Investigation and Characterization of Conductive DEAP Polymer Materials with Nickel Nanocomposites

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Investigation and Characterization of Conductive DEAP Polymer Materials with Nickel Nanocomposites

By
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Submitted in partial fulfillment of the requirements for the

Master of Science
In
Mechanical Engineering

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August 14th, 2015
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Abstract

Dielectric ElectroActive Polymers, or DEAPs, are devices with coupled electrical and mechanical responses that resemble stretchable parallel plate capacitors, that can act as actuators, sensors, or electrical generators. Currently, the electrode layers on the top and bottom are generally conductive carbon grease, which is dirty and also causes curing issues for certain polymers. This thesis explores several polymers and conductive fillers to identify a conductive nanocomposite material, to replace the grease electrode with a solid material and eliminate issues associated with grease electrodes. It then characterizes the mechanical and electric properties and how they change during cyclic loading, while augmenting an equibiaxial tensile testing machine and advancing the knowledge of equibiaxial characterization.

The most promising polymer/filler combination was found to be EcoFlex30, a platinum cure silicone rubber, containing seven volume percent of nickel nanostrands and three volume percent of 0.1 mm length nickel-coated carbon fiber. Using two conductive fillers of different sizes resulted in much higher conductivity than a single filler alone, and an enormous piezoresistive effect. This material gave weak conductivity at no load, increasing several orders of magnitude as strained and well surpassing the benchmark of 1.2 S/m set by conductive carbon grease. Elastomer materials were found to have conductivities as high as 275 S/m under peak strain, and changing the nickel-coated carbon fiber length allowed for strains over 120%. Equibiaxial stress-strain curves were also analyzed for energy lost through hysteresis, in order to compare to published results for DEAPs used as Dielectric Energy Generators. Results and recommendations are presented for using and further improving the materials for applications of DEAPs used as energy harvesters and capacitive sensors, using the material alone as a piezoresistive sensor, and improving the equibiaxial characterization process.
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<th>Symbol</th>
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<tbody>
<tr>
<td>A</td>
<td>Area</td>
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<tr>
<td>C</td>
<td>Capacitance</td>
</tr>
<tr>
<td>d</td>
<td>Distance</td>
</tr>
<tr>
<td>E</td>
<td>Energy</td>
</tr>
<tr>
<td>Q</td>
<td>Electrical charge</td>
</tr>
<tr>
<td>r</td>
<td>Radius</td>
</tr>
<tr>
<td>ROT</td>
<td>Rotation, either in X, Y, or Z directions, in ANSYS simulations</td>
</tr>
<tr>
<td>SDF</td>
<td>Stress decay factor</td>
</tr>
<tr>
<td>t</td>
<td>Thickness</td>
</tr>
<tr>
<td>U</td>
<td>Translations, either in X, Y, or Z directions, in ANSYS simulations</td>
</tr>
<tr>
<td>V</td>
<td>Voltage</td>
</tr>
<tr>
<td>v</td>
<td>Volume</td>
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<tr>
<td>(\epsilon)</td>
<td>Absolute permittivity of a material</td>
</tr>
<tr>
<td>(\epsilon_0)</td>
<td>Permittivity of free space</td>
</tr>
<tr>
<td>(\epsilon_r)</td>
<td>Relative permittivity of a material</td>
</tr>
<tr>
<td>(\epsilon_1, \epsilon_2, \epsilon_3)</td>
<td>Engineering strains in the three principal directions</td>
</tr>
<tr>
<td>(\lambda_1, \lambda_2, \lambda_3)</td>
<td>Stretch ratios in the principal directions, one more than the engineering strains</td>
</tr>
<tr>
<td>(\lambda_r, \lambda_z)</td>
<td>Stretch ratios for circular DEAP</td>
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<td>(\sigma_1, \sigma_2, \sigma_3)</td>
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1. Introduction

EAPs, or Electro-Active Polymers, are increasingly being used in new applications, such as sensors, actuators, switches, and generators, and as the number of EAP uses continues to grow, so too will the need for characterization techniques and material property data. DEAPs, or Dielectric EAPs, are considered a frontrunner in artificial muscle technology [1], due to their smooth motion, much smoother than current robotic prostheses. They can also be used to generate electricity for such applications as wireless devices, as the voltage difference across them changes when physically stretched and relaxed in a charged state [2], or sensors, due to their changes in capacitance and conductivity when strained [3]. A DEAP consists of a dielectric layer sandwiched between two conductive compliant electrodes, effectively forming a parallel-plate capacitor, so stretching the sensor causes a measureable change in capacitance, representative of the strain of the sensor [4]. This is a similar idea to resistance being used to measure strain in a conventional strain gauge, except the DEAP measures much higher strain levels.

The materials generally used for the conductive layers (the electrodes) of DEAPs are carbon black powder, or conductive carbon grease (CCG) [5]. Other methods of making compliant, conductive electrodes are being explored [6], such as overlaying conductive wires on the electrode layer, but these adversely affect the DEAP voltage distributions [7]. Both carbon materials are brushed on, temporary in nature, incapable of prolonged use and can contaminate anything they come in contact with. To address this need requires an electrode material that is adequately conductive, something that is a solid or self-contained layer within the DEAP, not an exposed powder or grease. That material, in addition to being conductive, would need to be very compliant and capable of reaching high strains, the higher the better [8]. The literature shows that this is a gap that needs to be addressed.

There are many benefits from and applications for DEAPs made using these materials. For instance, a very soft DEAP edema sensor for measuring edema (swelling) in human patients’ limbs was investigated at RIT [8]. This could benefit the estimated 140 to 250 million people affected worldwide by lymphedema, a major cause of chronic edema (fluid-
Currently, the “state of the art” clinical edema measurement consists of either pressing a finger into the skin and rating how long it takes to “rebound” on a scale of 1 to 4 [10], measuring the dimensions of an affected limb [11], or measuring the volume of water displaced [10]. Developing a conductive elastomer is the first step in creating a DEAP sensor such as this. The usefulness extends beyond high-deformation strain gauges and sensing applications, and into energy harvesting and actuation. Any technology that could benefit from conductive elastomers—high strain electrodes—stands to be advanced, in fact.

Uniaxial characterization is necessary to establish the material properties under most loading conditions, and equibiaxial characterization extends the material property knowledge to specific DEAP applications involving the previously mentioned sensor as well as power generation. Uniaxial loading is loading along a single axis, such as in a tensile test, whereas biaxial loading generally exists when a planar specimen is stretched in two principal directions. Equibiaxial loading is a special case of biaxial loading, when the stresses are equal in those principal directions. Situations with this loading scenario include planar sensors shaped like a circular patch, and Dielectric Elastomer Generators (DEG), or energy harvesters, as their power density is squared if it can be stretched in two directions instead of one. Such DEG’s could be used to power mobile devices, such as under the keys of a wireless keyboard, or in the soles of shoes to generate electricity while walking [3]. Conductivity is also paramount, in order to reduce resistive losses, both for DEGs and artificial muscle applications.

This work therefore set out to create a conductive elastomer that can be used as an electrode material in DEAPs, and establish the trends between conductivity, stress, and strain under uniaxial tension conditions and between stress and strain in equibiaxial tension. An existing equibiaxial tensile testing machine will be augmented to measure the electrical properties along with stress and strain data, testing samples to near their maximum strain under cyclic loading. While the characterization of the relationship between strain and conductivity of DEAP materials in biaxial tension is not within the scope of this thesis, the groundwork will be laid for future work.
2. Review of the Literature

This section will cover the necessary background and prior work. The background includes the fundamentals of equibiaxial tension, EAPs, capacitance and conductivity, and how the effects of nanoparticulate concentrations would benefit EAP applications. Prior work involves the equibiaxial characterizations of similar materials, studies on the effects of nanoparticulates on capacitance and conductivity, and examples of DEAPs under equibiaxial tension being used as pressure transducers and energy generators.

2.1 Equibiaxial Tension Testing
2.1.1 Equibiaxial stress state

Equibiaxial stress is a state of stress that occurs when a material element has one principal stress that is zero, and the other two principal stresses are nonzero and equal. As a special case of plane stress, this loading state typically occurs in membranes, but may be found in other geometries as well. An inflating alveolar sac wall, a blood vessel aneurism, the sclera of the eye, the fabric on a taut trampoline, or any thin member or plate being stretched equally in two directions experiences this stress state. Biaxial material properties are different from standard uniaxial properties, leading to the need to be able to test and characterize materials under biaxial loading.

Materials exhibit different stress-strain responses depending on the state of loading [12]. In uniaxial loading, a specimen’s cross-sectional area decreases as the specimen elongates, according to the Poisson effect. For the square specimen in Figure 2.1 below, the thickness and width both decrease as the length increases under uniaxial loading. In a biaxial test however, the specimen is being stretched in two directions. The width is not only constrained from contracting, but also being pulled at the same rate as the other direction. This results in a stiffer response as shown in Figure 2.2.
The state of stress changes depending on the specimen geometry, boundary conditions, and loading, and will usually be different in different regions of the same specimen. Biaxial tension is simply the case where a specimen experiences principal stresses in two directions, in contrast to uniaxial tension which has stress in one principal direction, and a generic stress state which has nonzero stress components in all three principal directions. For planar specimens, the difference between uniaxial and biaxial stress is shown in Figure 2.1. Equibiaxial tension is a special case of biaxial tension that occurs when the two nonzero principal stresses are equal, and the stress, in the out of plane direction is equal to zero by the assumption of plane stress.

2.1.2 Biaxial tensile testing equipment

Equipment capable of testing materials in biaxial tension is now commercially available [14-16] using hydraulic or servomotor actuation. The 574 Series of biaxial tension testers produced by Test Resources and shown in Figure 2.3, for example, use four independently controlled servoactuators to strain the specimen along the X and Y directions, achieving a wide range of load or strain profiles up to a frequency of 15Hz.
A simple equibiaxial tension tester design was developed by Brieu et al. in 2007 [17], and mounted on an Instron 4302 tensile testing machine as shown in Figure 2.4. The rising of the Instron machine’s crosshead extends the machine’s top vertical clamp (2), while linkages (7) couple the vertical motion to an equal extension of the horizontal arms (5). The ratio of vertical and horizontal extensions is a 1:1 ratio, for equibiaxial strain.

In 2009, Ferrara and a multidisciplinary senior design team at the Rochester Institute of Technology (RIT) modified Brieu’s design in order to be actuated by electric motors and lead screws, and run as a standalone system [18]. Smoger [19] modified the machine using minimum constraint design principles in order to eliminate binding, and performed extensive sensitivity analysis on the effects of clamp misalignment and non-equibiaxial loading. His implementation of the design, shown in Figure 2.5, was then used to test specimens of Medium Ultraflex, a hyperelastic polymer [19].
Figure 2.5: Redesigned equibiaxial tensile testing machine; CAD model (top) and physical machine (bottom) [20]

Figure 2.6: Range of motion of redesigned equibiaxial tensile testing machine; minimum extension (Left), maximum extension (Right) [20]
Figure 2.6 demonstrates the machine’s motion by showing the points of minimum and maximum extension. The system includes two 2-lb load cells to measure the forces applied in the x and y directions, and an optical technique is used to measure displacement. A dot pattern was marked on the specimen, visible in Figure 2.7, corresponding to the corners of the Central Diamond Region outlined in Figure 2.8 where equibiaxial conditions were present. These points were imaged at prescribed intervals during testing, in order to measure x and y displacements in the region of the specimen where equibiaxial loads were expected.

Equibiaxial stress-strain data was then calculated from these loads and displacements, and input to the curvefitting tool under ANSYS’s Material Models options in order to create a two-parameter Mooney-Rivlin material model. The Mooney-Rivlin two-parameter equation was chosen because it provided the best fit to the data, without the increased complexity of Mooney-Rivlin models with more parameters. This method is also a good choice for testing because of the strain range that it tests over. Many DEAP applications, including the edema sensor, should be capable of up to 100% strain, and this machine is capable of generating data up to 88% strain [20].

In 2013, Clarke et al. [21] created another method to test elastomers in equibiaxial tension. This testing machine consisted of a linear servo, connected to an assembly of wires,
clips, and pulleys to apply this force to the boundary of a circular specimen. Figure 2.9(a,b) show schematically how the force is applied to the specimen boundary, where $R_{pre}$ is the initial specimen radius and $R_{max}$ is the radius at maximum strain. Figure 2.9(c,d) show the specimen at its positions of zero and maximum strain.

![Diagram](image)

**Figure 2.9: Equibiaxial testing machine designed by Clarke et al. [21]**

2.1.3 Biaxial tensile testing specimens

The following equibiaxial tensile testing specimens were designed for machines capable of stretching specimens along their x and y axes, which was the case for all the machines presented except for the one in Figure 2.9.

In 2005, Sacks et al. [22] simulated different types of specimens used to test materials in equibiaxial tension. These included suture attachment (SA), clamped square specimens (CSS), and clamped cruciform specimens (CCS) with varying corner radii, shown in Figure 2.10. Sutures were found to be the “best” attachment technique, in terms of not interfering with the specimen’s interior stresses through Saint-Venants edge effects, and providing the largest area in the center of the specimen under an equibiaxial stress state. This can be seen
when comparing the SA Von Mises stress plot in Figure 2.10(a) with the CSS in Figure 2.10(b) and CCS in Figure 2.10(c) and (d). The clamped boundary conditions in Figure 2.10(b,c,d) prohibit the material from translating along the edge parallel to the boundary, while the sutures accommodate that motion, allowing the material to move sideways and decreasing the stress in the corners. The high stress levels at the corners present with clamped conditions cause a so-called “stress-shielding effect,” as much more of the load is carried at the corners and sides of the specimen, diverting it away from the center. By allowing the material at the edges to translate along the boundary, load can be more effectively transferred to the interior. Sutures allowed the stress to be transferred to the specimen center very effectively, but presented other challenges in the form of preventing damage to the sample at the suture locations. Figure 2.12 shows the resulting stress distributions as a function of location along the center axes for a direct comparison of the different geometries, and Figure 2.11 shows the loading for each type.

Figure 2.10: FEA stress distributions [22]
Smoger [20] later examined the design of planar specimens used for equibiaxial testing, using ANSYS finite element software, and chose clamped boundary conditions and the CCS specimen type. Clamps were chosen over sutures because the mounting method was more repeatable and the risk of tearing was lower. The CCS specimen type was chosen
over the CSS because it allowed for higher stress values (i.e., closer to the nominal applied stresses) in the interior of the specimen because of lower stress-shielding. Several parameters were then varied in order to achieve the largest region with equibiaxial stress at the specimen center. These parameters included the width and length of the legs and center, and the equibiaxial region was found to be especially sensitive to the fillet radii between the legs. The region of interest for characterizing mechanical properties was in the shape of a diamond at the center of the specimen as shown previously in Figure 2.8.

### Table 2.1: Specimen geometries examined by Smoger [20]

<table>
<thead>
<tr>
<th>Specimen Type</th>
<th>Description</th>
<th>Configurations</th>
</tr>
</thead>
<tbody>
<tr>
<td><strong>CLAMPED CRUCIFORM SPECIMEN (CCS) WITH LEGS</strong></td>
<td>Cruciform specimen with exposed legs (clamped at end of leg)</td>
<td>Fillet radius of R=1, 5, 10mm</td>
</tr>
<tr>
<td><strong>CLAMPED CRUCIFORM SPECIMEN (CCS) WITHOUT LEGS</strong></td>
<td>Cruciform specimen with no exposed legs (clamped at fillet foot)</td>
<td>Fillet radius of R=1, 4, 5, 6, 10mm</td>
</tr>
<tr>
<td><strong>CLAMPED SQUARE SPECIMEN (CSS)</strong></td>
<td>Square specimen with clamps inside the boundaries</td>
<td>Dimensions of 2” x 2”, 3” x 3”</td>
</tr>
</tbody>
</table>
The different geometry types are shown in Table 2.1 and the results of Smoger’s simulations are shown in Table 2.2. The SDF (Stress Decay Factor) is the percentage of the uniaxial edge stress that is experienced in the central diamond region, and the range represents the amount of the specimen (relative to the length of the specimen between the clamps) where equibiaxial stress conditions are present. Smoger identified the geometry boxed in Table 2.2 as the best candidate for testing: a Clamped Cruciform Specimen style with no legs, and fillet radii of 4mm. The table shows that equibiaxial stress conditions are present for 10.8% of the width between the clamps, and that the equibiaxial stress there is 83% of the uniaxial stress present at the specimen edge, as opposed to being carried near the fillet as in Figure 2.10(c). This is therefore the optimal specimen configuration, because the equibiaxial stresses are the highest and the central diamond region is the largest. This means that the data will reach higher values, allowing a broader dataset to be used for material characterization, and that the strain measurements can be taken over a larger area which is easier, and more accurate, since measuring larger distances is easier and gives a lower percent measurement error. Smoger’s recommended geometry will therefore be used for equibiaxial testing in this work.

<table>
<thead>
<tr>
<th>Specimen</th>
<th>SDF (%)</th>
<th>Range (mm)</th>
<th>Range (%)</th>
</tr>
</thead>
<tbody>
<tr>
<td>CCS R01 leg</td>
<td>80.74%</td>
<td>3.6</td>
<td>7.0%</td>
</tr>
<tr>
<td>CCS R05 leg</td>
<td>76.99%</td>
<td>3.6</td>
<td>7.0%</td>
</tr>
<tr>
<td>CCS R05 no leg</td>
<td>82.91%</td>
<td>3.6</td>
<td>10.2%</td>
</tr>
<tr>
<td>CCS R06 no leg</td>
<td>82.30%</td>
<td>3.6</td>
<td>9.6%</td>
</tr>
<tr>
<td>CCS R10 leg</td>
<td>74.48%</td>
<td>3.6</td>
<td>5.8%</td>
</tr>
<tr>
<td>CCS R10 no leg</td>
<td>77.98%</td>
<td>3.6</td>
<td>7.9%</td>
</tr>
<tr>
<td>CSS 2x2</td>
<td>83.10%</td>
<td>3.6</td>
<td>8.8%</td>
</tr>
<tr>
<td>CSS 3x3</td>
<td>69.18%</td>
<td>3.6</td>
<td>5.4%</td>
</tr>
</tbody>
</table>

Table 2.2: Analysis results to find specimen with largest equibiaxial stress region [20]

2.2 Electroactive polymers (EAPs)

Electroactive polymers are polymeric materials with piezoelectric characteristics, causing one of two effects: moving in response to an applied voltage, or experiencing a voltage change when deformed in a charged state. Other types of materials have similar properties coupling the deflection of the material to such things as chemical concentrations, magnetic fields, pressures, etc., but EAPs are unique due to the large strains they are capable
of creating through applied voltage [23]. EAPs constructed from VHB acrylic tape manufactured by 3M for example, were found to be capable of a maximum actuation strain of 380% [24].

The two main categories of EAPs are ionic EAPs and electronic EAPs. Ionic EAPs are driven by the diffusion of ions, and electronic EAPs by an electric field [25]. Electronic EAPs, which are the larger focus of this work, are the more powerful of the two classes with fast-acting power over a larger range of motion, but they require actuation voltages in the several kilovolt range. Ionic EAPs require much lower voltages, on the order of several volts, but must be contained in an electrolyte bath isolated from the environment, are much slower-acting, and produce much less actuation force. Examples of the two main types are shown in Table 2.3, and a summary of the advantages and disadvantages as actuators in Table 2.4.

<table>
<thead>
<tr>
<th>Electronic EAPs</th>
<th>Ionic EAPs</th>
</tr>
</thead>
<tbody>
<tr>
<td>- Dielectric EAP (DEAP)</td>
<td>- Carbon Nanotube (CNT)</td>
</tr>
<tr>
<td>- Electrostrictive Graft Elastomers</td>
<td>- Conductive Polymers (CP)</td>
</tr>
<tr>
<td>- Electrostrictive Paper</td>
<td>- Electrorheological Fluids (ERF)</td>
</tr>
<tr>
<td>- Electro-Viscoelastic Elastomers</td>
<td>- Ionic Polymer Gels (IPG)</td>
</tr>
<tr>
<td>- Ferroelectric Polymers</td>
<td>- Ionic Polymer Metal Composites (IPMC)</td>
</tr>
<tr>
<td>- Liquid Crystal Elastomers (LCE)</td>
<td></td>
</tr>
</tbody>
</table>

Table 2.3: Types of EAPs [26]

2.2.1 Ionic EAPs

An ionic EAP’s movement results from electrochemical reactions occurring at electrodes on the sides of a polymer. The ionic polymer membrane composite (IPMC), shown in Figure 2.13, is a type of ionic EAP composed of a center polymer gel, covered by a compliant electrode on either side [23]. The polymer gel must be electrically insulating and be conducive to ion diffusion, the electrodes must be electrically conductive, and both the polymer and electrodes must be capable of achieving high strains. The bending of ionic EAPs is due to the polymer matrix shrinking at extreme pH values [27], as shown in the chart of Figure 2.14 for polyacrylamide (PAAM) hydrogels, which is ultimately due to electrochemical reactions cause by the applied voltage.
<table>
<thead>
<tr>
<th>Type</th>
<th>Advantages</th>
<th>Disadvantages</th>
</tr>
</thead>
<tbody>
<tr>
<td>Electronic EAP</td>
<td>• Exhibit rapid response (milliseconds)</td>
<td>• Requires high voltages (~100 MV/meter). Recent development allowed for (~20 MV/meter) in the Ferroelectric EAP</td>
</tr>
<tr>
<td></td>
<td>• Can hold strain under DC activation</td>
<td></td>
</tr>
<tr>
<td></td>
<td>• Induces relatively large actuation forces</td>
<td></td>
</tr>
<tr>
<td></td>
<td>• Exhibits high mechanical energy density</td>
<td></td>
</tr>
<tr>
<td></td>
<td>• Can operate for a long time in room conditions</td>
<td></td>
</tr>
<tr>
<td>Ionic EAP</td>
<td>• Natural bi-directional actuation that depends on the voltage polarity.</td>
<td>• Requires using an electrolyte</td>
</tr>
<tr>
<td></td>
<td>• Requires low voltage</td>
<td>• Require encapsulation or protective layer in order to operate in open air conditions</td>
</tr>
<tr>
<td></td>
<td></td>
<td>• Low electromechanical coupling efficiency</td>
</tr>
<tr>
<td></td>
<td></td>
<td>• Except for CPs and NTs, ionic EAPs do not hold strain under DC voltage</td>
</tr>
<tr>
<td></td>
<td></td>
<td>• Slow response (fraction of a second)</td>
</tr>
<tr>
<td></td>
<td></td>
<td>• Bending EAPs give low actuation forces</td>
</tr>
<tr>
<td></td>
<td></td>
<td>• Electrolysis occurs in aqueous systems at &gt; 1.23 Volts</td>
</tr>
</tbody>
</table>

Table 2.4: Summary of EAP type advantages and disadvantages for actuation (modified from [25])

The direction of bending can also be changed by reversing the polarity of the voltage. A solution is acidic when it has a surplus of positive hydronium ions, $\text{H}_3\text{O}^+$, and basic when it has a surplus of negative hydroxide ions, $\text{OH}^-$. When an electric potential is applied across the ionic EAP’s electrodes, a portion of the water undergoes electrolysis [28] and breaks into hydronium and hydroxide ions. The positive charge at the anode creates an acidic solution by increasing the concentration of hydronium ions, which then attracts the negatively-charged hydroxide ions out of the polymer gel. The negative charge at the cathode likewise creates a basic solution, by increasing the concentration of hydroxide ions, which then pulls

Figure 2.13: IPMC in unactivated (Left) and activated (Right) states
out the positively-charged hydronium ions [27]. This results in the polymer shrinking at the electrodes as the contents of the polymer gel matrix are drawn out, as shown in Figure 2.15. Hydroxide ions are much smaller than hydronium ions, and can therefore diffuse much more quickly. This results in the polymer gel matrix shrinking faster in acidic solutions than basic ones, causing the EAP to bend toward the acidic anode side [27]. This is also the reason why ionic EAPs are typically not capable of maintaining an actuation force for a long period of time under a constant voltage [25].

Figure 2.14: Polymer shrinkage as a function of pH [27]

![Figure 2.14: Polymer shrinkage as a function of pH [27]](image)

Figure 2.15: Ion diffusion in Ionic EAPs [27]

![Figure 2.15: Ion diffusion in Ionic EAPs [27]](image)
As time goes on, diffusion occurring at the basic cathode side eventually catches up as the diffusion toward the anode slows. The pH values of the acidic and basic sides remain very different, but the ratio of their percent shrinkage values is much closer to one as the reaction tends toward steady state.

2.2.2 Dielectric EAP Overview

The most popular electronic EAP type, and the type targeted by this thesis, is the dielectric EAP (DEAP). DEAPs consist of a polymer sandwiched between compliant electrodes, as shown in Figure 2.16. This basic configuration looks identical to a parallel plate capacitor. The concept of a DEAP is simple; when a voltage is applied, the two electrodes are attracted to each other, causing the DEAP to get thinner and longer. Alternatively, straining the sensor longitudinally causes it to get thinner, changing the capacitance, which can be measured as an indicator of the state of the sensor [5]. Creation and characterization of the materials used in this type of EAP is the main goal of the proposed work.

![Figure 2.16: Basic construction and operation of DEAP actuator](image)

2.2.3 DEAP Layers

DEAPs have two types of layers. Essentially mimicking a parallel plate capacitor (see Figure 2.16), there are three distinct layers, of two different types. The outer two layers serve as electrodes, while the center layer is a dielectric medium.
The compliant electrodes on either side of the DEAP’s dielectric layer need to be capable of two things. The first is conducting electricity in order to energize the entire electrode, and the second is being subjected to high strains without permanently deforming or significantly resisting the strain, meaning that it has to have a very low elastic modulus and high percent elongation. By blending conductive nanoparticulates into a polymer base, the conductivity of the resulting composite matrix can be increased substantially [29].

How much nanoparticulate is required is a matter of reaching the percolation threshold. This point is where the concentration of conductive particles is high enough for them to form continuous paths through the composite matrix, causing the bulk conductivity to change from being dominated by the base polymer matrix, to being dominated by the more conductive network formed within. The volume fraction of nanoparticulate required to reach the percolation threshold is a function of polymer and particulate material properties, such as the interfacial energy and surface tension existing between the particles, and particle geometry, specifically the size, aspect ratio, and orientation. The change in conductivity caused by reaching the percolation threshold of carbon black in nylon is shown in Figure 2.17, for example.

Increased conductivity of the compliant electrodes is critical for avoiding a voltage drop across the electrode surface of the DEAP, representative of resistive losses, and a detriment to performance regardless of application. Graf and Maas [7] studied the magnitude of this voltage drop, and Figure 2.18 shows the voltage distribution across the DEAP from regions of positive voltage at the upper electrode contact point(s) to negative voltage at the lower electrode contact point(s). The voltage differential between the top and bottom
electrodes, or voltage gradient, is highest at the electrode contact points, but quickly decreases moving laterally away from them.

Figure 2.18: Simulation results showing voltage distribution around electrode contact points [7]

Figure 2.19: Analytical voltage drop as a function of position along the DEAP and the number of contact points, n [7]

Figure 2.18 also shows that having multiple contact points results in a much more uniform voltage across the electrode surfaces (less voltage drop), and Figure 2.19 shows this pattern for up to five contact points, where the height of the line represents the voltage differential between the top and bottom electrode surfaces. The voltage differential is critical for a DEAP used as an actuator, as any section of the DEAP with a lower voltage differential would have less attraction between the two electrode surfaces and produce less actuation force. Making the top and bottom layers of the DEAP more conductive will result in less voltage drop and a more uniform voltage over the entire DEAP electrode surface.

The center layer of the DEAP is the dielectric layer. As an electric insulator, it must be non-conductive, meaning it must have a high dielectric breakdown voltage. It also must be capable of reaching high strain levels, at least the same strain as the conductive electrode material, and for sensing applications, it is desirable to increase the capacitance to make it more measureable. This can be done by adding particulate, such as barium titanate powder, which can be mixed into the base polymer [31] in order to increase the dielectric constant. This has the effect of increasing the conductivity and decreasing the breakdown voltage of
the dielectric, but should only pose a problem at higher voltages. Higher amounts of barium titanate do alter the composite material’s stiffness however, and lower the elongation to failure [32], also effects that should be studied.

2.2.4 DEAP Applications

The first characteristic of EAPs, moving in response to an applied voltage, makes these materials useful as actuators, especially biomimetic actuators. Applying a voltage that causes the polymer to move would achieve continuous, fluid-like motion. This is highly desirable for creating an artificial muscle [28], something that has been difficult to achieve with current technology, and would greatly improve human prosthetic devices. Other noteworthy applications being considered for these actuators are in space vehicles, such as the need for low power consumption windshield wipers to continually clean solar cells on rovers [33, 34].

The second ability of electronic EAPs, specifically dielectric EAPs (DEAPs), is changing in capacitance when deformed. This makes them useful as sensors, as they can transform strain into a measureable capacitance output. One potential application under consideration at RIT is a DEAP-based edema sensor [8]. Edema is the accumulation of fluid in the body, the medical term for swelling [10]. It can be caused by congestive heart disease, inflammation, poor circulation of the lymphatic system, and a host of other conditions, and can be necessary to measure in a hospital. Currently, the only method for “measuring” an edema level is to push one’s finger into the affected area and observe how long it takes the “pit” formed by the finger to rebound [35], shown in Figure 2.20. The proposed sensor, shown conceptually in Figure 2.21, would be attached to the affected area with a non-irritating adhesive. The capacitance is the same as a parallel-plate capacitor [6], and governed by Equation 2.1, where \( \epsilon_0 \) is the permittivity of free space, \( \epsilon_r \) is the relative permittivity of the material, \( A \) is the instantaneous area of the capacitor, and \( l, w, \) and \( t \) are the length, width, and distance separating the electrodes of the DEAP at any given point in time, and change as force is applied.

\[
C = \frac{\epsilon_0 \epsilon_r A}{t} \tag{2.1}
\]
The product $\varepsilon_0 \varepsilon_r$ forms the absolute permittivity of the dielectric material forming the capacitor, and measures the electric field “stored” per unit charge, with units of Farads per meter.

The capacitance can be expressed in terms of $t_0$ (the initial distance between the electrodes), $A_0$ (the initial area), and the stretch ratios $\lambda_1$, $\lambda_2$, and $\lambda_3$. $\lambda_1$ is equal to the current length divided by the original length, and $\lambda_2$ and $\lambda_3$ are the same for width and thickness respectively. The sensor is assumed to be of constant width, making $\lambda_2$ equal to one, and $\lambda_1 \lambda_2 \lambda_3 = 1$ by conservation of volume, making $\lambda_3 = 1/\lambda_1$. For these assumptions, the formula for capacitance becomes Equation 2.2.

$$C = \varepsilon_0 \varepsilon_r \frac{A}{t} = \varepsilon_0 \varepsilon_r \frac{A_0 \lambda_1 \lambda_2}{t_0 \lambda_3} = \varepsilon_0 \varepsilon_r \frac{A_0 \lambda_1^2}{t_0}$$

2.2
Conceptually, straining the sensor increases the parallel plate capacitor’s electrode area, and brings them closer together by the Poisson effect, causing the capacitance to increase quadratically as the sensor is longitudinally strained. The capacitance of the sensor can then be measured to calculate the strain of the sensor, and correlated to a particular amount of swelling at that location.

Technology using DEAPs as sensors for biomedical applications has already been proven feasible. In 2010, Ozsecen used a DEAP within a rapid prototyped casing to create a pressure sensor for use in a blood pressure monitor. He found that the DEAP’s capacitance followed the applied pressure surprisingly well and claims that a proper calibration curve will allow the DEAPs to be implemented as pressure transducers in a new type of disposable blood pressure cuff [37]. The relations between the properties will be discussed in further detail in Section 2.2.5.

The third relevant characteristic of DEAPs do is that they change the voltage across them when strained in a charged state. The amount of charge on the DEAP is constant, causing the voltage across the DEAP to change inversely with capacitance by Equation 2.3. Stretching and releasing the DEAP results in a cycle such as shown in Figure 2.23 [2], and several of these in series would result in an array of miniature power-generating devices at a usable voltage, also called energy harvesting.

\[
Q_i = Q_f
\]

\[
C_i V_i = C_f V_f
\]

\[
V_f = V_i \ast \frac{C_i}{C_f}
\] 2.3
Kohnbluh [3] also presents a detailed list of applications that EAPs may be well suited for given sufficient development. Some examples, in addition the ones already mentioned, include MEMS devices, pumping mechanisms with unusual geometries, and flexible printed electronics.

2.2.5 DEAP Electrical Properties

A DEAP’s electrical properties, such as its capacitance, can be used to measure some other useful property such as pressure or strain (either uniaxial or equibiaxial). Others can be enhanced to optimize the DEAP for applications such as energy harvesting. Some of the work characterizing DEAPs in equibiaxial configurations has already been mentioned in earlier sections, but those studies will be revisited in this and the following section, emphasizing the DEAP electrical properties.

Goulbourne et al. [6] and Ozsecen [37] have both characterized the pressure vs. capacitance relationships, in order to use DEAPs as pressure transducers. In 2007, Goulbourne clamped a DEAP at its perimeter and pressurized one side, as shown in Figure 2.24. The elastomer used as the dielectric medium was 3M’s VHB 4905 acrylic tape, and the electrode materials tested were carbon grease, silver grease,
graphite powder, and graphite spray. Goulbourne also found that the greases, specifically the carbon grease, worked better than the powder and spray, as the powder and spray would develop microcracks at large strains resulting in a drop in conductivity across the surface. Goulbourne compared the DEAP’s capacitance against the applied pressure, and showed that there was a definite relation between the two. This is important for sensor applications, as it demonstrates using the DEAP capacitance to measure other physical quantities, in this case, pressure and volume. Both are graphed as a function of the volume of air displacing the DEAP membrane in Figure 2.25. Tests were also performed with different levels of pre-stretch; these results are shown in Figure 2.26.

![Figure 2.25: Comparison of pressure and capacitance [6]](image1)

![Figure 2.26: Capacitance curves at different initial pre-strains [6]](image2)
In a similar application, Ozsecen explored the use of a DEAP sensor encased in a rapid-prototyped plastic casing to measure blood pressure (Figure 2.27). As with Goulbourne’s study, the goal was to relate capacitance to pressure, using the former as a way to measure the latter. The author did not present the data in the form of capacitance as a function of pressure, but plots the two variables on the same graph. Figure 2.28 shows the measurements for the sensor when the pressure is directly applied, and Figure 2.29 shows capacitance vs. blood pressure when the sensor is used inside the blood pressure cuff. The author states that the capacitance closely matches analytical predictions, follows the pressure very closely when used inside the cuff, and that a proper calibration curve will make it possible to calculate the pressure from the capacitance change.

Figure 2.27: DEAP sensor in rapid prototyped casing [37]

Figure 2.28: Capacitance output of DEAP sensor in response to directly applied pressure [37]
A company called StretchSense has made, and is now selling, DEAP sensors as well [38]. These sensors use a silicone rubber and what appears to be carbon grease to make a capacitive DEAP sensor, and attach a circuit and Bluetooth module to it to transmit the signal. The sensor is capable of measuring human body movement, and variants of it are capable of being sewn into clothing, and sensing bending, shear, and even pressure. Because the sensor is encased in silicone, the electrode grease material is not exposed.

In 2013, Clarke et al. [21] characterized the capacitance of DEAPs in equibiaxial tension using the experimental system shown previously in Figure 2.9 and reproduced in part in Figure 2.9 and reproduced in part in to apply equibiaxial strain through radial loading. In keeping with analytical
models, the capacitance was found to be proportional to fourth power of the stretch ratio in the radial direction, $\lambda_r$ [21], which the authors note had not yet been taken advantage of in the literature for energy generation purposes. An energy density of 560 J/kg was achieved here, a substantial improvement over previously reported values ranging from 2.8 to 300 J/kg, as the DEAP is more fully stretched. The 4th order dependence on the radial stretch ratio is predicted by modifying Equation 2.2 for equibiaxial radial strain to give Equation 2.4. Note, by conservation of volume, $\lambda_z = 1/\lambda_r^2$.

$$C = \varepsilon_0 \varepsilon_r \frac{A}{t} = \varepsilon_0 \varepsilon_r \frac{\pi r^2}{t} = \varepsilon_0 \varepsilon_r \frac{\pi r_0^2 \lambda_r^2}{t_0 \lambda_z} = \varepsilon_0 \varepsilon_r \frac{\pi r_0^2 \lambda_r^2}{t_0 (1/\lambda_r^2)} = \varepsilon_0 \varepsilon_r \frac{\pi r_0^2 \lambda_r^4}{t_0 \lambda_r^2}$$

\[ 2.4 \]

2.3 DEAP Materials Summary

2.3.1 Typical Polymers

Polydimethylsiloxane (PDMS) is the polymer usually used to build DEAPs. As an engineering material with much prior art, it is not lacking standard uniaxial characterization, or even equibiaxial characterization [39].

PDMS was tensile tested by Lisitano in 2013 [40], studying the stiffness of Dow-Corning’s Sylgard 184, a commercially available formulation of PDMS, as a function of the percent of cross-linking agent used. The stress-strain curves for the samples with 20% crosslinking agent, for example, are shown in Figure 2.32, and the stiffness vs. weight percent of crosslinking agent plot is shown in Figure 2.33. The trend shown in Figure 2.33 shows that altering the mix ratio allows customization of the resulting PDMS, a characteristic common to many crosslinking elastomers.
PDMS fails at relatively low strain values when compared to other elastomers, generally below 200%. A company called Smooth-On, for instance, carries many types of polymers for do-it-yourself projects such as mold making or special effect costumes, with lower elastic moduli and capable of strains up to 1000% [41].
2.3.2 Enhancing DEAP Electrode Conductivity

As described in Section 2.2.3, the addition of conductive nanoparticles increases electrode conductivity. The most commonly studied nanoparticles have been carbon black and silver (abbreviated Ag). One study by Niu et al. [29] examined the conductivity, \( \sigma \), as functions of concentration and strain. Their silver particles were 1-2\( \mu \)m diameter platelets, and the carbon black particles were 40-100nm diameter spheres. Their results for conductivity change at the percolation threshold under zero strain are shown in Figure 2.34. Note the different scales for conductivity and weight percentages. The silver-PDMS conductivity is on a log scale, starting below \( 10^{-2} \) and increasing to \( 10^{4} \) Siemens per meter, whereas the carbon black conductivity varies from near zero to above 35 S/m and appears to continue to increase. A much higher concentration of silver is required than of carbon black to reach the percolation threshold however.

In addition to the higher cost of silver compared to carbon black, the resulting nanocomposite material becomes less and less like a polymer as the material becomes almost fully comprised of the conductor, making it stiff and brittle [29]. The silver PDMS also displays an enormous amount of hysteresis, as the conductivity increases with strain but hardly drops at all when the strain is released, as shown in the right side of Figure 2.35. This would make the material unsuitable for applications requiring the conductivity to be quantified, but would not be as significant if the goal was simply to increase conductivity without needing to relate it back to the value of the strain. The increasing stiffness would however be a detriment to a compliant sensor.
For material preparation, Niu et al. stated that they put the silver platelets used in their experiments through a cleaning procedure of acetone, ethanol, deionized water, and freeze-drying, but did not report any difficulties or preparation requirements for the carbon black powder [29]. This is in contradiction with preliminary work done by Spath [8], who was unable to get the carbon black and PDMS nanocomposite to cure regardless of curing time and temperature, using the same Sylgard 184 two-part PDMS produced by Dow Corning and Vulcan XC72 carbon black powder produced by Cabot Inc. The only observed differences in the reported methods and materials are that Spath’s particles had average diameter of 20-40nm, vs. 40-100nm for the Niu study. Spath then tried using silicone caulk instead of PDMS, which was successful, and believed to be due to the fact that Sylgard 184 has a platinum-catalyzed curing system, while the silicone caulk has a hydrolytic curing system. The difference may be that Niu et al. had been able to passivate the carbon black used in their experiments, perhaps by mixing the carbon black with a smaller amount of the base polymer to completely cover it, before adding the crosslinking agent and more base polymer, so that the crosslinking agent and carbon black did not interact, but the papers published did not contain these details.

Nickel nanocomposites produced by Conductive Composites were also selected as possible candidates. Their products include Nickel Nanostrands, which are extremely high aspect ratio nanoscale nickel slivers, and Nickel-Coated Carbon Fiber. Since these are fairly new materials, the available literature is somewhat limited, but it is clear that it is capable of
producing very conductive polymer nanocomposites. Most of the research performed on the materials has been either on making conductive coatings or paints, or focuses on the piezoresistive behavior of materials, assessing the feasibility of making a sensor that works by assessing the change in conductance of the material as it’s strained, but this research is fairly limited in scope. While the change in conductivity is sought to be maximized for creating a piezoresistive sensor, the DEAP conductive electrode simply requires the conductivity to be maximized. Examining the data from a study performed by Johnson, et al. in Figure 2.36 [42], the sample with 9 vol% nanostrands and 3 vol% nickel-coated carbon fiber has a resistivity around 3 Ω-meters at zero strain, or a conductivity of .33 Siemens per meter. At approximately 58% strain, right before the specimen breaks, the conductivity has increased two orders of magnitude to about 33 S/m. For comparison, the bulk conductivity of conductive carbon grease is about 1.2 S/m. The data for the stress required to achieve these strain levels is not given, so the material stiffness is unknown, but the conductivity at zero strain is at least comparable to carbon grease, which was established to by Goulbourne [6] to be an effective, though dirty, electrode material.

Figure 2.36: Volume Resistivity of Nickel Nanocomposites by Johnson, et al. [42]
3. Gap in the Literature and Statement of Work

3.1 Summary of the State of the Art

Biaxial tensile testing machines are available commercially, most being capable of straining a specimen along each axis independently. The one located at RIT is driven by an electric motor, and uses a set of linkages to couple the extension in the X and Y axes together at a fixed 1:1 ratio. Its range of motion makes it capable of straining specimens up to 88% [20], spanning the range over which DEAPs are typically used and making it a good candidate for the proposed work.

The Mechanical Engineering Department at RIT has seen multiple theses done in the field of electroactive polymers. This has included enhancing the actuation ability of ionic EAPs [43], assessing the feasibility of integrating together multiple types of EAPs to create a human bicep muscle [28], and now enhancing electrode conductivity. Work outside the university has resulted in implementing DEAP sensors in biomedical applications such as blood pressure monitors [37], as well as using them in other applications including power generation [21] and actuation imitating artificial muscles [34].

3.2 Summary of Needs

Currently, carbon grease electrodes generally make DEAPs impractical to use outside of the lab. Creating conductive elastomers with nanoparticulates will enable DEAPs that are self-contained and more practical, and the higher the conductivity, the further the resistive losses can be reduced. That results in stronger actuation forces for DEAPs used as artificial muscles, more efficient DEAP energy harvesters, and more accurate DEAP sensors.

Additionally, some applications employ DEAPs in an equibiaxial loading state, specifically certain sensors and Dielectric Elastomer Generators (DEGs). This creates a need for equibiaxial material properties in addition to uniaxial ones.
3.3 Statement of Work

The body of work to be done in this thesis was broken up as follows:

1. Preparation of the equibiaxial testing machine. This work will be presented in Chapter 4.
   a. Validation of the equibiaxial tension tester.
      i. Continue Smoger’s boiling flask work, experimental and ANSYS
      ii. Test pressurized planar membrane and model in ANSYS
   b. Augmentation of equibiaxial tensile testing machine and test method
      i. The machine will be modified to test stiffer materials
      ii. The machine will be modified to electrically isolate the specimen at the grips
      iii. Resistance measurement capability will be implemented with a National Instruments myDAQ device, and the existing LabVIEW program modified to support this
      iv. A more powerful optical strain measurement technique will be implemented to improve accuracy and determine the strain field across the entire specimen

2. Combinations of different elastomers and conductive fillers will be evaluated to identify one that best meets the criteria of high conductivity, low stiffness, and capable of withstanding high strains. This work will be presented in Chapter 5.
   a. Different base polymers will be evaluated in order to find one with low stiffness and high elongation to failure
   b. Different conductive particulates will be evaluated in order to make the resulting composite as conductive as possible, while minimizing the increase in stiffness, and maintaining the capability to withstand high strain
   c. Special consideration will be given to avoiding cure inhibition caused by incompatibilities between the conductive filler and base polymer

3. The most promising DEAP materials will be tested and characterized using the augmented tensile testing machine. This work will be presented in Chapter 5.
a. Flat, rectangular specimens will be tested uniaxially on the equibiaxial tension tester, by not attaching two of the clamps to the specimen. Uniaxial stress, strain, and conductance data will be generated.

b. Flat, cruciform-shaped specimens will be tested on the equibiaxial tension tester. Equibiaxial stress and strain data will be generated.

c. The suitability of the materials for use in different DEAP and piezoresistive application will be assessed.
4. Material Characterization Techniques

4.1 Boiling flask model

Smoger [20] completed the characterization of Medium Ultraflex, a nonlinear elastic polymer, under equibiaxial tension, and began the work of validating the results using an expanding spherical sac model of the same material. The apparatus in Figure 4.1 was originally created at RIT in support of lung flow field research being performed by Oakes [44], and the specimen was created by hot dipping a 50mL boiling flask into the molten polymer. The goal of the validation was to show that the material properties determined through the use of the equibiaxial tensile tester, developed by Smoger and Ferrara, accurately predicted the pressure vs. deflection behavior of the expanding spherical sac in an equibiaxial loading state. This would be done by measuring the deflection of the bottom of the specimen as a function of applied pressure, and comparing it to predictions generated in ANSYS using the material properties that were the results of Smoger’s characterization.

Figure 4.1: Boiling flask experimental setup
4.1.1 Boiling Flask – Experimental

The specimen was previously created, by hot dipping a 50mL boiling flask into molten Ultraflex, allowing the Ultraflex to solidify, and removing the newly-formed membrane from the outside of the flask [45]. This specimen was then zip-tied around the bottom of the graduated cylinder and placed inside the acrylic enclosure as shown Figure 4.1. The tank and graduated cylinder were both filled with glycerin, and the pressure differential across the boiling flask specimen was controlled by adding or removing glycerin either from the tank using the syringe pump, or from the graduated cylinder protruding from the top using a pipette. The inner and outer pressures were both measured using an inclined manometer, and a picture was taken at each data point using a Nikon D3100 14-megapixel camera. The pressures on the inside and outside of the inflating model were determined from height measurements in a pair of manometers with Equation 4.1, where θ is the angle of inclination of the manometer with respect to horizontal and \( l_{\text{glycerin}} \) is the length of glycerin column as measured in the inclined manometer, defined in Figure 4.2.

\[
P = \rho gh = \rho_{\text{glycerin}} \cdot g \cdot h_{\text{glycerin}} = \rho_{\text{glycerin}} \cdot g \cdot l_{\text{glycerin}} \cdot \sin \theta
\]  

Figure 4.2: Inclined manometer variables and setup

The vertical deflection of the bottom of the specimen was found by analyzing the pictures of the specimen in ImageJ [46], which essentially counts pixels between two points in an image. Pixels in the vertical direction were measured between the bottom of the membrane and a reference point, and converted to distance by measuring a known length on a machinists ruler in order to calculate a conversion factor of pixels/inch. The vertical deflection at each pressure was simply the difference between the current and initial vertical
An example of processing an image in ImageJ is shown in Figure 4.3. The pressure difference was increased steadily, and data was recorded from a zero pressure differential to 350 Pa.

Figure 4.3: Distance measurement in ImageJ

4.1.2 Boiling Flask – Finite Element Analysis

The boiling flask model was then simulated in ANSYS using Smoger’s Medium Ultraflex material properties in order to compare to the experimental results. The model was meshed with Shell281 elements, as this element type was found to be the best suited for the boiling flask application, and more accurate than Shell181 due to the addition of midside nodes [18]. The model was then constrained in all degrees of freedom at the boundary representing the mouth of the flask, and an outward pressure was applied on the model as shown in Figure 4.5.

As explained in detail in the following section, the physical boiling flask model was found to have a non-uniform thickness. The change in thickness as a function of height was accounted for by measuring the wall thickness in ImageJ, plotting thickness vs. height in
Excel (Figure 4.6), and fitting a cubic polynomial to the data. This trendline equation was used to create a thickness function within ANSYS (a .func file), which was applied to the Shell elements to define the thickness as a function of the Y-coordinate.

Figure 4.5: Cross section of boiling flask with pressure, constraints, and thickness definition

![Cross section of boiling flask with pressure, constraints, and thickness definition](image)

The vertical deformation and Von Mises stress are shown in Figure 4.7 and Figure 4.8. Note that the stress is highest at the top, where the model is clamped and also the thinnest, and where the elements must bend more as the model expands.

![Thickness function data and fit](image)

The equation for the thickness function is:

\[ t = (1.929 \times 10^{-5})y^3 - (1.481 \times 10^{-3})y^2 + (4.379 \times 10^{-2})y + 0.4776 \]

\[ R^2 = 0.94 \]
4.1.3 Boiling Flask – Results and Interpretation

The experimental and ANSYS results for displacement of the bottom of the expanding sac were plotted in Figure 4.9 alongside the linear elastic model originally used by
Oakes [44] and the material model defined by Ferrara [18]. The lines in the plot show how the predicted equibiaxial material properties of Medium Ultraflex have changed with improvements in test and analysis techniques, culminating with Smoger’s properties, and are compared with the new experimental boiling flask data.

There were, however, uncertainties in the deflection and thickness measurements. Previous studies assumed that the specimen’s initial inner diameter was the same as the outer diameter of the glass boiling flask upon when it was molded on, but it was found that the Ultraflex expanded upon removal from the flask. Several pictures were taken around the zero-pressure area, and these data points formed a small cluster around the origin in the deflection vs. pressure plot of Figure 4.9. The initial point of zero deflection was chosen to be roughly in the middle of the cluster, at an estimated initial diameter of 58±1mm, a significant difference from the flask’s outer diameter of 52.0mm.

The larger source of deflection measurement error was unwanted rotation/translation occurring at the same time as the inflation that was supposed to be measured, as the specimen wasn’t initially taut. Analogous to the inflation of a typical balloon, the specimen doesn’t inflate uniformly in all directions, and also required a significant amount of pressure to appear taut and spherical. It was observed to take as much as 100 Pa during experimentation before specimen motion appeared to be due to inflation only. The graph also shows the experimental data being shifted to the right (i.e., material behaves as stiffer than the model predicts) of the Smoger ANSYS Equibiaxial prediction once the pressure is above 100Pa.
The last uncertainty stems from measuring the specimen’s wall thickness. The latest ANSYS simulation incorporates a wall thickness that varies from .6 mm at the neck to 2.0 mm at the bottom instead of the previously assumed constant 1.05 mm, but this assumes there is no refractive error. The measurements are made optically on a picture taken with a near zero pressure differential, looking at the side of the model, and the result is the length of the line shown in Figure 4.10. Any error in the wall thickness measurement would change the thickness profile applied to the ANSYS model, causing it to predict a different response.
The uncertainties listed above cast doubt onto the thickness profile, and more importantly the amount of deflection due to radial expansion during the initial slack-removal phase. As a result, agreement between the ANSYS experimental pressure-deflection curves was not obtained. However, qualitative agreement can be shown by shifting the experimental pressure-deflection curve by 110 Pa on the x-axis and 1 mm lower on the y-axis in Figure 4.11, in an attempt to remove the effect of the initial slack-removal phase. As stated by Buerzle, uncertainty in the location of the “zero-stress state” can potentially shift the entire pressure-deflection curve [47], which is the case here, as the initial point of zero deflection is the only difference between the original data of Figure 4.9, and the modified data of Figure 4.11. These results seemed to show that the equibiaxial material characterization approach using the equibiaxial tensile tester at least gave a reasonable approximation of material properties. However, future work is recommended in order to further refine this approach.

Figure 4.11: Comparison of predictions with shifted experimental data
4.2 Pressurized Planar Membrane

The results of the boiling flask model led to the creation of another compliant model for additional validation Smoger’s material properties, using a geometry that would not be subject to the difficulties found with the boiling flask model. This geometry, a pressurized planar membrane (PPM), is essentially a circle clamped around its perimeter, subjected to a pressure achieved through a difference in water column height on either side of the specimen. The approach was similar to that of the boiling flask: record experimental pressure vs. deflection data and compare it to ANSYS simulations. The geometry was also nearly identical to that used by Goulbourne to test dielectric elastomer sensors [6], discussed in Section 2.2.5.

4.2.1 Pressurized Planar Membrane – Experimental

An aluminum mold was constructed to make a four inch diameter specimen up to a quarter-inch thick. As shown in Figure 4.12, a piece of aluminum roughly three inches in diameter was placed between the hot plate and the mold, in order to minimize the amount of heat flowing into the specimen from the edge of the mold. Without it, the temperature at the edge of the specimen would be higher than in the center because it was contacting more mold surface (i.e., the side of the mold), causing the polymer to start to burn, evidenced by yellowing and smoke coming from the edges. The flat specimen shape was much easier to mold than the boiling flask was to hot dip, because it only had to be hot enough to remove the air bubbles, and the challenge of achieving a uniform thickness was eliminated. Removing the air bubbles consisted of taking care to place flat slices of material into the mold, pressing the air out from under the material as it began to melt, and using a pin, needle, or other small sharp object to remove remaining bubbles or move them to the edge of the specimen.
The PPM test fixture used to test this specimen consisted of two pieces of \( \frac{1}{4} \)” wall, 3” ID polycarbonate pipe, with buckles on the sides to clamp them to each other end-to-end with the specimen in between, and two barbed fittings in the side. The concept is shown in Figure 4.13.

An alignment fixture was also created that could align the fixture in a vertical milling machine. The bottom half of the assembly fixture was clamped in the vice, while the upper half was secured in a collet in an unplugged milling machine. The two halves were lined up horizontally, and the head was lowered to pre-compress the specimen prior to latching the test fixture. This pre-compression forces the material outward, cancelling the material being forced inward when the pipe walls come together, resulting in a taut specimen. Figure 4.14 shows a specimen mounted by hand, and Figure 4.15 shows the assembly fixture in use. The rolling at the edges comes from the misalignment, and buckling is caused when the latches pull the two halves together and force material to the inside of the fixture.
Figure 4.14: Sample clamped without assembly fixture

Figure 4.15: Assembling with assembly fixture in milling machine

The specimen and test fixture were then placed into a glass fish tank, manometer tubes were connected to the ports in the fixture, and the tank filled with water and air removed from within the tubing and fixture. The manometer, Nikon D3100 camera, and ImageJ were used as in the boiling flask trials to record the inner and outer pressures, and take and process images, as shown in Figure 4.17. ImageJ’s small measurement line can be
seen in the picture on the right, extending from the bottom of the membrane to the edge of the fixture.

Figure 4.16: Pressurized planar membrane setup

Figure 4.17: Image processing in ImageJ

4.2.2 Pressurized Planar Membrane – Finite Element Analysis

The pressurized planar membrane was modeled in ANSYS in cylindrical coordinates as a 4° sector of a 76.2 mm diameter plane. This was done using the ANSYS verification manual example VM218 [48], changing geometry and material properties, and remeshing. It was meshed with Shell181 elements, given the specimen thickness of 5.715 mm as measured
on the actual specimen with a set of calipers, given symmetry boundary conditions at the sides, constrained in all degrees of freedom at the clamped boundary, and loaded with a higher pressure than it was capable of withstanding. The model was solved using automatic substepping, incrementing until the simulation diverged. Exporting the bottom deflection from the Time History postprocessor, the simulation time and percent of the load applied are directly proportional, so multiplying by the original applied load maps the simulation time into applied pressure. Figure 4.18 shows the meshed model, and the deflection and Von Mises stress results at the highest pressure differential that converged, 1246 Pa. There are two images for Von Mises, as the values are different on the top and bottom sides of the plane.

Figure 4.18: FEA analysis: a) Meshed model, b) out of plane deformation UZ, c) Von Mises strain - top view, d) Von Mises strain - bottom view

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As a planar specimen, each element is under conditions of biaxial stress. For conditions of equibiaxial stress, the two principal stresses will be equal. Figure 4.19 shows the principal stresses plotted as a function of radial position under a pressure of 1000Pa, and Figure 4.20 shows the ratio of these principal stresses, which would be one under equibiaxial conditions.

Figure 4.19: Stress distributions across specimen

Figure 4.20: Ratio of principal stresses across specimen
4.2.3 Pressurized Planar Membrane – Results and interpretation

The pressurized planar membrane results are shown in Figure 4.21. Though the edges of the specimen are physically clamped, which theoretically does not allow rotation at the boundary, it was found that allowing rotation improved the ANSYS convergence without greatly altering the results, so both curves are shown below in order to extend the curve. The pressure vs. deflection data curves from the experiment and ANSYS predictions had very similar shape, but the ANSYS simulations predicted a less stiff response than what was seen experimentally. In the end this was insufficient to validate Smoger’s approach for determining material constants for materials in equibiaxial tension [20], and this was attributed to the fact that conditions in much of the specimen were not equibiaxial. It was reassuring to see that the experimental model became unstable (began to explode) as predicted in the simulation, though at a higher pressure. This is shown in Figure 4.22. Increasing the pressure expands the model, which increases the area, and increases the force exerted by the pressure, at a rate higher than the material can handle. If not constrained, and the pressure held constant at this point, it would essentially pop like a balloon.

![Deflection vs. Pressure Differential Results](image-url)

Figure 4.21: Deflection vs. applied pressure differential
Figure 4.22: Experimental model becoming unstable

Figure 4.22 also shows why the clamped boundary condition in ANSYS prevented convergence; the material is so flexible that allowing rotation at the boundary during the simulation is actually a better representation of the physical model.

4.3 Variable Stress Decay Factor

In Smoger’s method, the stress decay factor is assumed to be constant with respect to load, a necessary condition for the equibiaxial characterization process [20]. It was realized during the course of this thesis, however, that the assumption is only valid if the slope of the material’s stress-strain curve is constant, that is, if the material is linear elastic. If that is the case, then a given load will cause a certain stress field and a certain strain field, and doubling that load will double the stress and strain values. Any ratios between stresses at different locations will thus remain constant, since all the values scale together. If the stress-strain curve is not linear, then different parts of the specimen will be exhibiting different tangent moduli, and those ratios cannot remain constant. The material used for developing the characterization and for the validation attempts was extremely nonlinear.

Figure 4.23 shows an example for why the stress decay factor cannot remain constant for a nonlinear elastic material, even within a specimen of a given geometry, as it is loaded and deformed. Assume that all deformation is elastic. At state 1, the edge of the specimen (in the leg), denoted edge$_1$, and the central diamond region, CDR$_1$, have certain stress and
strain values. As the specimen is loaded, the stresses and strains increase along the curve, to state 2. If the material were linear elastic, the stresses (and strains) would double when the load doubles, as stated above, and the ratio of the stress in the central diamond region to that at the edge (the definition of SDF) would remain constant. Since this material is not linear elastic, however, the ratio of the stresses at state 1, $\sigma_{CDR_1}/\sigma_{edge_1}$, cannot be equal to the ratio of the stresses at state 2, $\sigma_{CDR_2}/\sigma_{edge_2}$, and the SDF is nonconstant.

![Graph showing stress-strain relationship](image)

Figure 4.23: Example case of why SDF is not constant

In order to make the equibiaxial characterization procedure valid and accurate, the variable stress decay factor must be taken into account. For a given geometry and given material model, the SDF can be fit to simulation data as a function of nominal stress, but as a function instead of a constant. The equibiaxial stress can then be determined from experimental data and the simulated (nonconstant) SDF as before, and the entire process can be iterated as described by Smoger, with more steps in the iterative process. This would result in a back-and-forth approach, between improving the equibiaxial stress data obtained experimentally, improving the material model (the constitutive equation between equibiaxial stress and equibiaxial strain), and improving the (variable) stress decay factor. These three steps would be performed iteratively until the data, SDF function, and material model all converged. It may even be possible to streamline the process, by rolling the stress decay function into the material model.
4.4 Summary of Equibiaxial Tensile Tester Validation

The Boiling Flask model and Pressurized Planar Membrane (PPM) were two test geometries used to try to validate the equibiaxial stress-strain characterization process developed by Ferrara [18] and Smoger [20]. ANSYS simulations predicted a less-stiff response than what was seen during experimentation, and though qualitative agreement could be found by adjusting the data from the Boiling Flask model, it was clear that the characterization method was flawed from a fundamental assumption that was inaccurate. The material characterization technique assumed a constant stress decay factor, which is only valid for linear elastic materials, but it is now understood what the steps are to correct the approach. The validation will require additional future work, and was not pursued further for this thesis.

4.5 Augmentation of Testing Equipment

4.5.1 Augmentation overview

In order to measure the electrical characteristics using the existing test platform, the machine needed to be modified in order to isolate the specimens from the rest of the machine and computer electrically, and the resistance measurement had to be taken and recorded with the rest of the data in LabVIEW. Additionally, the stiffer specimens tested in this work required larger load cells (10 pound capacity) than Smoger and Ferrara’s load cells (2 pound capacity) [18, 20]. Finally the method of optical strain measurement was improved from the displacements of four points, to calculating a continuous strain field.

4.5.2 Electrical Isolation of Specimen from Machine

To isolate the specimen electrically, one half of each specimen grip was replaced with a grip made from acrylic, which is an excellent electrical insulator. The incorporation of the acrylic piece broke the electrical path between the specimen and the machine, as illustrated in Figure 4.24.
The other half of the grip does not electrically contact the rest of the machine, and was left as aluminum, so that resistance measurements can be made by attaching multimeter leads to the clamp tightening bolt, which screws into the aluminum clamp. This is much better than attaching the multimeter leads directly to the specimen, because alligator clips have the potential to bite the material and cause it to tear prematurely when being stretched. Additionally, some contact resistance variability between the specimen and alligator clips was observed during preliminary testing, and this was eliminated by tying the leads in to the aluminum clamp.

4.5.3 Resistance Measurement

To take resistance measurements, a National Instruments myDAQ was used, as the device features a built-in multimeter, and several virtual instruments (VIs), or programs, in the National Instruments ELVIS software for use with LabVIEW [49]. One of these is the Digital Multimeter (DMM) VI, capable of making a resistance measurement once the measurement range is specified. However, the programs for the device are designed to run through their own Graphical User Interface, and the sub-VIs require the user to select a measurement range and do not incorporate autoranging. If the selected range is too low then the VI returns an over-range error, and if the range is too high then the measurement can
become inaccurate. The specimen resistance can also change by multiple orders of magnitude during a test, so the VI was modified to perform auto-ranging. The DMM VI was modified and incorporated such that the myDAQ would adjust its measurement range until it was correct, and then record multiple readings in order to reduce the possibility of a random measurement error. If the resistance is too high for the myDAQ to measure (above the 20 MΩ), then the program will remain in an open loop until it receives user interaction. There are three cases where the resistance is above 20 MΩ: The first is if the specimen simply has very high resistivity at low strain, the second is if the specimen has broken or come out of the clamp in which case the test should be stopped, or if a multimeter lead isn’t making proper contact in which case the test should be stopped also. Further details of the operating procedures for the machine and programs are included in Appendix A – Machine Operation Instructions.

4.5.4 Optical Strain Measurement Improvement

In Smoger’s work, the strain in the equibiaxial area was determined by optically measuring the displacements of four points at the corners of the central diamond region show in Figure 4.25 using ImageJ [46]. These points were marked using a stencil with a diamond pattern. This had the drawback of basing the strain data on the movement of only four points, or the strain in a given direction from the difference in displacement of two points.

In order to improve this, a method called Digital Image Correlation (DIC) was introduced. DIC is another way to optically measure strain, by tracking the differences in the greyscale color of the specimen surface [50]. It requires the specimen to be coated with a “speckle pattern,” which is a partial covering of spray paint misted over the surface of the specimen as shown in Figure 4.26. A MATLAB package called Ncorr [51] is then used to
analyze the series of images that were taken as the specimen was strained. This is similar to the previous method but determines the strain field across the entire specimen, by tracking the movement of subsets of greyscale values. Also similar to the previous method, the pixel distances can be converted to inches or millimeters by using a reference image to measure the number of pixels in a known length. Ncorr runs calculations within the user-defined Region Of Interest, and stores this data within a multidimensional data array that can be accessed and used in other calculations. The image analysis process and the additional code is included in Appendix B – Image Analysis Process in Ncorr.

The DIC method was convenient because it made the strain measurement much more accurate, and required no modifications to the physical test equipment other than the spray paint application to the specimens. The same LabVIEW compatible Pixelink camera was used as in previous work. Ncorr is open source and free to use, and has been verified to produce results comparable to VIC-2D, the standard for DIC software produced by Correlated Solutions [53]. Applying the speckle pattern was also fairly easy, and the paint spattering was so thin and discontinuous that it had no effect on the specimen stiffness.

Following the modifications, the experimental test machine with an equibiaxial specimen with the DIC speckle pattern is shown in Figure 4.27. This is the typical test configuration used for all tensile testing done for this thesis.
Figure 4.27: Experimental testing machine following modifications
5. Creation and Characterization of Conductive Elastomers

5.1 Polymer Selection

As noted earlier, Dow Corning’s Sylgard 184 PDMS is typically used in the making of DEAPs, due to low stiffness (high compliance) and fairly high elongation to failure. However, there are other silicone-based elastomers that have even lower stiffness and higher elongation values, which were explored as alternatives to PDMS.

For making conductive elastomers, the crosslinking reaction that occurs when the polymer cures also becomes a significant consideration. There are two main categories of crosslinking reactions that most silicone-based polymers can undergo to cure. These are addition reactions, also called platinum cure systems, and condensation reactions, also called tin cure systems. Whether a polymer is a platinum-cure or a tin-cure refers to the atom involved in catalyzing the crosslinking reaction, which is what causes the individual polymer chains to link together into larger bulky molecules that form a solid material.

Compliant silicones made using either cure system can be bought commercially from Smooth-On [41], but platinum-cured ones are available in lower stiffnesses. The softest, least-stiff is the Ecoflex series, which recently expanded to include even softer polymer than those available at the beginning of this work. The chosen elastomer was Ecoflex OO-30, with Shore OO hardness of 30, a low elastic modulus of 10 psi (over the first 100% strain), and high elongation to failure of 900% with excellent tear strength. Platinum-cure silicones, however, were very sensitive to cure inhibition due to contact with other chemical species, specifically carbon. Hence, blending carbon black into the specimens resulted in specimens that either would not dry, or dried hard and brittle, cracking while drying. The specimen in Figure 5.1 was created by combining a silicone elastomer with Cabot carbon black, and the one in Figure 5.2 is a urethane rubber that cured with smaller amounts of carbon black, but that became brittle with larger amounts.
Toluene was used as a solvent to thin mixtures that were too cake-like, and all specimens were mixed in a Thinky ARM-310 centrifugal planetary mixer, as mixing by hand was insufficient and did not achieve complete dispersion, as evidenced by the nonhomogenous appearance in Figure 5.3. The only specimens that did not use toluene and were not mixed using the centrifugal mixer were those with Medium Ultraflex thermoplastic, as the material had too high of a melting temperature and would rapidly solidify in the mixer.
To improve on the EcoFlex and carbon black results, a different conductive filler could be used, a different cure system could be used, or steps could be taken to attempt to passivate the carbon black and keep it from causing cure inhibition. Different polymers were explored, including urethanes and tin-cure silicones from Smooth-On, as well as a thermoplastic elastomer.

Medium Ultraflex, the thermoplastic elastomer from Douglas and Sturgess [54] was used by Smoger in his work. It is similar in appearance to ballistic gel with Shore hardness 00-24, and has a fairly low melting temperature for a thermoplastic elastomer. The goal of using a thermoplastic material was that the polymer could be melted, the filler mixed into solution, and then be allowed to cool (as opposed to cure). Though the overall idea worked, resulting in a mildly conductive material that was very stretchy, it was impractical as the material was very prone to “crumbling.” It appeared as though the polymer and filler were not mixed together to form a homogeneous phase, but rather the carbon black remained somewhat clumped together, which was very obvious when stretched, as voids developed at the carbon pockets and small black rubber pieces crumbed off very easily (Figure 5. 4).

The inability to achieve a uniform particulate dispersion was due to the stiffness of the melted mixture and the high melting temperature. A high loading fraction was necessary to reach the percolation threshold and achieve conductivity, and the mixture became very stiff and hard to mix at 20 weight percent. A centrifugal mixer may be capable of handling this type of mixture, except that the temperature quickly falls below the polymer’s melting.
point when removed from the hot plate. Solvents can be used to reduce the viscosity of thermosetting polymers, but Ultraflex’s high melting temperature of approximately 175°C prohibits this, as it is well above the boiling points of solvents such as toluene (111°C) or acetone (56°C). A heated centrifugal mixer would possibly be able to successfully mix it, but this equipment is not present in the lab and is impractical, as commercial mixers generally are not capable of reaching these temperatures [55].

Figure 5.4: Voids evident in thermoplastic elastomer blended with carbon black when stretched

The summary of polymer suitability is given in Table 5.1, and includes the pure elastomer’s Shore hardness. Strictly speaking, Shore hardness is not a measure of stiffness, but is often more convenient, at least preliminarily, as softer elastomers are more compliant, and elastic modulus and strain at failure are not always listed in material datasheets like Shore hardness is. Shore A is the most common hardness scale for soft polymers, but supersoft polymers are rated on the Shore OO scale, which is lower than the Shore A scale.

The conclusions from this benchmarking and preliminary testing are that thermoplastic elastomers are impractical due to the high temperatures to melt them, and the best elastomers are platinum-cure silicones since they are the most compliant materials available. Platinum-cure silicones experience cure inhibition in the presence of certain materials, specifically carbon. Urethanes, specifically Vytaflex10 worked reasonably well and had less cure inhibition, but became hard and stiff at higher carbon loadings. A tin-cure
silicone rubber, specifically Oomoo30, would be the best material to use if carbon black is being used as the conductive filler. However, a platinum-cure silicone such as EcoFlex30 would be far superior, if a conductive particulate was found that didn’t cause cure inhibition.

<table>
<thead>
<tr>
<th>Polymer Name</th>
<th>Polymer Type</th>
<th>Elastomer Hardness</th>
<th>Elongation @ Break</th>
<th>Results and Observations From This Study</th>
</tr>
</thead>
<tbody>
<tr>
<td>Sylgard 184 PDMS</td>
<td>Platinum-cure silicone</td>
<td>Shore A-43</td>
<td>&lt; 200%</td>
<td>Cure inhibition with carbon</td>
</tr>
<tr>
<td>EcoFlex OO-30</td>
<td>Platinum-cure silicone</td>
<td>Shore OO-30</td>
<td>900%</td>
<td>Cure inhibition with carbon</td>
</tr>
<tr>
<td>Oomoo30</td>
<td>Tin-cure silicone</td>
<td>A-30</td>
<td>250%</td>
<td>Slightly stiffer than desired</td>
</tr>
<tr>
<td>Mold Max10</td>
<td>Tin-cure silicone</td>
<td>A-10</td>
<td>529%</td>
<td>Cure inhibition with carbon</td>
</tr>
<tr>
<td>Vytaflex10</td>
<td>Urethane</td>
<td>A-10</td>
<td>1000%</td>
<td>Cure inhibition at higher carbon loading</td>
</tr>
<tr>
<td>Vytaflex10 with So-Flex softener</td>
<td>Urethane</td>
<td></td>
<td></td>
<td>Cure inhibition at higher carbon loading</td>
</tr>
<tr>
<td>ClearFlex50</td>
<td>Urethane</td>
<td>A-50</td>
<td>500%</td>
<td>Very thin solution, particulate settles out. High stiffness, low tear strength</td>
</tr>
<tr>
<td>Medium Ultraflex</td>
<td>Thermoplastic Elastomer</td>
<td>OO-24</td>
<td></td>
<td>Too hot to mix; was very porous upon cooling (not curing) and “crumbed” apart</td>
</tr>
</tbody>
</table>

Table 5.1: Polymer suitability summary [40, 41, 54, 56]

5.2 Conductive Particulate Selection

Other conductive particulates that were investigated besides carbon black powder included Zoltek Panex30 milled carbon fiber, Alfa Aesar conductive grade graphite powder, and Nickel-based fillers. Carbon fiber resulted in cure inhibition issues in platinum-cure silicones, as expected, and the resulting samples in tin-cure silicones were fairly stiff. Graphite was found to need exceedingly high loading fractions to become even weakly conductive, and the resulting specimens were very heavy but tore apart very easily. The conductivities of the specimens made with these particulates are shown in Figure 5.5. None of the specimens met the goal equal or greater bulk conductivity compared to carbon grease [57]. The limiting factor for increasing weight percent carbon and carbon fiber was the specimens becoming very brittle, or stiff, from the reinforcing fillers.
Nickel-based conductive particulates, produced by Conductive Composites Company, were tested next. These products include nickel-coated carbon fiber (NCCF), referred to as Precision Converted Fiber or PCF by Conductive Composites, in Figure 5.6 [58], and nickel nanostrands, Figure 5.7 [59], which are nanoscale nickel slivers with extremely high aspect ratios. Both are made through Chemical Vapor Deposition (CVD). Though the NCCF contains a significant amount of carbon, the carbon is coated with nickel, essentially passivating it. Conductive Composites recommended using either 0.1 mm or 0.25 mm NCCF with a 20 weight percent Nickel coating, along with nickel nanostrands, in order to make a more conductive product. This was confirmed through testing, as using both NCCF and nickel nanostrands significantly reduced the percolation threshold and gave a much more conductive material than either product alone could give.
Additionally, the more compliant platinum-cure silicones were able to be used with NCCF, since the nickel coating prevents the elastomer from significantly contacting the carbon fibers, and therefore prevents cure inhibition. This simplified the elastomer decision to choosing the material with the lowest elastic modulus and highest elongation, which was EcoFlex30.

Figure 5.8 shows the preliminary data of conductivity vs. particulate concentration for unstrained samples. Some of the specimens with high filler concentrations exceeded the target, which was the level of conductivity of carbon grease. Since the conductivity will increase dramatically when the sample is strained, the nickel nanostrands and nickel-coated carbon fiber system will be a suitable conductive filler. (Specimens with filler combinations that had resistances too high to read with a handheld multimeter were not shown in Figure 5.8. Specimens with resistances that were too high to measure were deemed unusable, as their conductivities were approximately zero.) Because nickel has such high density,
5.8 plots both volume and weight percentages. Table 5.2 provides a summary of the results of particulate comparisons.

![Conductivity vs. Particulate Concentration](image)

Figure 5.8: Bulk conductivities of specimens made with nickel nanocomposites

<table>
<thead>
<tr>
<th>Particulate</th>
<th>Preliminary Conductivity (S/m)</th>
<th>Preliminary Results and Observations</th>
</tr>
</thead>
<tbody>
<tr>
<td>Cabot XC72 carbon black</td>
<td>0.26</td>
<td>Caused cure inhibition in platinum cure silicones - the lowest stiffness polymers</td>
</tr>
<tr>
<td>Zoltek Panex 30 milled carbon fiber</td>
<td>0.03</td>
<td>Caused cure inhibition in platinum cure silicones - the lowest stiffness polymers</td>
</tr>
<tr>
<td>Alfa Aesar graphite powder</td>
<td>0.01</td>
<td>Specimens had exceptionally low tear strength</td>
</tr>
<tr>
<td>Conductive Composites NCCF</td>
<td>0</td>
<td>Essentially non-conductive without nanostrands</td>
</tr>
<tr>
<td>Conductive Composites nickel nanostrands</td>
<td>1.6e-5</td>
<td>Essentially non-conductive without NCCF. Also cracked while curing and was fairly weak</td>
</tr>
<tr>
<td>NCCF + nickel nanostrands</td>
<td>3.6</td>
<td>Most conductive option. Max strain varies, some over 35%, some of which is non-recoverable.</td>
</tr>
</tbody>
</table>

Table 5.2: Conductive particulate summary for unstrained samples

Microscope images of specimens with nickel-coated carbon fiber and nickel nanostrands are shown in Figure 5.9 through Figure 5.11. Also visible in these images, is the
paint from the DIC speckle pattern, and evidence that the length of the nickel-coated carbon fibers appears longer than advertised by the supplier, Conductive Composites. The fibers appear as the long needle-like spindles, and the nanostrands, which are indistinguishable from the base polymer, give the remaining material a sponge-like appearance.

Figure 5.9: Microscope image of torn edge of specimen with 7 volume percent Nickel Nanostrands, 3 volume percent 0.25 mm NCCF

Figure 5.10: Microscope image of flat face of specimen with 7 volume percent Nickel Nanostrands, 3 volume percent 0.25 mm NCCF
Based on all of the preliminary testing of polymers and particulates, the most promising conductive filler was a combination of 7% nickel nanostrands and 3% nickel-coated carbon fiber by volume. Both 0.1 mm and 0.25 mm fibers were promising enough to continue testing. This caused no cure inhibition in platinum-cure silicones, which allowed EcoFlex30 to be used, a significant step toward a more stretchable final material. This polymer composite composition was used for the remainder of this work.

5.3 Uniaxial Stress-Strain Testing

Measurement data collected on the uniaxial specimens in this set of tests included specimen geometry, load cell data, optical strain data, and resistance across the specimen. As detailed in Section 4.5, the load cells recorded the force required to stretch the specimens, pictures of the specimen were taken at each load step and run through Ncorr in MATLAB to extract the strain values, and the resistance across the specimen was measured by a National Instruments my-DAQ. All values and images were recorded through LabVIEW for further analysis. An example of a uniaxial specimen being tested, showing the DIC speckle pattern, is in Figure 5.12.
5.3.1 Preliminary Tests

After selecting nickel-coated carbon fiber and nickel nanostrands as the conductive fillers, and Ecoflex OO-30 as the elastomer, several specimens were made and tested with varying volume fractions of the fillers. These mixtures were guided by the work of Johnson et al. [42], who had recorded volume resistivity vs. strain for several mixtures of Sylgard 184 PDMS with nanostrands and NCCF (20 weight percent nickel coating, 2 mm length). They had attempted to maximize what they defined as a “gauge factor”, the ratio of the change in resistance over the initial resistance of the specimen as it was strained, in order to find the best material for a piezoresistive sensor, which is a strain gauge where the resistance can be related to strains, at levels higher than conventional strain gauges are capable of. The authors strained them until failure but did not present stress-strain data, or results for specimens that were cyclically loaded.

5.3.2 Results Sought from Uniaxial Testing

In order to be used in a compliant sensor -whether the sensor output is based on resistance or capacitance- the material must have low stiffness, be capable of high strains, and be conductive. The resistance vs. strain results are the most important, as that is the relation that would be used by a piezoresistive sensor, and the ability to conduct is what would be of importance for a material used as a compliant electrode of a capacitive sensor. The stress-strain results and high-strain capability then determine the measurement range of a sensor made of that material, and what types of materials it is suitable to measure. For
example, a sensor with a stiffness higher than that of skin would not be able to accurately measure the flexing of a muscle or joint, since the presence of the sensor would affect the movement of the skin that it was attached to.

The questions to be answered are:

- What length of nickel-coated carbon fiber is optimal?
- Is it feasible to use the material as a piezoresistive sensor, or will hysteresis affect the resistance readings causing a difference between extension and contraction?
- Will cyclic loading cause changes in the material, that would appear as drift if used as a piezoresistive sensor?
- Is the material conductive enough to be used as a compliant electrode for a capacitive sensor?

5.3.3 Results – NCCF length

Tests were performed on specimens with the same compositions in terms of volume fractions, but with different lengths of nickel-coated carbon fiber, in order to find which fiber length gave better conductivity and compliance. In order to compare results across different specimens, resistance values are presented as conductivity (the reciprocal of resistivity), as resistance and conductance are dependent on the length of the specimen while conductivity (or resistivity) is a material property independent of specimen geometry. This conductance can be calculated using the original specimen dimensions, or the actual dimensions as the specimens are stretched. This is similar to true strain vs. engineering strain, as the cross-section shrinks and the specimen elongates. The “engineering conductivity” approach was chosen, as this would scale with the measurement being taken but still be independent of different geometries across specimens. “True conductivity” shows that the material becomes more conductive when the smaller cross-section is taken into account, but that doesn’t directly relate to the actual resistance measurements.

The 0.1 mm length NCCF clearly made a more conductive material than 0.25 mm NCCF. Both materials were made with 3 volume percent of NCCF, 7 volume percent nanostrands, and EcoFlex30, and the only difference was the length of the fibers. The material with 0.1 mm NCCF had an initial conductivity of 0.25 Siemens per meter in the tests
shown in Figure 5.13, reached 100 S/m by 23% strain, and 240 S/m before unexpectedly failing at a strain of 34%. The 0.25 mm fiber material had an initial conductivity of only 0.0002 S/m and maximum of 26 S/m at nearly 40% strain, and was stiffer, but was capable of higher strain. The 0.25 mm NCCF specimen was not strained until failure here, but had reached nearly 40% while the 0.1 mm NCCF specimen began to fail at 34% strain. A comparison of the stress-strain curves is shown in Figure 5.14. In the following figures, “nickel nanostrands” is abbreviated “NiN”.

![Conductivity Comparison of Specimens made with .1mm and .25mm NCCF](image1)

Figure 5.13: Conductivity of specimens made with 0.1 mm and 0.25 mm length NCCF, under cyclic loading (3 cycles shown)

![Stress-Strain Comparison of Specimens made with .1mm and .25mm NCCF](image2)

Figure 5.14: Stress-strain curves of specimens made with 0.1 mm and 0.25 mm length NCCF, under cyclic loading (3 cycles shown)
Specimens were also strained until failure, or until the tensile testing machine was fully extended, which was the case of the specimen with .25mm. These results are plotted in Figure 5.15 and Figure 5.16, and showed the maximum strain the specimens were capable of reaching, and the conductivities at those strains. The specimen in this test made with 0.1 mm fibers also withstood much higher strains than the previous test, indicating the failure in the previous test was likely due to a flaw in the specimen that initiated a tear earlier than otherwise expected. The small drops in stress seen in the stress-strain curve for the specimen made with 0.25 mm NCCF are due to tightening the grips during the test to prevent slipping. Though the clamps are tight at the beginning of the test, at high strains, the material has stretched enough that the grips begin to loosen, and the material can start to slide out. The small drops are points where the test was paused in order to tighten the clamps, and then the test resumed. Again, this specimen withstood over 120% strain and never failed.

![Conductivity vs. Strain to Failure for Different NCCF Fibers](image)

Figure 5.15: Conductivity vs. strain, tested to failure (0.25 mm NCCF specimen did not fail)
Figure 5.16: Stress vs. strain, tested to failure (specimen with 0.25 mm NCCF did not fail)

5.3.4 Results – Hysteresis and Cyclic Loading

The data revealed significant hysteresis loops in the stress-strain curves, as shown in Figure 5.17. The conductivity vs. strain curves did not have this effect as much, but conductivity did decrease as the specimens were cycled. This is likely due to the fact that the nickel nanostrands break as they are repeatedly stretched, making for a less effective conductive network within the material. For a sensor, this means the values would be continually drifting, preventing a specific resistance reading from being able to refer to a specific strain since the strain vs. resistance relation is continually changing. Interestingly, the conductivity of the specimens made with 0.25 mm length NCCF seemed to decrease more slowly than that of the specimens made with 0.1 mm length NCCF. Figure 5.18 shows this effect for 0.1 mm NCCF and Figure 5.19 shows it for 0.25 mm NCCF.
Figure 5.17: Hysteresis effect in stress-strain curves

Figure 5.18: Conductivity decreasing as specimen is cycled in specimens with 0.1 mm NCCF
Since the conductivity drop is likely due to the nanostrands breaking with increased cycling, it is possible that the conductivity vs. strain curve will eventually stop changing, as all the nanostrands are broken. Another way that the nanostrands are broken is through the mixing process during specimen creation. This reveals itself in significantly reduced conductivity, as shown by the results of Hansen et al. [60] in Figure 5.20, showing the conductivity normalized against the conductivity seen with a mixing time of 60 seconds. This data is from specimens made of Minwax® Polycrylic® acrylic/urethane, but the trend is likely the same for elastomeric composites made using any other base polymer. Given that the specimens in this work are mixed for approximately 30 seconds, it appears as though the resistance vs. strain curve would continue to drift until the conductivity had dropped by an order of magnitude, at a minimum, as the trend in Figure 5.20 does not appear to show signs of approaching an asymptote.
To test the effects of nanostrand breakage in the mixing process, the material for a specimen was intentionally overmixed, similar to the process in Hansen’s study. This specimen was mixed in the Thinky AR-310 mixer for 500 seconds instead of 30 seconds, and then cycled to see if the conductivity was still dropping.

Figure 5.21: Cycling of intentionally overmixed specimen
Figure 5.21 shows that the conductivity was similar to the normally mixed specimen in Figure 5.19 at higher strain. At lower strains, the specimen actually had higher conductivity, meaning that the increased mixing time may have led to better dispersion of the conductive particles that was more significant than the nanostrand breakage.

The trend of conductivity decreasing with cycling, however, continued unabated after the increased mixing. In light of Figure 5.20 and Figure 5.21, it appears as though it would take a considerable number of cycles before the conductivity stopped changing. If the specimens were in fact cycled until the nanostrands stopped breaking, this would resemble a specimen that was made with regular nickel nanopowder instead of nanostrands. Nickel nanopowder is much more commercially available and much less expensive than the nanostrands, so if this approach were pursued, it would make sense to start with NCCF and nickel nanopowder. A nickel nanopowder with dendritic morphology is also available from Alfa Aesar [61], which should lead to fairly high—not as high as nanostrands—conductivity given its high aspect ratio. The nanopowder is not expected to be as brittle as nanostrands. It also costs on the order of $100 per pound, instead of several hundred dollars per pound of nickel nanostrands. The resulting conductivity would likely be lower than with nanostrands, but certainly higher than other systems such as carbon black, and hopefully not experience the cyclic degradation seen with nanostrands. One of the keys remains to use two conductive phases, preferably with high aspect ratios, one larger (the nickel-coated carbon fiber), and one smaller to bridge the gaps (the nanostrands or nanopowder) between the larger phase.

It is also possible that the drop in conductivity is due in part to localized plastic deformation in the elastomer, resulting in voids. In order to determine the exact reason, “damaged” specimens would likely have to be examined beneath an SEM (scanning electron microscope), to see what type of damage is occurring at the microscale level.

5.3.5 Results – Conductive Enough for Electrode for Capacitive Sensor

As noted previously, the material with 0.1 mm NCCF had an initial conductivity of .08 S/m, reached 100 S/m by 23% strain, and 240 S/m before failing at 34% strain. Conductive carbon grease, by comparison, has a conductivity of 1.21 S/m, and is a grease so
its conductivity doesn’t change as it is strained. The conductivity of the nickel-based nanocomposite with 0.1 mm NCCF increased to a level higher than carbon grease in the neighborhood of 7.5% strain, after three cycles to 23% strain and back, as shown in Figure 5.22. Similarly, the material with 0.25 mm NCCF varied from 0.0002 S/m to 26 S/m at just under 40% strain. Its conductivity surpassed carbon grease by 25% strain on the second cycle, but had degraded to require 34% strain by the tenth cycle, as shown in Figure 5.23.

Figure 5.22: Conductivity comparison of material with 0.1 mm NCCF with carbon grease

Figure 5.23: Conductivity comparison of material with 0.25 mm NCCF against carbon grease
The conductivity of the material with 0.1 mm NCCF is certainly sufficient at higher strains, as it is higher than the currently used material. Whether or not it would be sufficient at lower strains is likely application specific.

5.3.6 Comparison with Prior Work

The set of data found in the literature that was most similar to data gathered through this work was generated by Johnson et al. [42]. Data is presented as resistivity, in units of Ω-cm, as those are the units used by the other authors. Figure 5.24 and Figure 5.25 were generated by converting the data to resistivity in the proper units, and using Johnson’s published data as the background of the graph.

![Resistivity vs. Strain - Comparison with Prior Work](image)

Figure 5.24: Resistivity comparison of material with 0.1 mm NCCF, against prior work by Johnson with 2 mm NCCF [42]
The testing performed by Johnson did not include a specimen with the same volume percentages used in this work, and they used Sylgard 184 PDMS instead of EcoFlex30 and 2 mm NCCF, instead of 0.1 mm and 0.25 mm. Also, their stated objective was to maximize the change in conductivity, as opposed to simply maximizing conductivity, in order to investigate using the material as a resistive strain gauge based on its piezoresistive response. Finally, since the results from the current study have clearly shown that resistivity increases with repeated cycling, current study results are plotted three times in each figure for comparison: 1st cycle results, additional cycle results, and test to failure results.
Interestingly, Johnson’s measured resistivity with 2 mm NCCF was lower at low strains than the specimens tested in the current work with the 0.25 mm NCCF, in Figure 5.25. Their specimens also all failed at lower strain levels. Consistent with expectations, that the resistivity of the material made with 0.1 mm NCCF (Figure 5.24) is much lower than the material made with 2 mm NCCF.

Figure 5.24 and Figure 5.25 also each show data from only one individual specimen tested in this study. It can be seen in both specimens that at a given strain value, resistivity was higher in each subsequent cycle. What it is interesting however, is that when a later cycle is straining a specimen farther than it had ever been strained before, when the strain of the later cycle approaches the maximum strain that the specimen had previously seen, that the conductivity approaches its previous maximum as well. This can be best seen at approximately 33% strain in Figure 5.24, and approximately 56% strain in Figure 5.25.

5.3.7 Summary of Uniaxial Testing

The uniaxial testing provided the answers to the questions posed prior to testing, listed below.

- Optimal NCCF length
  - The optimal fiber length for high conductivity is .1mm, which is the shortest length that Conductive Composites Company currently manufactures
  - The optimal fiber length for high strains for .25mm, as specimens made with it reached the limits of the machine without failing at 120% strain
- Hysteresis
  - The conductivity didn’t appear to be significantly different between extension and retraction
- Cyclic loading and effect on conductivity
  - Conductivity was largely dependent on cyclic loading, and continually decreased as the specimens were cycled due to “cumulative damage”,

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suspected to be due to nanostrands breaking. This will require future work to verify.

- **Suitability for a DEAP conductive electrode material**
  - In specimens made with 7 volume percent nanostrands and 3 volume percent 0.1 mm length NCCF, the conductivity surpassed that of conductive carbon grease around 7.5% strain, peaking at 275 S/m. This is clearly conductive enough once it surpasses the conductive carbon grease.
  - In specimens made with 7 volume percent nanostrands and 3 volume percent 0.25 mm length NCCF, the conductivity didn’t surpassed conductive carbon grease until approximately 28% strain, and peaked near 45 S/m. This is not as viable as material made with the shorter fiber length, unless higher strain capability is required.

Additionally, the material was compared to previous work and found to be comparable given the discrepancies in specimen materials and compositions. Each material tested in the current work maintained its advantage over the prior study; material with 0.1 mm NCCF was much more conductive, and material with 0.25 mm NCCF was capable of much higher strains.

5.4 Equibiaxial stress-strain testing

5.4.1 Specimen configuration

The equibiaxial specimen configuration used for this study was based on the optimal one found by Smoger [20], a Clamped Cruciform Specimen with a 4 mm fillet radius, and shown below in Figure 5.26. The specimen was tested with exposed legs, instead of having the clamps come all the way to the base of the legs, in order to be able to track the uniaxial strain during testing.
5.4.2 Hysteresis and Equibiaxial Characterization

This test of EcoFlex30 with 7 volume percent of nickel nanostrands and 3 volume percent of 0.25 mm nickel-coated carbon fiber exhibited a large amount of hysteresis, and, therefore, the stress-strain curve is path dependent and the material doesn’t have a singular stress-strain curve. Also, the test stopped at approximately 8.3% equibiaxial strain, as the material started to tear at the inside radius of the specimen between the legs. This area experiences higher stress, and the rest of the specimen could have been strained much more.

![Figure 5.26: Equibiaxial specimen being tested](image)

Figure 5.26: Equibiaxial specimen being tested

![Figure 5.27: Stress-strain curve during equibiaxial test, showing hysteresis](image)

Figure 5.27: Stress-strain curve during equibiaxial test, showing hysteresis
This is shown in Figure 5.27. The stress-strain curve drops slightly with each subsequent cycle. Since the sample exhibits hysteresis and is not stabilized, an elastic stress-strain characterization is not applicable, as there is no definitive curve that a material model of that type can be fit to at the present time.

5.4.3 Variable Strain Ratio

For the specimen shown in Figure 5.26, strains were measured in Ncorr in the equibiaxial region of the sample as well as the in the legs of the sample. The equibiaxial strain in the central diamond region could then be compared to the uniaxial strain in the legs, and the ratio of those values found at each step in the test. This ratio averaged .59, compared to the stress decay factor (SDF) of .77 that was expected based on Smoger’s work [20]. This is a strain ratio instead of stress ratio, but the two values should be approximately the same. The more significant finding, was that the strain ratio varied from .48 to .85 depending on the level of applied strain (0-14.8% uniaxial strain in the legs, corresponding to 0-8.3% equibiaxial strain in the central diamond region). This supports the argument about a variable SDF made in Section 4.3, Variable Stress Decay Factor. As stated previously, this will need to be the subject of future work.

5.4.4 Energy loss in the material

Lastly, the energy loss per cycle was estimated using the area enveloped by the hysteresis loops, in order to compare to published results on the energy produced by DEAP energy harvesters. This was done by numerically integrating the stress-strain curve to calculate the difference in areas under the extension and retraction portions of the curve, or the difference in energy absorbed vs. released. This was done over the three cycles in Figure 5.27 and then averaged.

In order to arrive at the equibiaxial stress-strain data to do this, the stress decay factor approach was used for this approximation because, on inspection of Figure 5.27, the extension and retraction portions of the curve (not the point of load direction change) are fairly close to linear. The data isn’t being made linear or given a linear fit, this is only a
justification for using the SDF, for this particular set of data. The linear assumption also
does not necessitate the assumption that the extension and retraction parts of the curve are
co-linear. The two parts can have different slope, as changing the slope scales the stress
values together and the ratios between them (the SDF) remains constant. Thus, using the
SDF is not as inaccurate an assumption as it was for the highly nonlinear Ultraflex data
discussed in Chapter 4.

The average strain ratio of 0.59 was used as the SDF. The stress in the specimen legs,
as calculated from load cell measurements, was multiplied by this SDF to approximate the
equibiaxial stress in the central diamond region. The stress-strain curve to be analyzed then
consists of the experimental equibiaxial strain data on the x-axis, vs. the approximated
equibiaxial stress on the y-axis. This is shown for one cycle in Figure 5.28. The higher
curve is the original data. Multiplying the stress by the average strain ratio, or approximate
SDF, gives the approximated equibiaxial stress. The area inside the lower curve then
represents energy losses to be calculated.

![Equibiaxial Stress vs. Equibiaxial Strain](image.png)

Figure 5.28: Stress-strain curve during equibiaxial test, showing hysteresis

Performing numerical integration, and normalizing by the specimen volume, gives an
energy loss per unit volume of just over 1234 Joules/m³ when strained up to the 8.3%
equibiaxial strain and then unloaded, quasi-statically. This value applies only to the central diamond region, meaning it is only valid for equibiaxial conditions.

Values were then calculated from the results published by Clarke et al. [21], which were also under equibiaxial conditions and presented in Section 2.2.5. Clarke’s DEAP generator consisted of a DEAP with a center dielectric layer that was only 40 millimeters in diameter and .5 millimeters thick, with conductive carbon grease electrodes. Using their published values for energy density and the dimensions, the energy output was calculated to be 1.43 Joules when cycling to 440% strain and back. Incorporating the stated efficiency of 27%, the mechanical energy input to the generator is approximately 5.3 Joules. Arbitrarily normalizing on a “per 100% strain basis”, the work input to the system would be 1.21 Joules, with .33 Joules of electrical energy out.

Should the nickel nanostrand and nickel-coated carbon fiber elastomer material be used as conductive electrodes on either side of Clarke’s DEAP generator, with the same .5 millimeter thickness and diameter as the center dielectric layer, the energy loss would be approximately .0093 Joules per layer, again normalized for the same 100% strain cycle. This lowers the efficiency from 27%, to 25.4%. This is strictly the percentage of electrical energy recovered from the generator compared with the amount of energy put into it.

It is worth noting that the Clarke study claimed a far, far higher energy efficiency (27%) than previous studies (7.5%) [62], so that should be kept in mind when looking at this comparison. A decrease of 1.6% out of 7.5% efficiency is much more significant than 1.6% out of 27% efficiency. The values for the nickel-based nanocomposites were also based on data taken from tests that were quasi-static, while Clarke’s DEAP was being cycled at .5Hz. The strain range for a DEAP with the new electrodes would be much smaller than the range for their DEAP with conductive carbon grease, since sample failures were observed in this study at strains as low as 34% for one specific trial, though the 0.25 mm NCCF material strained 120% never failed. Overall, the efficiency decrease and limitations may be an acceptable compromise for a solid electrode instead of a conductive grease layer for certain applications. One prominent concept for implementing a Dielectric Energy Generator is
putting a DEAP in a shoe sole [63]. Different configurations require different amounts of strain, but they are generally less, and within the range of the new materials. Solid electrodes, in fact, may be able to alleviate issues with the carbon grease electrode layers trying to rupture.
6. Conclusions and recommendations for future work

6.1 Conclusions

The result of this work is an assessment of new conductive elastomer materials for uses in Dielectric ElectroActive Polymers, and as piezoresistive strain gauges. It is a solid, conductive elastomer, composed of a platinum-cure two-part silicone base polymer, with two types of nickel-based conductive particulates: nickel nanostrands and nickel-coated carbon fiber. It is superior to current DEAP electrode layer materials in that it is self-contained, and not a messy grease or powder. This makes DEAPs much more useful for practical purposes outside the laboratory, but the materials also have other drawbacks.

The material evaluated in this study exhibits large amounts of hysteresis in its stress-strain curve, which partially reduces its usefulness as an electrode for DEAP energy harvesting applications, as the area enclosed by the hysteresis loops is the energy dissipated by the material each cycle. The benefits of having solid elastomeric electrodes may outweigh the drop in efficiency due to this energy loss, however.

The largest target applications of these materials, piezoresistive strain sensors, is not practical with this exact mixture, but changing one of the components may make it viable. The idea behind a piezoresistive strain sensor is that stretching the material causes the conductive fillers to line up, reducing the distance that electrons must travel through the nonconductive polymer matrix in order to traverse the length of the sensor. The resistance of such a sensor then drops by several orders of magnitude, from megaohms to tens of ohms, as the sensor material is strained. Mapping the resistance measurement back to strain is the foundation for a high-deformation strain gauge, but this material is not repeatable in that the response changes as it’s cycled. High strain levels are hypothesized to gradually break the high aspect-ratio, fragile nanostrands, and the resistance curve steadily rises with the number of cycles. If a conductive powder that didn’t break like nanostrands were to be used, possibly dendritic nickel nanopowder, this problem might be eliminated.
The materials are very conductive once modest strain is achieved, and there is the possibility of using them as conductive electrodes for capacitive strain sensors, sensing the change of capacitance of a DEAP, instead of the change in resistance in a single layer. For a capacitive sensor electrode, the value of the conductivity isn’t as critical, as long as the material is in fact sufficiently conductive in the intended operating range. The current material combinations tested had lower conductivity than conductive carbon grease until a minimum of 7.5% strain was reached, but reached over 200 times the conductivity at higher strains.

The work included augmenting an equibiaxial tensile testing machine in order to characterize conductive materials, by electrically isolating the specimens and implementing resistance measurements. The optical strain measurement method was also substantially improved, by implementing Digital Image Correlation using Ncorr.

Progress was also made in understanding the equibiaxial characterization process. The current characterization method uses a stress decay factor, the simulation-found ratio of stresses in the equibiaxial central diamond region compared to the nominal uniaxial stress in the leg of the specimen. This was found to be a flawed approach since that value does not remain constant as a specimen is loaded.

Overall, the goals of identifying and characterizing a conductive elastomeric material for use as a DEAP electrode were accomplished. Augmentation of the equibiaxial tensile testing machine was completed in order to take this data, and comparisons were made relating the results to several DEAP applications. The equibiaxial characterization method was not validated, but a flaw in the method was found with steps identified to correct the method.

6.2 Recommendations for future work

Recommended future work includes continuing to investigate using nickel nanocomposites as DEAP materials and as piezoresistive sensors, incorporating nickel nanopowder. This thesis found the stress-strain and conductivity-strain results for elastomers
made with nickel-coated carbon fiber and nickel nanostrands, but nickel-coated carbon fiber and nickel nanopowder appear to be a more promising combination for most of the applications noted above in section 6.1. Nickel-coated carbon fiber with a higher nickel coating level could also be tested, as the fibers that were tested in this work all had a 20% nickel coating by weight, and other coating levels are also available.

Since the addition of conductive particulates makes a resulting nanocomposite stiffer, it becomes a tradeoff of conductivity vs. compliance. Using the above mentioned material combination, work could be done to determine the optimal point where the electrode is conductive, and balance the conductivity with material stiffness and required layer thickness to keep the overall DEAP sensor or energy harvester as compliant as possible. Given that the conductivity increases exponentially with conductive filler concentration in the region of the percolation threshold, this will likely be the highest filler concentration that allows for sufficient strain, and the layer can then be made only as thick as necessary.

Replacing DEAP layers with different materials, whatever they may be, will alter the characteristics of the final DEAP device. Certain attributes such as DEAP capacitance can be calculated analytically provided that the dielectric constants of the constituents in the different layers and their mass fractions are known, but DEAPs should be made and tested with solid electrodes. This is particularly true given the fact that conductors, by definition, have infinite dielectric constants because they cannot support an electric field within them since electrons are free to move, and the nickel containing layers are essentially a blend of conductors in a dielectric matrix. Testing the entire DEAP would also mean testing at the device level rather than at the material level, and would move a DEAP with solid electrodes –for whichever application is targeted– much closer to reality.

Another means to improve DEAPs may be to improve upon the principle of the StretchSense capacitive sensor [38] using different materials. The sensor appears to be made of a silicone rubber and conductive carbon grease, with two carbon grease layers sandwiching a layer of rubber to create a classic DEAP, all enveloped in another protective layer of rubber. It was observed through other experimentation in the lab that the elastomer used to make the StretchSense had comparably low tear strength, and the StretchSense
sensors broke fairly easily. It may be speculated that a polymer with lower tear strength was used because of constraints imposed by the carbon grease. In other words, platinum-cure silicones may have been ruled out, in order to avoid cure inhibition from the carbon. This is the exact reason that nickel-based conductive fillers were found for this work, and it may be possible to use them in a grease form as well, as shown in Figure 6.1.

![Figure 6.1: DEAP sensor configuration with nickel nanocomposite electrodes](image)

Replacing the conductive carbon grease with a grease loaded with nickel-coated carbon fiber and nickel nanostrands or dendritic nanopowder would allow the platinum-cure silicones with higher tear strength to be used, resulting in more durable sensors. This would also likely resulting in higher electrode conductivity given the higher aspect ratios of the nickel-based fillers. No conductive nickel grease like this is currently known to be commercially available, though there are silver greases that have extremely high conductivity (greater than 10,000 S/m) [64]. It should also be noted, that in other experiments in our lab, EcoFlex 30, a platinum-cure silicone and the same elastomer used in this work, was in fact found to cure when up against carbon grease, but it is unclear if the elastomer immediately contacting the carbon grease cured or if there was a thin uncured layer. The StretchSense sensors are only a single millimeter in total thickness, so a thin uncured layer at each interface between the elastomer and carbon grease may be significant.

Alternately, the grease layer could be replaced by a thin layer of pure nickel nanocomposites (nickel-coated carbon fiber and/or nickel nanopowder or nanostrands), again with the goal of having excellent in-plane conductivity and eliminating the presence of grease with a solid electrode. Given that the pure layer of nickel particles would be extremely conductive, the electrode layer would be very thin, and should maintain a low overall stiffness.
In the field of equibiaxial characterization, the method with which equibiaxial stress data is obtained can be improved. This would most likely involve incorporating a variable stress decay factor, which could lead to iteratively improving the equibiaxial stress data, stress decay factor, and material model, or possibly incorporating the variable stress decay factor into the material model itself.
7. References


[61] Aesar, A., 2015, "7440-02-0 | Nickel Powder, -325 Mesh, Typically 99.8% (Metals Basis) | 43214 | Alfa Aesar| Alfa Aesar."


A. Appendix A – Machine Operation Instructions

Programs required:

- LabVIEW, must be 32-bit version
- National Instrument Vision Acquisition System (required to interface with camera through LabVIEW)
- National Instruments ELVIS driver package for use with the NI myDAQ
- Anaheim Automation SMC60WIN (compatible with newer operating systems despite manufacturer stating otherwise)
- PixeLINK Capture OEM

Devices being interfaced with:

- Anaheim Automation DPY50611 motor controller, via USB. Wiring connections are shown in Figure A-1.
- (2) Interface SML-10 10-pound load cells with Interface SGA amplifiers, through National Instruments SCB-68 terminal board, via National Instruments PCI-6024E DAQ card in the computer
- PixeLINK PL-E531MU monochrome imaging camera with Fujinon 1:1.4/12.5 mm HF12.5HA-1B lens, via USB
- National Instruments myDAQ, via USB

At the start of the test, use SMC60WIN, Figure A-2, produced by Anaheim Automation, to move the motor until the specimen is taut. If running a biaxial test, make sure the legs all have equal tightness. If one leg is tighter than the others, the stress and strain won’t be equibiaxial, and the clamp(s) need to be loosened to adjust the specimen.
The quality and orientation of pictures being taken by the camera can also be checked and adjusted within the Capture OEM software produced by PixeLINK, Figure A-3. If the camera is recognized by the computer, it should appear in the Camera Select dropdown. Pressing Video Preview from the Video Capture tab gives a live feed. SMC60WIN and PixeLink Capture OEM must be closed before running the LabVIEW VI since only one program can interface with a device at a time.
Figure A-3: PixeLINK Capture OEM program to view camera feed

The test rig is operated by the program Biaxial Membrane Tester with Image Grab and DMM.vi, the front panel of which is shown in Figure A-4. Before running the machine, the desired motion parameters are entered on the Configuration tab in the Biaxial Membrane Tester with Image Grab and DMM.vi. The motor controller is currently set to control the stepper motor with the smallest step size, so if those settings are changed, then the move increment will have to be lowered. The Filename Prefix is also important. Ncorr expects images with a naming convention of Name_####.tiff. The relevant portion of that is that there must be a name (numbers are fine as well), then an underscore, and then the image number. The LabVIEW program automatically appends the underscore and the image number to the file, the user just has to not use any spaces or underscores in the “Filename Prefix” field. Other image formats are also acceptable, though some formats will cause a loss of image quality which may have an effect depending on the resolution and accuracy required.
Once all parameters are set, a dialog box will appear asking where to save the data when the program is started. After telling it where to save the files, the testing may be monitored from the Acquisition tab, shown in Figure A-5.
Figure A-5: Acquisition tab on front panel of main VI

DMM_Voltage.vi, shown in Figure A-6, may also be opened in order to more directly monitor the resistance readings being taken by the NI myDAQ, as the readings are only sent to the main VI if they’re in range and going to be logged. Everything is still controlled from the main VI, however. If the specimen resistance over 20 MΩ, the program won’t proceed until it has an in-range reading (so that it will stop if the specimen fails or the multimeter lead comes off), so a wire has to be put across the multimeter terminal to let it take a reading and proceed to the next step.
If the load readings start looking like Figure A-7, it likely means the specimen is slipping and the clamp needs to be tightened, or less likely, that the specimen is starting to fail. Both slipping and the beginning of tear are usually visible. Generally, the clamps can’t be tightened enough at the beginning of the test, and after applying some load and having the clamped material stretch thinner, clamps become loose. The test can be paused simply by taking one the multimeter leads off (the program continues to check the resistance endlessly until it gets an In-Range reading), tightening the clamp again, and putting the multimeter lead back on.
Figure A-7: Slipping of specimen in clamps
B. Appendix B – Image Analysis Process in Ncorr

Image analysis using Ncorr is fairly straightforward. After collecting the images during the test run, and installing the Ncorr software (instructions are available online [51]) and adding it to the MATLAB file path, “handles_ncorr = ncorr” is typed into the MATLAB command window (without the quotes).

The first step is Load Reference Image. This is the image that the other images are going to be compared against to determine how much the specimen has moved in future images. Next, under Load Current Image(s) >> Load Lazy, load all the images including the reference image (this makes it clear what the first image is, when viewing a GIF of that the program can later make, which endlessly loops). Next, define the Region Of Interest. Region Of Interest >> Set Reference ROI >> Draw ROI, and draw the region of the specimen that you want to analyze. For the analysis shown in Figure B-1, the area near the clamps was excluded, in order to stay away from edge effects.

![Figure B-1: Creating Region Of Interest](image)

The next step, Set DIC Parameters, is the step where adjustments are made between accuracy and computation time, aside from the amount of area selected. The two parameters that can be adjusted are Subset Radius, and Subset Spacing. The values used for this work
were 18 for Subset Radius, and 3 for Subset Spacing, though the optimal choices for these parameters will vary with the quality of the speckle pattern and required accuracy, etc. The criteria for optimizing them, is making the analysis settings coarser until it starts producing inaccurate results. These values worked well and little effort was put into making the analysis run faster. Also, be sure to check the Enable Step Analysis and Auto-Propagation under High-Strain Analysis, as this allows the program to update the Reference Image. If the specimen begins to tear during the test, also enable Subset Truncation, as this will prevent the program from calculating strain across a crack.

Figure B-2: DIC parameter selection

Select Perform DIC Analysis. When prompted to seed the images, select an area where higher strain is experienced, making sure that the seed point never leaves the field of view. It should also be near the center of the region, if possible. After the images are seeded, wait for the new window appear. The ease with which the program achieved
convergence is shown at the bottom right. After clicking Finish, the program will continue and complete the analysis. Then Format Displacements, and Calculate Strains, both are fast. View Strains to see the chosen strain, as in Figure B-3, and select File >> Create GIF. You can also set the maximum and minimum values on the image and Apply To All, for better GIFs, and set the time delay between frames to .2 seconds as well.

Then, run the additional MATLAB script called Ncorr_postprocessToGetStrains.m. This code averages the strain values of the entire Region Of Interest for each load step, and returns an array of average strain values over that region with a single value in the array for each load step. This strain data can then be plotted against the stress at each step, etc.
Lastly, be sure to save the DIC data before closing Ncorr, so that the data can be reloaded in the future without performing the DIC analysis again.

%% Code to process DIC data into a strain array with a single value for each load step

numberofimages = length(handles_ncorr.data_dic.strains);  % Ncorr stores all data in larger structures. I.e., strains is a matrix within data_dic, which is within handles_ncorr

for i=1:numberofimages
    exx_ave(i) = mean(handles_ncorr.data_dic.strains(i).plot_exx_ref_formatted(~~handles_ncorr.data_dic.strains(i).plot_exx_ref_formatted));  % Average data and put into array
    eyy_ave(i) = mean(handles_ncorr.data_dic.strains(i).plot_eyy_ref_formatted(~~handles_ncorr.data_dic.strains(i).plot_eyy_ref_formatted));
    exy_ave(i) = mean(handles_ncorr.data_dic.strains(i).plot_exy_ref_formatted(~~handles_ncorr.data_dic.strains(i).plot_exy_ref_formatted));
end

exx_ave = exx_ave';  % Transpose x-direction strain data into a column vector
eyy_ave = eyy_ave';  % Y-direction strain data
exy_ave = exy_ave';  % XY shear strain data