computational process is less costly since much lighter data are processed.

As we can see from Figure 48, only the red color channel is allowed to pass from the original image. Using this filtered data, the image can be reconfigured as greyscale, and then turned into binary image by selecting sufficient threshold that, again, leaves the information that I am interested.
2. Cleaning

![Image of cleaning process]

**Figure 49. These images show the clean-up process of an image in order (from top to bottom)**

Here, to eliminate unnecessary information in the image, I normalized the image by dividing it with the background image. This way, errors that are caused by shading in the original image, coming from the angle of light and other factors in the setup, can be minimized during the analysis. Once the image is normalized, I make the edge of object (dark area) smoother by using dilating gradient mask. Other small objects that we are not interested can be eliminated by selectively filling the small dark area with white pixels, using component labeling algorithm that are commonly used in visual control. Finally, threshold is applied to the image to make it binary, so that, when the orientation of fibers are automatically measured in the next step, the system can take advantage over the strong polarization between the object and the background, and make easier decision.
3. Micro FFT

![Micro FFT Process](image)

**Figure 50.** These images shows the micro FFT process of an image in order (from top to bottom)

As mentioned from the previous section, Fourier transformation can measure the orientation of the object very accurately. However, to measure the orientation of object that is not in a straight line but in a curvature, like what is shown in Figure 50, some preprocessing of the image is required. To do this, the image is segmented into many small square tiles which contain equal number of pixels, named ‘macropixel.’ Each macropixel are sized to contain at least one edge of the object. Also, for the same reason, the object is thinned down by finding the median of object in each row of the macropixel. This way, we can optimize the processing time. Fast Fourier transform (FFT) is used to calculate the orientation of processed object.
Figure 51. In a), it displays the result of micro FFT with sufficiently large macropixel. In b), it displays the result of micro FTM with smaller macropixel.

Here we see the final result of the entire process. As we can see, there are many erroneous interpretations in the result, but, by modifying the macropixel size, we can minimize the errors in the analysis, and maximize the accuracy and resolution. Overall, we observe that this process gives sufficient orientation and curvature information of the objects.
Figure 52. This image shows the progression of image process, discussed in this chapter, on the CNT forest in order (from top to bottom).

We apply the macropixel method onto much denser and highly oriented CNT forest. The last picture demonstrates the orientation of CNT with the gradient field as the representation of the fiber orientation information. The average orientation of fiber’s curvature is calculated to be 36.76 degree.
4. **Macro FFT**

![Macro FFT Image](image-url)

**Figure 53.** These images show the macro FFT process of the CNT forest in order (from top left to bottom)

Here we apply the FFT to entire area of the picture as the reference has it done from the introduction. We were also able to measure the general direction of fibers, which is found to be 120 degree.

Indeed, applying the Fourier transform method to the whole picture definitely gives good idea of how the fibers are oriented. However, if we want to find the curvature of the fiber, the micro FFT method is ideal.

5. **Periodicity Measurement**

Fourier series (FS) is a tool that is often used to fit the data using compound of multiple sinusoidal functions with various magnitudes and frequencies. Therefore, if we have periodic trend in our data, FS can give us good representation of the periodicity in data. I used this aspect of FS on our buckling analysis.

As described in eq. 6, we are interested to know the spacing between constraining fibers, and its relation to the wavelength of buckles that takes place in between two fibers. First, in order to impose FS fitting onto an image, some preprocessing is required.
First, I manually crop a section of the image that is significant. Of the cropped section, I convert the image format to greyscale, and then set a sufficient threshold to make it binary. Next, I use rectangular macropixel (n by m, n>m) to polarize the area of macropixel with whichever information (white or black) is dominant. Using this process, I sweep across the columns of the image, then its rows.
Figure 55. It shows the progress of applying FS on the preprocessed image (left). The preprocessed image is simplified with a new image indicating the location of extremes (middle). The location of extremes is identified using FS.

Using p-norm, I find the sufficient threshold for the system to decide whether it is trough or peak of the buckles, or whether it is the fiber constraints or the membrane. Then, as we can see from the Figure 55, we can obtain the general information about the buckling characteristics, and the spacing between fibers. Using this information, we can find the wavelength of the wrinkles and the strain value of the actuator.