Active Fiber Composites for Structural Actuation

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Aaron Alton Bent

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Signature of Author

Aaron Alton Bent

Department of Aeronautics and Astronautics
January 31, 1997

Certified by

Professor Nesbitt W. Hagood
Thesis Committee Chairman, Associate Professor, Department of Aeronautics and Astronautics

Certified by

Professor S. Mark Spearing
Assistant Professor, Department of Aeronautics and Astronautics

Certified by

Professor Michael J. Cima
Professor, Department of Material Science

Accepted by

Professor Jaime Peraire
Chair, Graduate Office

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Aaron A. Bent

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The performance of a number of aerospace applications could be greatly improved by the introduction of active materials to the task of vibration suppression. However, traditional actuator/sensor technology has precluded the widespread use of active materials in applications due to limitations in robustness to damage, actuation capability, and difficulty in integrating large systems of actuators into structures. A new concept for the actuation and sensing of structures, Active Fiber Composites, was introduced and explored in this thesis. This material incorporated unidirectional PZT fibers into a hybrid matrix, producing a highly conformable and anisotropic actuator material. A separate electrode layer with an interdigital pattern provided the primary piezoelectric effect in the direction of the fibers, and electrical connections which remained insensitive to damage at the ceramic level. This thesis followed a natural progression in the development of this technology, beginning with an investigation of component materials, and ending with the integration of these composites into structures.

A number of methods for characterizing bulk materials were developed with a particular emphasis on metrics for structural actuation. A linear micromechanics model based on a coupled field generalization of the Mechanics of Materials approach was developed to predict the effective properties of the composite, and validated with the use of a commercial finite element code. The utility of this model was demonstrated in the design of active fiber composites by examining the change in composite properties as a function of the bulk constituents. The ease of processing, performance, and operational risk were weighed for different composite geometry, PZT fiber materials, and matrix compositions.

A method for repeatable manufacturing of active composites was developed using a hand lay-up procedure for batch processing. Composites were characterized in low and high field regimes, and for free and laminated stress conditions. Low field composite properties showed excellent agreement with the models as a function of varying ceramic fraction, and for a variety of different fiber materials and matrix compositions. High field performance demonstrated that these new materials had significantly higher actuation capability than monolithic piezoceramics for planar actuation. In order to demonstrate their use in structures, these composites were applied as the actuators in two programs for rotorblade control: a trailing edge flap actuator and an integral twist blade spar. These applications demonstrated the merit of active fiber composites for structural control, and illustrated several important issues of structural integration and operation.

Thesis Supervisor: Prof. Nesbitt W. Hagood, Ph. D.,
Associate Professor of Aeronautics and Astronautics
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Chapter 1

Introduction

1.1 Motivation

Actuators and sensors are identified as one of the critical component technologies for the advancement of intelligent structures (Crawley, 1992). Although intelligent, smart, active, controlled and adaptive are often used interchangeably, a useful paradigm is the one presented by Wada et. al. (1989). This identifies intelligent structures as a subset of all other types of structures, capturing the highest level of complexity and functionality of system components. Intelligent structures are those that contain 'actuators and sensors that are highly integrated into the structure' and 'control logic and electronics that provide the cognitive element of a distributed or hierarchic control architecture'. Thus, they can sense and respond to stimuli within their environment. An example might be self repairing structures: structures that can sense and locate damage, and act to either repair or minimize the effects of the damage by redirecting stresses through undamaged areas. Although discrete portions of each of these exist, at present, the combined technology does not exist for this level of 'intelligence'. Actuators and sensors are not yet suited to highly distributed actuation and sensing, nor does the current control and processing allow the level of interaction needed for such applications.

This work deals with the development of actuator and sensor technology. In particular, this thesis focuses on active materials as actuators and sensors for control of structures. Aerospace applications are a particularly demanding group of structures, and place significant constraints on these materials associated with performance and robustness. The applications that follow not only demonstrate the benefits and feasibility of active materials to realizing significant improvements in performance of aerospace applications, but also show the limitations of the current active materials. Three areas of application are discussed below: aeroelastic control of fixed wing aircraft, aeroelastic control of rotary wing aircraft, and acoustic control of fuselage structures.
Applications of Solid State Actuator Materials

Active materials have been investigated extensively for the active control of **aeroelastic structures**, both rotary wing and fixed wing. In both cases, the emphasis has largely been in active vibration suppression, as opposed to static shape control. For fixed wing craft, the potential benefits include larger flight envelopes through extending the flutter boundary, increased aircraft life through reduced wing root loads, and improved passenger comfort. Early work employed the available control surfaces (flaps), but was limited by the slow response of hydraulic and electric actuators.

The first example of the application of active materials to this problem was performed at MIT (Lazarus et al., 1991), and involved bonded piezoceramic actuators to an aluminum plate. Several researchers (Ehlers and Weisshaar, 1992) showed that significant improvements in actuation authority could be realized by employing host structural coupling. Lazarus and Crawley (1992) utilized bend-twist coupled graphite/epoxy plates to demonstrate 30, 10 and 20 dB reductions in the first three modes (respectively) from an initial 1% structural damping. Relatively few large scale demonstration programs have been carried out. One experiment of significant size did show the performance feasibility of large scale aeroelastic control (Lin and Crawley, 1995). A 5 foot span wing with 50% piezoceramic coverage was tested in the Langley Transonic Dynamics Wind Tunnel. Significant vibration suppression was achieved using modern control methodologies, reducing the first mode response by an order of magnitude and extending the flutter boundary by 12%. This experiment was significant for another reason - it demonstrated the difficulties of current active material technology applied to large structures. It highlighted difficulties in implementing dozens of individual actuators, and the need for almost total coverage in order to achieve useable performance do to the limited authority of current actuators.

Application of active materials to **aeroelastic control of rotorblades** provides similar benefits, with perhaps greater payoff: reduced operator fatigue, a reduction in acoustic signature and overall noise, significant increases in payload, faster cruising speeds, and reduced maintenance. High levels of vibrations are caused by unsteady aerodynamic loads acting on the rotor system, particularly in forward flight. These vibrations could be greatly reduced by altering the twist distribution of the blade in flight as a function of the azimuth angle (Derham and Haggood, 1996). The approach centers around controlling the pitch of each blade, independent of the collective pitch needed for hovering/forward flight control. Like fixed wings, early work employed the available actuators (swash plate), but was limited by the low bandwidth and inability to control the blades independently (see du Plessis, 1996, for an overview of this subject).
The earliest use of active materials to enable individual blade control (IBC) employed a piezoceramic bender which actuated a trailing edge flap (Spangler and Hall, 1990). This work illustrated the need to consider the efficiency of actuator/linkage designs in order to permit sufficient authority at higher rotational speeds, where aerodynamic forces were greater. Since then, additional work with improved load transfer mechanisms and high force stack actuators has demonstrated the feasibility of rotorblade control at reduced scales (Hall and Prechtel, 1996). A set of general requirements for active servo-flap control on realistic rotorblades has been compiled by researchers at the University of Maryland (Giurgiutiu, et al., 1994). These include the needed flap deflection (±5°), total power consumption (10-12 kW), and actuator weight budget (10 kg/blade).

The use of actuators integral to the blade to generate full blade twist is an alternative approach to control. In this case, the actuators must operate against the torsional stiffness of the blade itself, since they are in the structural load path. Unlike fixed wing structures, rotor blade control is centered almost exclusively on obtaining maximum twist. Host structure twist-extension coupling can be employed, using unidirectional composites in conjunction with isotropic actuators. A more efficient manner of twist control could be achieved using directional (or anisotropic) actuators, which could directly induce twist and permit greater freedom in the host structure design. For integral actuation, the actuator system must be capable of blade pitch changes of ±2° at the tip (Shaw and Albion, 1981). One such conceptual design employed directionally attached piezoceramics (Barrett, 1992), and demonstrated that torsional blade modes could be significantly damped on a practical blade structure (Nitzsche and Breithbach, 1994). A comparison of anisotropic actuator concepts applied to both integral and flap actuated blades is given in duPlessis and Hagood (1995). Neither concept has been demonstrated in a full size blade to date.

Another application of active materials to aerospace structures is in the area of Active Structural Acoustic Control (ASAC) of sound transmission through aircraft and helicopter fuselages. High acoustic levels are present within these enclosures (von Flotow and Mercadel, 1995), typically caused by propellers or engine fan spool speed imbalances transmitted via the wing structure, or periodic pressures exerted on the cabin from passing propeller blades (airborne noise). Unlike aeroelastic control applications, much of this work has been tested in full scale structures. Early control concentrated on noise cancellation technologies, employing microphone sensors, and speakers as acoustic actuators. These works typically demonstrated 10-15 dB reductions in sound pressure levels over the first several harmonics of the blade passage frequency. However, reductions were highly spatial, and required many sources for achieving control - up to 32 for small turboprop aircraft (Elliott, 1989). These numbers are required due to the spatial
mismatch between the primary acoustic field and the field produced by the interior sources (speakers), creating an interior acoustic field that is more broadband in nature (Silcox, 1990).

A more direct approach at acoustic control makes use of the fact that the acoustic field must pass through the structure. Vibration suppression of the fuselage structure has been studied using both point force devices (shakers) and in-plane bonded piezoceramics as actuators (Silcox et al., 1993; Fuller et al., 1992), illustrating reductions in interior noise levels similar to those obtained with speaker sources. More importantly, these reductions can be broadband in nature, are global throughout the structure, and can be achieved with far few actuator sources. Several full scale demonstrations have also shown that piezoceramics are feasible for this application (Fuller and Gibbs, 1994; Mathur and Tran, 1993). In fact, distributed active materials appear to be ideal for active structural acoustic control. Only small out-of-plane structural displacements (on the order of 25 μm) are required to obtain significant control authority. Studies indicate that ‘extended’ actuator patches are more effective in reducing controller spillover, which is the limiting factor in control performance (Lester and Silcox, 1992). In a more general sense, it is advantageous to tailor the size, location and number of actuators in order to optimize the energy coupling between acoustic and structural responses (Silcox et al., 1993). Both of these conclusions point to the need for directional and (semi)continuous actuator materials.

There are numerous other aerospace and related field applications that can or do utilize active materials to achieve improved performance: structural health monitoring, damage mitigation, de-icing, etc. to name a few. The above three examples discussed are applications currently receiving significant attention in the aerospace field, and these are used to drive the requirements for the next generation of active materials.

Work in the above applications has defined a set of desired actuator qualities for dynamic applications in aerospace structures. These requirements can be organized into three main categories:

- **Performance**: the need for high actuation authority, highly directional actuation, and moderate bandwidth (1 Hz to 10 kHz).

- **Robustness**: the need for actuator materials that are more robust to damage during use and handling (i.e. less brittle than piezoceramics), and have higher tensile strengths.

- **Manufacturability**: the need for highly integrated actuators that can cover entire structures, and actuators that are conformable to curved structures such as shell and tube sections.
Available Actuation Materials

There are a number of ways to compare potential solid state actuation devices. The standard manner in which to compare these materials are actuator-based measures of performance: actuation strain, induced stress, stiffness, bandwidth, or power consumption (i.e. efficiency). Energy-based comparisons are a convenient method of combining several of these performance metrics. The most useful is the Actuation Energy Density (J/kg), which is the maximum amount of work that can be delivered to an ideal load, normalized by the density of the actuation material. Several works have been dedicated to compiling a comprehensive comparison of actuators based on this metric (Giurgiutiu et al., 1995). However, the reader should be cautioned in making final conclusions based entirely on this metric, as the interpretation of maximum actuation strain varies with application. Furthermore, nonlinear effects often act to reduce the true available work that can be delivered to the structure, making this comparison misleading. Other considerations, such as parasitic mass of supporting components, require comparisons from a total system design point of view.

Many comparisons stop there. However, the proposed application places additional restrictions on the actuator material. In the case of aerospace structures, it is important to also consider factors such as weight, cost, and temperature range of operation (or environmental stability to be more complete). Also in this category is an item that is often overlooked when describing actuation materials: strength and reliability. There is a need for the actuation material to be both damage resistant and damage tolerant (Lagace, 1992). Planar actuation of structures typically involves high levels of stress, which are transmitted through the same transduction path that provides actuation authority over the structure. Thus, the material must be load bearing. Finally, realistic structural lifetimes will demand actuator materials with significant resistance to fatigue, both electrically and mechanically. For a typical rotorblade, the expected life is on the order of 2e8 rotation cycles, necessitating an actuator operating life of 6e8 cycles with 3/rev control.

Numerous solid state actuation materials have been investigated for aerospace applications. Perhaps the most widely used of these are piezoceramics, due to their relative ease of implementation and use. Actuation and sensing is applied through electrical signals, and their low field linear behavior has aided in modeling for transducer applications. Their high stiffness gives adequate energy densities, and their fast response times provide high bandwidth. In comparison (Table 1.1), other solid state actuation materials are less well suited for the desired applications. Piezo-polymer films (PVDF) are robust to damage, but lack the high stiffness to provide significant actuation authority.
Table 1.1 Comparison of Solid State Actuation Materials (Commercial and Potential)

<table>
<thead>
<tr>
<th></th>
<th>PZT 5H</th>
<th>PVDF</th>
<th>PMN</th>
<th>Terfenol D</th>
<th>Nitinol</th>
</tr>
</thead>
<tbody>
<tr>
<td>Actuation Mechanism</td>
<td>piezoceramic(31)</td>
<td>piezo film</td>
<td>electrostrictive</td>
<td>magnetostrictive</td>
<td>shape mem. alloy</td>
</tr>
<tr>
<td>Max Strain</td>
<td>0.13%</td>
<td>0.07%</td>
<td>0.1%</td>
<td>0.2%</td>
<td>2%-8%</td>
</tr>
<tr>
<td>Modulus (GPA)</td>
<td>60.6</td>
<td>2</td>
<td>64.5</td>
<td>29.7</td>
<td>28 m, 90 a</td>
</tr>
<tr>
<td>Density (kg/m$^3$)</td>
<td>7500</td>
<td>1780</td>
<td>7800</td>
<td>9250</td>
<td>7100</td>
</tr>
<tr>
<td>Actuation Energy Density (J/kg)</td>
<td>6.83</td>
<td>0.28</td>
<td>4.13</td>
<td>6.42</td>
<td>252-4032</td>
</tr>
<tr>
<td>Hysteresis</td>
<td>10%</td>
<td>&gt;10%</td>
<td>&lt;1%</td>
<td>2%</td>
<td>High</td>
</tr>
<tr>
<td>Temp Range</td>
<td>-20 to 200°C</td>
<td>low</td>
<td>0 to 40°C</td>
<td>high</td>
<td>—</td>
</tr>
<tr>
<td>Bandwidth</td>
<td>100 kHz</td>
<td>100 kHz</td>
<td>100 kHz</td>
<td>&lt;10 kHz</td>
<td>&lt;5 Hz</td>
</tr>
</tbody>
</table>

Electrostrictive materials (PMN) have low hysteresis losses and high stiffness, but have poor temperature stability, and require high currents to operate due to their high material dielectric. Shape memory alloys (Nitinol) are capable of very high strains, but are limited to ultra-low bandwidth applications (<5 Hz) due to the time needed for thermal dissipation/heating. Finally, magnetostrictive actuators (Terfenol) have similar actuation energy density and bandwidth as piezoceramics, but are much less effective when the coils and flux path materials are accounted for. Although piezoceramics at first seem ideally suited for planar structural actuation applications, they suffer from distinct disadvantages associated with reliability, performance, strength, and large scale distributed control.

In reality, no current solid state actuation material is capable of meeting all of the demands of large scale planar actuation of structures. A new material must be developed that combines the best attributes of available actuation materials. i.e. a material that has the structural nature of composites, yet has actuation performance that equals or surpasses current piezoceramics. This thesis is motivated by this challenge.

1.2 Objectives

The major goal of this work is to develop a new high strain composite actuator which comprises electroceramic fibers and passive matrix, and which meets the demands of active control applications in aerospace structures. Supporting tasks, which highlight the major contributions of this thesis, are the following:

- Develop methods for characterizing constituent materials for active composites
• Advance the field of composite modeling for coupled field materials
• Identify the key issues and tradeoffs in the design of active fiber composites
• Develop the ability to manufacture and characterize active fiber composites
• Demonstrate the capability of active composites in structural control of laminated structures

The ultimate objective is to enable highly distributed actuation and sensing for active structural control applications.

1.3 Background

1.3.1 Piezoelectric Fiber Composites

Conventionally electroded Piezoelectric Fiber Composites (PFCs) were introduced (Bent and Hagood, 1993) as a means for actuation that overcomes weaknesses in monolithic piezoceramics by providing increased strength, conformability, and reliability. This concept is illustrated in Figure 1.1. Unlike previous active composites which utilized ceramic rods oriented through the thickness to maximize the longitudinal coupling factors for ultrasonic transducer applications (Smith and Auld, 1990), the piezoceramic needed to be aligned in the plane of the structure to couple into stresses and strains associated with planar bending and twisting. The piezoceramic fibers were unidirectionally aligned to maximize the volume fraction (and hence the actuator authority), and to maximize the anisotropic nature of the actuation. Uniform electrodes, applied using vapor deposited metals above and below the compositected ceramic, delivered the electric field to the active ply. In this configuration, the poling direction is oriented through the thickness of the composite, and the planar actuation is due to the transverse (d_{31}) piezoelectric effect. As the illustration shows, both planar directions actuate in phase, although the induced stress is less in the direction perpendicular to the fibers due to the softer matrix in series with the fibers.

In particular, PFCs were shown to offer the following advantages over monolithic ceramic actuators:

1. **Performance**: Use of piezoceramic fibers maintained the majority of the stiffness and bandwidth of monolithic piezoceramics. The unidirectional fibers created anisotropic actuation. Thus, twist could be directly induced into isotropic host structures, for example.
2. **Strength and Conformability**: The combination of fine ceramic fibers and soft matrix provides load transfer mechanisms that increases robustness to damage, and allows static ultimates higher than monolithic ceramic.

3. **Large Scale Actuation**: PFCs could enable realistic applications which require highly distributed actuation and sensing. A single actuation ply can be fully integrated into the existing structure.

A number of models were developed to predict the linear composite properties from the bulk constitutive relations (Bent, 1994). Three variations of models were based on a *Uniform Fields* method, which is a generalization of the Mechanics of Materials approach to coupled field materials. This method provided a closed form solution, enabling an understanding of the major effects governing the composite response. An elasticity solution, based on a *Self Consistent* methodology, was also advanced for the prediction of properties. This method was not closed form, but was more accurate at low fiber volume fractions because it could capture the non-uniform concentration of electric and mechanical fields within the materials. Finally, a *Finite Element* model was employed to confirm the assumptions in the previous models. All models had good agreement, and provided a means for design and experimental validation of the composites.

A method for manufacturing piezoelectric fiber composites was also developed in the past work. Commercially made PZT fibers (130 μm diameter) had just become available, and were manufactured through extrusion techniques. These were used to manufacture single fiber thick plies for comparison to the above models. Manufacturing was done by hand, laying the fibers into a mold which was constructed on an aluminum cure plate. The matrix material was a two-part epoxy, which was (in some cases) doped with PZT filler particles to increase the matrix dielectric. These composites were cured in a manner similar to graphite/epoxy composites, using an autoclave to supply simultaneous heat and pressure to the specimen.
Later work demonstrated that these active composites were feasible for actuation applications. A twist-extension bimorph was constructed to demonstrate the possibility of induced twist on isotropic substrates (Bent, Hagood, and Rodgers, 1993). These PZT fiber composites were also embedded within laminated graphite/epoxy host structures, illustrating the feasibility of these materials as highly integrated active materials (Rodgers, 1995).

Piezoelectric fiber composites demonstrate substantial advantages over monolithic ceramics, and represent the future in active materials. However, one major drawback remained from the previous work in piezoelectric fiber composites - low actuation performance. The high dielectric mismatch between fiber and matrix (approximately three orders of magnitude in size) seriously reduced the electric field available to the fiber material for actuation. Hence, piezoelectric fiber composites had very low actuation energy density, comparable to PVDF. Furthermore, high field concentrations in the polymer-based matrix often caused dielectric breakdown prior to poling. Efforts to increase the matrix dielectric using high dielectric fillers were not sufficient to increase performance to levels comparable with bulk PZT.

1.3.2 Interdigital Electrode Monolithic Ceramics

The interdigital electrode (IDE) concept was introduced as a 'performance enhancement technology' for monolithic ceramic actuators (Hagood et al., 1993). Although interdigital electrodes have been used extensively in the past for surface acoustic wave (SAW) transducers (Feuillard et al., 1993), the idea was never applied to ceramics for the purpose of actuating structures until recently\(^1\). Figure 1.2 shows the concept of IDEs. Electrode fingers of alternating polarity are arranged on the top surface of a monolithic ceramic wafer, with an exact mirror image on the bottom. This arrangement of electrodes causes a portion of the electric field to be aligned within the plane of ceramic, as it travels between the electrode fingers. The percentage of field oriented in this direction depends upon the various geometry parameters: electrode finger width (w), wafer thickness (t), and electrode finger spacing(p). In general, for a large electrode spacing, and small electrode width and wafer thickness, the majority of the field is aligned in the plane, with little field inhomogeneity under the electrodes.

Such an electrode arrangement offers several advantages. First, the poling direction is now in the plane, allowing use of the much larger primary piezoelectric effect (d\(_{33}\)). Second, the actuation is anisotropic: the planar actuation is out of phase in the two

\(^1\) IDE concepts have been also suggested for stack actuation using multilayer ceramics (Yoshikawa and Shrouth, 1993).
Figure 1.2 Concept of the interdigital electrodes on monolithic ceramic (IDE).

directions, as illustrated in the figure. Such directionality is advantageous for tailoring actuation and sensing to particular modes of a structure. Models developed for prediction of the device response verified these benefits. An energy technique (Rayleigh-Ritz) using assumed modes for the displacement and voltage degrees of freedom was developed to provide a closed form solution of the effective constitutive properties. This model showed that an increase of almost two times in actuation strain was possible for geometry ratios p/h of 6 and w/h of 1. More importantly, an increase in induced stress capability (i.e. actuation authority) of three times was possible in the primary direction. The IDE device was also modeled using a commercially available finite element code. This model revealed the details of the internal electric fields, and validated the assumed modes used in the Ritz model in regimes of high electrode spacing, where field distortions near the electrodes had been ignored.

For the experimental work, the electrodes of a number of monolithic wafers were etched into the interdigital pattern using photolithography techniques. Measurements did show an increase in free strain similar to that predicted in the models. However, it was also found that many of the ceramics failed due to catastrophic cracking during poling. Furthermore, unpublished results later found that it was difficult to obtain significant induced stress in samples laminated to passive materials. The difficulty originates in electric field gradients near the electrodes, causing stress concentrations in the so-called ‘dead areas’ of the actuator. A second difficulty may be due to local areas of high electric field near the electrode edges and terminations where charge can collect.

A similar problem occurs in multilayer ceramic (stack) actuators, where microcracks are preferentially initiated near the electrodes, leading to crack propagation under stress and eventual actuator failure (Pan and Yoshikawa, 1996). Researchers have visually detected these cracks using microscope CCDs in a number of active materials, including electrostrictors, switching ceramics, and piezoceramics (Aburatani et al., 1994). Several alternative electrode schemes have been investigated to reduce the stress concentrations in
multilayer actuators, including floating electrodes, slits in the ceramic, and insulator materials (Yoshikawa and Shront, 1993). However, these methods increase cost and do not directly solve the problem. An ideal solution is to remove the electrode material from the ceramic, thus desensitizing the actuator performance from damage caused at the electrodes by field concentrations in these areas. Electrodes on separate materials, such as an interlaminar electrode in the PFC design would accomplish this.

1.4- Approach

1.4.1 Advanced Smart Composite Materials Concept

The future vision for smart materials in structures was developed at MIT, in the Active Materials and Structures Laboratory (Hagood, 1994). This vision was driven by the need to provide a means for non-intrusive integration of active materials into current and future aerospace applications. Ideally, the active materials are seamlessly laminated within a host composite structure (Figure 1.3), covering the entire area of a critical component: the spar of a rotorblade or wing, or the fuselage of an aircraft. This is only possible if the active material can be designed with the strength, conformability and manufacturability similar to other advanced aerospace materials, such as graphite/epoxy or glass/epoxy. The active material will then be strong enough to permit the replacement of passive plies with active ones, creating dual purpose materials: plies that provide passive stiffness and strength, and are able to actively control the structure. Such plies will also have high levels of actuation authority due to complete coverage. This is unlike past embodiments of active materials, which are capable of only the second function. Active materials such as monolithic piezoceramics can not contribute to the strength of the load bearing structure, adding dead weight to the structure. Furthermore, only non-continuous coverage is possible due to the maximum available size of tape-cast ceramics, causing difficulties in stress concentrations and lead wire attachment within a laminated host.

The advanced composite is moving toward the defined concept of intelligent structures, with distributed actuators, sensors, processing, and control. Each layer is multifunctional in design. The actuation lamina (ply) is composed of the active material and the interlaminar electrode. The active material is in a composite form to take advantage of the benefits of composite damage tolerance through alternate load paths. Fine ceramic fibers provide the majority of strength and stiffness of the composite, while the matrix material surrounds and protects the fine fibers and provides load sharing and stress distribution. The active fibers may also be surrounded by reinforcing materials to increase the strength of the actuation ply. The interlaminar electrodes deliver the electric field to the
Figure 1.3 An illustration of the concept of an advanced smart composite material. active material and insulate the active material from adjacent passive or active plies. The sensor/processing layer incorporates sensors for structural output monitoring, logic circuitry for real-time control decisions, and amplifiers to drive the active lamina layer. Both plies are designed for distributed (hierarchic) control and robust operation. This will eventually be made possible through redundant components and an adaptive control architecture.

This thesis focuses on developing the actuation layer, both through developing manufacturing methods, and by advancing models for prediction of composite properties. Work in the area of hierarchic control (How and Hall, 1991), embedded electronics (Warkenti, et al., 1992), and shaped sensors (Miller et al., 1990) is being carried out in other programs, and will contribute to the overall goal of developing true intelligent structures.

1.4.2 IDEPFC Actuators Concept

The concept of the interdigitated electrode piezoelectric fiber composite (IDEPFC) is a fusion of the concepts presented in the earlier section: interdigitated electrode monolithic ceramics (IDEs) and piezoelectric fiber composites (PFCs). The premise is shown in Figures 1.4 and 1.5. Figure 1.4 illustrates the geometry of the IDEPFC device. Fibers are again aligned transversely to couple to in-plane forces, with fiber orientation taken to be
along the 3-axis. The electrode patterns have fingers of alternating polarity, and are exact mirror images on the top and bottom faces. Poling is predominantly along the 3-axis direction. Application of an electric field as shown produces primary actuation along the fibers (shown as an extension in the figure), and an out of phase transverse actuation (a contraction) perpendicular to the fibers.

A quarter model section of the IDEPFC actuator is shown in Figure 1.5. This cross-section is taken along planes of symmetry in all three axes, and will be used as the representative volume element for modeling. Geometry for this model is adopted from the previous work on IDEs, where $h$, $w$, and $p$ still represent the wafer thickness, electrode width, and electrode spacing, respectively. Previous work with the monolithic wafers incorporating IDE patterns showed a relatively uniform equipotential distribution away from the electrode, especially for larger electrode spacing. A similar result is expected here, although a percentage of the electric potential will now be concentrated in the low dielectric matrix. This distribution is reflected in the estimated pattern shown in the figure, illustrating both equipotential and electric field lines.

The previous work determined that maximum actuation capabilities were achieved for geometries where the resulting electric field was as homogeneous as possible. Thus, for high values of $p/h$ and $p/w$, the electric field was most nearly aligned completely in the plane of the ceramic, and very little of the field was involved in the local behavior near the electrode. For a value of $w/h=1$ (i.e. equal electrode and wafer thickness), the piezoelectric free strain and clamped stress along the 3 direction were shown to asymptote to the bulk ceramic longitudinal values $(d_{33}, e_{33})$, as $p/h$ went to infinity\(^2\). These values were within 90% of the asymptote at a $p/h$ as low as 6. This previous research chose these values for the geometry of the experimental section of that work in order to get the maximum benefits

\[^2\] Normalized by electric field, rather than voltage
of the IDE and maintain the ability to pole and actuate at reasonable voltages. The same values will be used in this present work.

There are several distinct advantages in the combining of the two previous concepts. As in the IDE device, the interdigitated electrode pattern should greatly enhance the performance of active fiber composites. This arrangement would increase the effective ceramic volume fraction along the electric field pathlength, making the higher strain levels possible at lower voltages. In addition, the primary axis of piezoelectricity now lies along the fiber direction, greatly increasing the maximum strain. Another advantage, illustrated by previous work in anisotropic actuation (Bent et al., 1993), is the increased level of anisotropy possible for the composite actuators. Although the actuation anisotropy is not as high as in the IDE device due to soft matrix in the 1-axis direction, the amount of ceramic can be tailored to maximize this effect. In addition to overcoming the weaknesses in monolithic ceramics, the composite form also removes the previously discussed drawbacks to the IDE technology. The interdigitated electrode pattern which was previously etched directly onto the ceramic is now applied to a separate interlaminar layer. This enables the composite to remain functional in the presence of damage to the ceramic.

1.5 Scope of Thesis

The organization of this PhD work (and subsequently this thesis) is based on the philosophy of vertical technology integration, applied to the problem of active composites design and implementation. Figure 1.6 offers a pictorial view of the thesis, breaking down the items by chapter. The flow-up represents the increase in design/manufacture capability, which supports the application of the technology to the next level. As in any design process, the effort is often circular in nature, whereby re-designs occur as a result of the
Figure 1.6 Outline of thesis, illustrating natural progression from constituent materials to structural applications. Numbers correspond to chapters in thesis.

outcome of testing or implementation. The flow-down of requirements supports this redesign.

Chapter 2 details the characterization of the constituent materials used in the active fiber composites. The purpose of the characterization is twofold: to provide model inputs of the material properties, and to allow design of optimal actuators based on a combination of processing and performance. The characterization is divided into the two ‘bulk’ materials, fibers and matrix. Fiber material choice is done through examining the response of the bulk ceramic of different PZT types. Chosen bulk materials are characterized in a series of poling, aging, and actuation free strain studies. Actual PZT fibers are also characterized, in order to determine the relationship of bulk-to-fiber properties. The matrix material, although passive, plays an important role in the electrical, mechanical, and actuation properties of the composite. Studies are undertaken to understand the tradeoffs existing between processing ease and composite performance by adding fillers to the pure epoxy starting material. Rheological, electrical, and dielectric breakdown tests are performed on the matrix in order to quantify these effects.

Chapter 3 assumes knowledge of the material constitutive properties and applies linear modeling techniques to determine the effective (or averaged) composite properties. Two electro-mechanically coupled models are developed to predict the composite response: a Uniform Fields approach, and Finite Elements. The Uniform Fields method is an generalization of the well known Mechanics of Materials approach to coupled field materials, allowing a closed form solution which provides physical insight to the IDEPFC problem. A commercially available Finite Element program, containing coupled field
elements is also used to model the IDEPFC structure, and provide validation of the simplifying assumptions made in the previous model. The objective of this work is to develop methods for modeling coupled field, multiphase materials which can be generalized to any composite geometry and any combination of (linear) constituents.

Chapter 4 applies the models developed in the previous chapter, and the bulk material characterization of Chapter 2, to the design of active fiber composites. This is done by examining the change in composite properties as a function of the different fiber materials and matrix compositions. Metrics for composite performance and reliability are introduced to quantify the tradeoffs in materials. From the design studies, several designs are suggested for experimental work.

Chapter 5 details the manufacture of individual IDEPFC specimens used in the model validation and performance tests. Preparation of the composite molds, lay-up of fibers, introduction of matrix, and curing is described for the different series of specimens. Post-cure preparation of the specimens for testing involves removal from the mold, poling, and bonding to a substrate material for the clamped stress testing. Poling methods for bulk ceramic, drawn from Chapter 2, serve as guidelines for composite poling. Finally, issues of active composite lamination and integration into passive host materials, dealt with during the manufacture of a model blade and bender test articles, serve as guidelines for future adaptive structures utilizing IDEPFCs.

Chapter 6 describes the experimental determination of composite properties. The objectives of this Chapter are to validate the model predictions of Chapter 4, and investigate the performance of laminated structures that incorporate active composites. Methods for measuring the various electrical and piezoelectric properties are described, including direct measure (dielectric and free strain) and indirect measure (clamped stress) techniques. Low field properties are compared to the Uniform Fields model of Chapter 3, as a function of ceramic fraction to demonstrate the effect of increasing authority. Results are also compared for a fixed fiber fraction, but using different fiber materials and different matrix compositions. The performance of the composites at high fields is examined to evaluate the model's capability to extend to nonlinear regimes of operation and to compare the capability of the different materials. Finally, the results for the two laminated structures for rotorblade applications that apply IDEPFC technology will be described.

Chapter 7 closes the thesis with principal conclusions from each chapter, and recommendations for future work in active fiber composites.
Chapter 2

Characterization of Constituent Materials

2.1 Introduction

One of the key challenges facing this work is the need to increase the performance of piezoelectric fiber composites. One way to improve performance is approached at the structural level, through the use of interdigitated electrodes. A second way is to approach the problem at the material level, by auspicious selection of the bulk properties. The issues affecting material selection in piezoelectric fiber composites were originally identified in the first work on this topic (Bent, 1994). The single most important factor affecting performance is the large dielectric mismatch between the fiber and matrix materials. Methods for reducing this mismatch include considering both the fiber and matrix materials. However, the choice of these materials directly affects other issues, such as the ability to process the composites. At the same time, material selection gives the added freedom of optimizing for specific applications. For example, it may be possible to tailor the matrix and fiber electrical properties to provide passive damping, maximized for a particular frequency range.

This chapter deals with fundamental issues of the constituent (or bulk) materials, and understanding how the choice of these materials can greatly affect the overall composite behavior and ease of manufacturing. The objectives for this chapter are

- to identify what information is important for the design of active composite materials, and
- to develop methods for characterizing bulk material properties, and use these methods to obtain the information needed.

The property characterization in this chapter is geared toward issues defining composite performance for structural actuation. However, the methods for characterization and the design tradeoffs can also be applied to other composites where active and passive materials are combined to optimize the overall properties (such as ultrasonic transducers, etc.).
This chapter is divided into two major sections: characterization of active (PZT) materials, and characterization of passive matrix materials. Different PZT ceramic materials will be characterized for performance, specific to needs for structural actuation. A single PZT type is then selected and characterized in bulk form for the application to composites, including a study in poling and aging. Actual PZT fibers are also characterized, for a comparison of properties to bulk ceramic, and to determine product variability and what effect it may have on measured composite properties. The matrix material is characterized for its affect on both composite performance and composite processing. Conductive and dielectric fillers are added to the matrix to tailor the dielectric properties. How this affects viscosity (processing), dielectric constant (performance), DC resistivity (poling), and dielectric strength (stability/safety) are studied. Although matrix and fibers are dealt with separately, it is impossible to consider the final selection independently. The next chapter (3.0) will provide models for the combination of the properties, and the subsequent chapter will show the tradeoffs in material selection on composite properties.

2.2 Piezoceramic Characterization

2.2.1 Ceramic Material Selection

Introduction

Electroceramics (in the current context) can be loosely defined as those ceramics which display a mechanical-electrical conversion of energy. Several of these were discussed in Chapter 1.0, and include ceramics whose energy conversion is based on the piezoelectric effect, the electrostrictive effect, or the ferroelectric-antiferroelectric switching effects. As previously discussed, piezoelectric materials were baselined for a number of reasons. However, even within the piezoelectric ceramic families there are a number of types, which range from ‘soft’ to ‘hard’ piezoceramics. These differences are caused by slight variations in oxide compositional ratios, or additional dopant materials. These compositions affect the mobility of the domains to spontaneously polarize with a given electric field. This, in turn, affects the degree of ‘hardness’ of the ceramic. A soft piezoceramic has higher domain mobility, and thus is easier to pole, often has a lower coercive field, and has higher electrical-mechanical coupling factors (a measure of energy conversion efficiency). By the same token, however, easier domain mobility also results in easier ceramic depolarization from stress or electric field, higher hysteresis and nonlinearity, and higher rates of piezoelectric ‘creep’. Transducer applications that are subject to mechanical shock and high stresses are often designed with hard piezoceramics.
to avoid depolarization in service, and hysteretic heating. Structural actuation has typically utilized the softer ceramics for high levels of actuation. Heating is generally not an issue for low frequency dynamic problems such as structural vibration suppression.

Piezoceramics are identified by Navy designations (Navy types I, II, IV, etc.), or more commonly by the Morgan-Matroc trademark names, which will be used here. Four of these are shown in Table 2.1 (PZT 5H, 5A, 4S, 8M), which represent a range of piezoceramics from very soft (5H) to very hard (8M). Applications design (including structural actuation) has usually been done using the low field linear properties. These properties are conveniently described in the linear constitutive relationships, which demonstrate the electrical and mechanical coupling through the piezoelectric properties. One form often used for structural control applications is (IEEE, 1986),

$$\begin{bmatrix} D \\ S \end{bmatrix} = \begin{bmatrix} \varepsilon^T & d \\ d & s^e \end{bmatrix} \begin{bmatrix} E \\ T \end{bmatrix}$$

(2.1)

where $D$ is the electrical displacement (Coulombs/m²), $S$ the strain, $E$ the electric field (kV/cm), and $T$ the stress (MPa). These complementary electrical and mechanical fields are related through the material properties: the constant stress (unclamped) dielectric ($\varepsilon^T$), the induced strain constant ($d$), and the constant field (shorted leads) compliance ($s^e$). Subscript ‘t’ denotes matrix transpose. Another common form is expressed in terms of the stress as the dependent variable, which is convenient for describing the induced piezoelectric stress possible,

$$\begin{bmatrix} D \\ T \end{bmatrix} = \begin{bmatrix} \varepsilon^s & e \\ -e & c^e \end{bmatrix} \begin{bmatrix} E \\ S \end{bmatrix}$$

(2.2)

where $\varepsilon^s$ is the clamped dielectric, $c^e$ is the constant field stiffness, and $e$ is the induced stress. A more complete description of the constitutive equations and relationships between the various constants are contained in Appendix A.

These properties will be important in the design of composites, as they dictate the actuation capability when combined with the passive matrix properties (using the models of Chapter 3.0). However, it is unclear from the linear material properties alone which piezoceramic is best suited for the composite application. For true structural actuation applications, it is also important to know how the material performs in the high field nonlinear regime. Quantities such as the coercive field (field to polarize and depolarize the ceramic), remnant polarization, and maximum induced strain levels are of great interest, as they describe the maximum performance limitations of the piezoceramics. These quantities
are less well defined and need to be determined experimentally, and in the framework of the current problem.

**Experimental Method**

A materials testing device was designed for the characterization of the different piezoelectric types. The device needed to be capable of testing at voltages significantly above the breakdown of air (~24 kV/cm), and so was designed to use oil as an insulating medium surrounding the sample being tested. This had the added benefit of permitting temperature control when connected to an oil bath. Since the interdigitated electrode will align the field predominantly along the fiber, it was important to characterize the strains of the PZT samples through the thickness - a difficult task using strain gages. Instead, laser interferometry techniques were applied for measurement of the ‘33’ strains.

Figure 2.1 shows the design of the characterization device. A Lexan enclosure was built with oil ports to allow circulation of oil (only room temperature tests were done on the PZT), and a sealed top to contain the oil. The specimen was held between two conductive points; one that was moveable on a micrometer stage, but was locked in place during testing, and a second that transmitted the sample displacement outside of the enclosure. This was done using a rigid aluminum rod, which was sectioned as shown in the figure. A silicone diaphragm provided the seal for the oil, while dual flexures mounted on the inner and outer sides of the housing ensured linear motion of the rod. Displacement was measured using laser interferometric equipment, which was used with a setting resolution.

![Figure 2.1](image)

*Figure 2.1* Interferometric measuring device designed for high field controlled temperature testing of piezoceramics. *(a)* Isometric view of device, *(b)* Cross section illustrating mode of operation.
of 1.28 μm/5 volts (maximum resolution is 19 nm/5 volts).

Four types of piezoceramics provided by Morgan Matroc were tested (PZT 5H, 5A, 4S, 8M), in order to represent the range of commercially available ceramics. These ceramics were provided in the form of disks, approximately 12 mm in diameter and 1.75 mm in thickness. All disks were nickel-plated, and were provided in both poled and unpoled states. Low field constants were taken directly from the specification sheets provided with the samples. Experimental testing was limited to high field measurements. The first tests were conducted to determine the coercive field (Ec) of the individual ceramics, in order to set voltage limits in operation of the composite actuators. A sinusoidal voltage (1 Hz) was applied at levels high enough to cause complete repolarization of the ceramic, giving the well-known butterfly curve. Values for the maximum, or saturation, strain were recorded for each type of ceramic from this test. For the soft ceramics (5H, 5A), this level was taken to be at a voltage twice the coercive field, while for the hard ceramics (4S, 8M) the maximum voltage was capped at a practical limit - twice the breakdown strength of air (~ 48 kV/cm). In all cases at these voltage levels, complete repolarization of the ceramic dipoles was occurring. Current measurements were provided by the output monitor of the high voltage amplifier, which could be used for determining the remnant polarization.

Although the saturation strain is a measure of the maximum possible induced actuation strain, actuators are not used in voltage cycles that cause the material to undergo repolarization. This can cause material fatigue in as few as 1e3 to 1e6 cycles (Jiang et al., 1994), and is difficult to control in a closed loop manner due to the reversal in gain sign with repolarization. Instead, ceramics that had been poled were used for further testing (provided poled by the manufacturer). After poling, most piezoceramics are used in the low field regime, with applied voltages far away from the coercive field to prevent repolarization. To maximize possible performance for structural actuation, a Representative Work Cycle was determined for each material tested. This cycle used a bias voltage\(^1\) to permit higher driving voltages in the direction of poling. For each ceramic type, the strain was measured for a voltage cycle of -75% Ec against the direction of poling, to the value where saturation strain was reached. This was chosen as +200% Ec in the soft ceramics, and 48 kV/cm (twice the breakdown in air) for the hard ceramics. The corresponding strain is reported as the Representative Work Cycle Strain in Table 2.1. These voltage limits have been found to maximize the available actuation strain without causing significant fatigue

---

\(^1\) A dc bias can be used for low field drive of piezoceramics in order to increase the sensitivity due to increased dipole alignment (Jaffe et al., 1971).
problems or introducing a danger of repolarization in IDEPFC samples (Rodgers et al., 1996).

**Results and Discussion**

A summary of low field material properties and high field properties is given in Table 2.1. Comparisons are made considering a one dimensional mode of operation (along the 33 direction). The low field properties are typically measured using well established resonance techniques (IRE, 1976), taken using very low voltages. The piezoelectric constants can also be determined using quasistatic methods, which give values that are within 1.5% of the values measured using resonance techniques (Bent, 1995). PZT 5H has very high induced strain constants (strain per unit voltage) due to the domain mobility of the ceramic. More importantly, PZT 5H has the highest induced stress constant (blocked stress per unit electric field), indicating high actuation authority. Since the induced stress is a combination of both induced strain and ceramic stiffness, the hard ceramics (which happen to be stiffer) have comparable performance using this quantity. This is even more evident in the coupling coefficient, which is a measure of the fraction of electrical energy converted to mechanical energy and vice versa, which also takes into account the dielectric constant. For PZT based electroceramics, this value is quite high (as opposed to 0.1 for Quartz, and 0.4 for Barium Titanate), and similar among the different PZT compositions.

The effect of ceramic composition is also apparent in the high field constants, which are the shaded quantities in Table 2.1. The quantities come from the results shown in Figures 2.2 and 2.3. In Figure 2.2, the left column contains plots of the strain versus electric field, also known as the butterfly curves. The softer ceramics have sharp, well defined coercive fields where repolarization occurs, whereas the hard ceramics (4S, 8M) appear to have a greater distribution of domain coercive fields, spreading the net ceramic repolarization over a range of voltages. The soft ceramics also have high saturation strains, as expected, taken as the total strain in the butterfly curve. The right column of Figure 2.2 shows the electrical displacement versus electric field, which is the well known hysteresis curve. It describes the nonlinear polarization switching behavior as a function of applied field. The hysteresis comes from the energy needed to reverse the metastable dipoles during each cycle (Jaffe et al., 1971). These curves demonstrate the maximum remnant

---

2 This measures the *converse* piezoelectric effect (strain for applied field), and is described in Chapter 6 in measurement techniques. Quasistatic methods are also reported for measuring the *direct* piezoelectric effect (charge for applied stress) in Bertincourt (1964).
Table 2.1 Comparison of longitudinal properties for four PZT ceramics. Low field data provided by Morgan Matroc, Bedford, Ohio. Shaded area shows high field data measured here.

<table>
<thead>
<tr>
<th></th>
<th>PZT 5H</th>
<th>PZT 5A</th>
<th>PZT 4S</th>
<th>PZT 8M</th>
</tr>
</thead>
<tbody>
<tr>
<td>Dielectric $k_{33}^T$</td>
<td>3350</td>
<td>1770</td>
<td>1500</td>
<td>800</td>
</tr>
<tr>
<td>Induced Strain $d_{33}$ (mV)</td>
<td>555</td>
<td>392</td>
<td>360</td>
<td>215</td>
</tr>
<tr>
<td>Induced Stress $e_{33}$ (N/C)</td>
<td>23.2</td>
<td>15.8</td>
<td>14.6</td>
<td>17.3</td>
</tr>
<tr>
<td>Coupling Coefficient $k_{33}$</td>
<td>0.790</td>
<td>0.774</td>
<td>0.861</td>
<td>0.767</td>
</tr>
<tr>
<td>Young's Modulus$^3$ (GPa)</td>
<td>48</td>
<td>53</td>
<td>60</td>
<td>77</td>
</tr>
</tbody>
</table>

Depole Stress $T_{33}^{\text{max}}$ (MPa) | 44$^4$ | 72 | >150 | >150

polarization for each bulk ceramic (see the next section for more on polarization). This will be important when comparing composites that use different PZT fiber types.

The Representative Work Cycle Strains are shown in Figure 2.3. These curves can be considered a portion of the butterfly curve, but are somewhat lower in value due to the more limited voltage drive range used to avoid repolarization. Note the high level of hysteresis and nonlinearity accompanying the higher strains of the soft ceramics. The actuation energy density here is the same as defined in Chapter 1, but based on the Representative Cycle values measured for each ceramic. Since the energy density is proportional to the square of the strain, and only linear with stiffness, PZT 5A has substantially higher Rep Cycle actuation energy than others.

$^3$ This is the Young's Modulus in the poling or '3' axis ($1/t_{33}^E$), and not the commonly reported Y33, which is a 3D stiffness constant.

$^4$ Depolarization Stress taken as reduction of $d33$ to 75% of its stress-free value, as provided in Zhang et al. (1994).
Figure 2.2 Nonlinear strain versus electric field (butterfly curves) and electrical displacement versus electric field (hysteresis curves), illustrating full polarization reversal.
Figure 2.3 Maximum Representative Work Cycle Strains for four PZT types.

A final property of the ceramic comparison is the compressive stress depolarization level. A compressive stress aligned with the direction of poling (hence, along the fibers in the composite) will tend to reorient the dipoles 90° to the original poling axis, causing a loss of piezoelectric properties. The highest depolarization stresses are found in the hard ceramics, making them suitable for high stress environments. At this point, PZT 5H was chosen as the ceramic for use in the initial composite design and manufacture. This choice was made based on the high actuation strains (and stresses), ease of polarization, and the commercial availability of fine PZT 5H fibers. The other ceramic compositions were not available in fibrous form at the time. The next section continues with the characterization of PZT 5H.

2.2.2 Ceramic Polarization Study

Introduction

Polarizability is a property inherent in all dielectrics (Buchanan, 1986). One type is *electronic polarization*, caused by shifts in electron clouds of the material away from their equilibrium positions for an applied field. Polar materials have additional contributions to
the material polarizability from orientation polarization of the dipoles for an applied electric field. In all cases, application of the electric field to the dielectric material induces a volume polarization. The field created in the material is offset by additional charges that collect on the electrodes (for a parallel plate capacitor analogy). Thus, the dielectric constant is used as a measure of the charge storage capability of the material.

One way to describe the polarization state of the material is the volume charge density. Alternatively, it can be described in terms of the surface charge on the material which exactly cancels the net volume charge. The polarization (\( \mathbf{P} \)) of a material is related to the electrical displacement (\( \mathbf{D} \)) and applied electric field (\( \mathbf{E} \)) through the following:

\[
\mathbf{D} = \varepsilon_0 \mathbf{E} + \mathbf{P}
\]  

(2.3)

where \( \varepsilon_0 \) is the dielectric of vacuum. Note that these parameters are vector quantities. Since the electrical displacement and electric field are related through the material dielectric constant (\( \varepsilon \)) in linear dielectrics by

\[
\mathbf{D} = \varepsilon \mathbf{E},
\]

(2.4)

it is possible to write the polarization in terms of the applied field as

\[
\mathbf{P} = (\varepsilon - 1) \mathbf{E}.
\]

(2.5)

Generally, \( \varepsilon \) is much larger than one in most dielectrics, so that the polarization can be approximated by the electrical displacement, as written in Equation 2.4. In most cases, it is easier to measure the electrical displacement directly, and the terms are often used interchangeably.

Note that the polarization vanishes with zero electric field. That is, the removal of the field causes the material to return to its original unpolarized state, and the internal field no longer needs to be canceled by additional charges on the electrodes. In contrast to pure dielectrics, is the phenomenon of piezoelectricity, described as electric polarization produced by a mechanical stress or strain. In these cases, the polarization of piezoelectric materials is non vanishing even after the field is removed.

The piezoelectric phenomena was originally studied in single crystals of Rochelle salt. The lack of center symmetry of these single crystals causes a net movement of the positive and negative ions with respect to each other upon application of a stress. This results in an electric dipole, and charges appear on the surface of the crystal. This external surface charge (polarization) is a quantity with the units of charge per area. Many crystals make up a polycrystalline ceramic, whose individual dipoles are initially randomly aligned, showing no net polarization. That is, unpoled polycrystalline piezoceramics show no piezoelectric effects because the individual dipole moments within the ceramic cancel.
Figure 2.4 Hysteresis plot, illustrating saturation polarization ($P_{sat}$) and remnant polarization ($P_r$).

Ferroelectricity is defined as the existence of 'reversibility in a polar crystal of the direction of the electric dipole by means of an applied electric field' (Jaffe et al., 1971). With the application of a high electric field, these dipoles align preferentially, giving rise to the piezoelectric effect and a net polarization. Thus, polycrystalline piezoceramics owe their usefulness to the existence of ferroelectricity.

The magnitude of the piezoelectric coupling coefficients have been shown to depend on the degree of polarization (Berlincourt, 1964). In other words, full polarization is necessary to realize the maximum benefits of the material. Evidence of polarization in a piezoelectric is usually described by the hysteresis plot of the $D$ versus $E$ curve, shown in Figure 2.4. For a non-ferroelectric, this curve would be a straight line (ideal capacitor), with its slope equal to the dielectric of the material. For a ferroelectric material, the flow of charges from dipole alignment causes a hysteretic path. The value for electrical displacement depends on the time history of the ceramic. The curve is shown for one complete cycle of dipole reorientation. The maximum level of instantaneous polarization is the saturation polarization ($P_{sat}$), and the polarization remaining after the electric field has been removed is the remnant polarization ($P_r$). It is the remnant polarization that indicates the degree of poling in the ceramic. For a non-ferroelectric material, this value would always be zero. Finally, the point at which the electrical displacement vanishes is the coercive field ($E_c$), corresponding to the coercive field found in the butterfly curve.

The degree of remnant polarization has been shown to be dependent on the magnitude of the electric field, the temperature of the ceramic during poling, and the length of time the poling field is applied. The best technique is to apply the poling field while the ceramic cools through its Curie temperature\(^5\) (Jaffe et al., 1971), where the dipoles align as

---

\(^5\) Curie temperature is defined as the temperature where the material becomes non-polar, which is $>200^\circ C$ for most PZT ceramics.
they appear spontaneously. This is difficult to do for all but the softest ceramics, such as Barium Titanate which has a Curie temperature around 130°C. Other ceramics (even PZT 5H with a Curie temperature of 193°C) would require temperatures damaging to the composite structure. However, some level of increased temperature may improve the degree of polarization. The need for higher temperatures may also be offset by applying the field longer or at higher levels (at some percentage of the coercive field).

Although the level of remnant polarization is easy to measure for a sinusoidally varying electric field, it is difficult to accurately measure the charges associated with dipole switching in a long duration poling process (on the order of minutes)\(^6\), due to leakage currents and other difficulties. Since the remnant polarization is likely a function of time, it is important to be able to assess other than the AC value. Instead, this experimental study will use the change in dielectric constant of the piezoceramic as an indicator of the degree of remnant polarization. This is a natural choice, considering the dielectric is the low field polarization, contributed to by each polarization phenomenon, including piezoelectricity. Higher values of the dielectric constant will indicate a higher degree of remnant polarization. This section determines the degree of polarization of PZT 5H as a function of the three above mentioned variables, by monitoring the increase in dielectric constant.

In the previous section, several piezoceramics were compared on the basis of their material constants, particularly for the magnitude of the piezoelectric effect in both low field and high field regimes. These quantities were based on the assumption that the materials were already fully poled. Polarizing of ceramics is not a well documented phenomenon, and the conditions used to accomplish poling are often proprietary to the manufacturer. Since the piezoceramic fibers will not be poled prior to composite manufacture, it is necessary to pole the ceramic in-situ. Once surrounded by matrix, it is difficult to establish whether the measured composite piezoelectric constants represent fully poled ceramic. However, the coercive field is a quantity which is easily measured for both the bulk ceramic and composite, and can be used to represent the level of electric field in the ceramic, regardless of configuration. This section establishes the necessary poling conditions in bulk ceramic using the coercive field as a metric. Then, the necessary poling conditions are easily identified for the composite form, once the composite coercive field is measured. It is important to accurately determine the poling field, not only to achieve full poling, but to avoid using fields that are unnecessarily high that may damage the composite.

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\(^6\) One method applied to bulk ceramics measures the charge released from depolarization caused by applying a compressive stress, to determine the effectiveness of the poling conditions.
Directly related to the phenomenon of poling is ceramic aging, or the post-poling change in properties with time. Aging has been observed for a variety of ceramics, with different aging rates dependent on the hardness of the ceramic. Changes in material constants include a decrease in dielectric constant, dissipation factor, piezoelectric constants, and the coupling factor, and an increase in the stiffness and mechanical quality factor. This aging is likely caused by the relief of various internal stresses that arise from the spontaneous polarization (Jaffe et al., 1971). Following poling, domains are slowly re-oriented into positions of lower energy. For Barium Titanate, the trend of aging has been shown to be linear when plotted against the log of time:

\[
\begin{align*}
\varepsilon_{33}^T & \quad 4\% \text{ per time decade} \\
 k_{33} & \quad 2.5\% \text{ per time decade}
\end{align*}
\]

A similar trend might be expected for PZT 5H, although the per-decade changes will be different. Such a change in material properties implies that a potentially large discrepancy in measurements are possible if taken at different periods in the aging process. Thus, the aging process of PZT 5H is characterized here by following the material constants as a function of time after poling. Again, the capacitance will be used as an indication of the remnant polarization, and hence, the trends in all piezoelectric constants. Piezoelectric constants are not measured, as the application of electric field to the sample at levels high enough to get significant strains may affect the aging rate (Jaffe et al., 1971).

**Experimental Method**

**Poling Study**

Piezoceramic wafers from a single lot were cut into thirty-six smaller pieces for the poling study, approximately 12.7 mm (1/2") wide in both directions, and 0.3 mm (12 mil) thick. The samples were thermally depoled in an oven at 225 °C for 30 minutes. One sample was tested to ensure complete depolarization by confirming the lack of piezoelectricity at fields below the coercive field. The samples were allowed to thermally stabilize for 24 hours in silicone oil, held at room temperature (23°C). The need for stability was important, as capacitance is extremely sensitive to changes in temperature. The pre-pole capacitance was measured for each sample in the same test fixture used for the dielectric breakdown tests (Figure 2.11), in oil at room temperature. The capacitance values were taken at 1 kHz, using an HP Impedance Analyzer\(^7\). Each sample was then poled at a different condition:

---

\(7\) HP 4194a Analyzer
- **Temperature** Samples were poled at three different temperatures in a heated oil bath: at 23°C, to examine the possibility of room temperature poling, and at 60°C and 90°C to examine the higher temperatures often used by manufacturers.

- **Time** Samples were poled for three different logarithmic times: at 10 seconds to evaluate ‘flash’ poling often used for hard ceramics, and at 100 and 1000 seconds (16.7 min) to evaluate intermediate and long duration poling.

- **Field Level** Samples were poled at four fields that were fractions of the room temperature coercive field8 (found for PZT 5H in the previous section): 50%, 100%, 150%, and 200% Ec.

After poling, the samples were aged for 24 hours. The post-pole capacitance was again measured in oil, at 23°C, recorded within ±2 min of 24 hours after the poling to avoid discrepancies caused by different aging times.

**Aging Study**

Two piezoceramic disks, approximately 12.7 mm (1/2”) in diameter and 1.75 mm (70 mil) thick were used in the aging study. Both disks were thermally depoled using the same conditions as above, and again allowed to thermally stabilize for 24 hours. The poling needed to be carried out at room temperature so that the change in capacitance could be measured shortly after poling. Full poling was attempted by using a poling field 300% of the room temperature Ec for 20 minutes, applied at room temperature (not shown in previous study). The post-pole capacitance was measured immediately after poling (2 min), and then logarithmically with time (10, 100, 1000, 10000 min).

**Results and Discussion**

The results of the poling study are shown in Figure 2.5, as surface plots for the change in capacitance between the pre-pole and post-pole values. Note that the values on the surface have been linearly interpolated between the measurement points of field and time. The maximum recorded change in capacitance was approximately 62%, reached at several combinations of time, temperature, and field level. Time of poling seems to have the least effect on polarization, making only a small difference at low field levels (50% Ec), and virtually no improvement for higher fields. The effect of temperature is much more pronounced, as it actually changes the coercive field, so that the poling becomes more complete at the lower electric fields. For 90°C poling, polarization is complete at 100% Ec.

---

8 The coercive field was measured at 1 Hz - low frequency so that dynamic effects in the composite can likely be neglected, and the composite coercive field can be related to the bulk value.
and almost 90% complete for 50% Ec (indicating that the coercive field has probably been reduced to approximately 1/2 of its room temperature value). In fact, increased temperatures may be necessary for full polarization. The increase in capacitance for room temperature poling is relatively constant for multiple field levels (100%, 150%, 200% Ec) and time (10, 100, 1000 seconds), and yet the poling is incomplete. It is important to stress that full poling is not accomplished at field levels equal to the measured so-called ‘coercive’ field, or at even twice the value.

Given these results for the bulk ceramic, it is possible to apply successful poling conditions to the composite material. This is done by measuring the composite coercive field at room temperature, and applying the required field according to this metric. The conditions chosen for poling the composites are 60°C poling, with a field of 200% Ec applied for 1000 seconds. Temperatures above this level do not improve polarization, and are too high for the epoxy used in the M series composites. These levels are used for all specimens manufactured for comparison with models, described in Chapter 5.0.

The results of the aging study are shown in Figure 2.6, illustrating the change of capacitance over the unpoled value, as a function of time for two separate PZT 5H samples. Both show a similar trend - an increase within the first 10 minutes of poling, followed by a steady decrease. As predicted in the reference (Jaffe et al., 1971), the decrease is linear with decade changes in time. For PZT 5H, this change appears to be about 2% change in dielectric constant per decade. What is not explained is the initial increase in capacitance, seemingly indicating that dipole alignment is occurring after removal of the poling field.

It is also interesting to note that the change of capacitance at 24 hours (approximately 1400 min) is significantly less than that found for fully poled ceramics in the poling study. In the latter, an increase of 62% was found, compared to 46% here. This may confirm the need for increased temperature to complete poling. (Remember that
Figure 2.6 Illustration of aging phenomenon in poled PZT 5H, shown as a change in dielectric constant from unpoled value as a function of time.

the disks used in this study were poled at 300% Ec, at room temperature). Since the composites will be poled at 60°C, they will be fully poled. Interestingly, it is likely that the increased temperature poling also accelerates the aging process (Mason, 1955). This would have the affect of compressing the first two decades in Figure 2.5. Regardless, the linear reduction per time decade means that testing after aging for at least 24 hours, with a window of 9 days, will result in errors of only 2% for the dielectric, and likely less for the coupling coefficient (and other piezoelectric constants). This is to be expected for a ceramic that is harder than the Barium Titanate, which had a 4%/decade decrease in dielectric and 2.5%/decade decrease in coupling coefficient (Jaffe et al., 1971).

2.2.3 Fiber Characterization

Introduction

All circular piezoceramic fibers used in this study and in all previous work in piezoelectric fiber composites (Bent, 1994; Rodgers et al., 1996) were commercially produced by CeraNova Corporation. This company has been involved in manufacturing PZT fibers since approximately 1992, and are currently the only manufacturers of PZT fibers at this scale and semi-continuous in length. The process involves processing a mixture of oxides and binder which is extruded through a small orifice die as a continuous green (unfired) fiber strand. This spooled green is cut to length (3.5” to 5”) in order to allow easier handling, and sintered (fired) in a lead atmosphere to avoid loss of the lead oxide volatiles. The firing process is responsible for densifying the ceramic, grain growth, and creating intergranular bonds which provide the basic polycrystalline ceramic strength.
Achieving full piezoelectric properties in the final ceramic product is a difficult task. Some of the key parameters affecting the quality are:

- raw oxide purity and particle size distribution
- thoroughness of oxide preparation/mixing
- firing atmosphere in calcination and sintering for retention of volatiles
- firing time and temperature profile for optimal grain growth and densification

Each of these parameters can substantially affect the final strength, size, and piezoelectric properties, for very small variations in the conditions. This difficulty is compounded by the small size of the parts being manufactured. This section examines the commercially available fibers in order to determine whether the material piezoelectric properties are similar to the bulk reported properties for PZT 5H. This information can then be used when considering the material properties to be used as inputs for the composite models.

**Experimental Method**

The data taken experimentally in this study involved measuring the free strain and coercive field of individually electroded fibers. The entire length of a PZT fiber (~20 mm) was electroded with colloidal silver, except for a center region of 2-3 mm, which was kept as the 'active length' (Figure 2.7b). Since the measurements were performed quasi-statically (1 Hz voltage signals), it was assumed that the effect of electrode material would be negligible. In this configuration, uniform potential is maintained under the entire electrode region at each end, with a uniform potential gradient across the active length. A small amount of distorted field will occur in the transition area (similar to that for interdigitated electrodes). The effect of field fringing outside the fiber is negligible due to the high dielectric constant of the fiber, as compared to air. This was confirmed in finite element models (Chan and Lilienkamp, 1996) for high aspect ratio ceramic cubes, electroded at the ends. Due to the small scales, it was difficult to a priori set the active length for each fiber. The lengths varied between 1.56 mm and 3.13 mm (to give an aspect ratio of active length to fiber diameter between 12 and 24 for the 130 μm fiber). Difficulty with breakdown in the lead wire attachments and other areas prohibited larger active lengths from being used. Following electroding, the colloidal silver was dried in an oven at 120°C for 20 minutes to maximize conductivity. Finally, the active length was measured accurately under a microscope at 40x magnification, using a graduated reticle.

The same laser interferometry system used for the PZT disk studies was also used here. A test fixture was designed and manufactured for general planar strain measurement, including fibers and composites (Figure 2.7a). The maximum specimen length that could
Figure 2.7 (a) Planar displacement measuring device, shown as used for measuring free strain properties of single PZT fibers, (b) The test fiber dimensions, illustrating active length.

be accommodated was 8", and the minimum was restricted only by the laser resolution and the ability of the sample to move the sliding reflector. One end of the sample was held stationary by a fixed clamp, while the other end was held in a sliding clamp that incorporated a retroreflector for displacement measurement. All surfaces were covered with Teflon to minimize frictional forces. For measuring individual fiber strains, each fiber was bonded to glass microscope slide covers with epoxy, which were then clamped as described. Lead wires were conductively attached to the exposed ends of the fibers using the same colloidal silver. The active length of each fiber was coated with silicone oil to avoid breakdown for fields moderately above the breakdown in air. An estimate of frictional force was calculated, and found to have negligible effect on fiber displacement.

The outputs that were recorded consisted of the applied voltage, displacement, and current. However, the signal noise was too high on the current output (without amplification circuitry) to provide polarization curves. For this same reason, capacitance measurements were not performed on the fibers. Such a capability does exist, however, at Penn State University, where devices for measuring the capacitance and polarization of single fibers have been developed (Yoshikawa et al., 1995). The fibers were not poled, due to the difficulty of poling at elevated temperatures (full poling is not achieved for room temperature poling). Thus, representative cycles and $d_{33}$ constants are also not reported for the fibers.
Table 2.2 Measured and expected results for single fiber testing. Standard deviations are shown as ± after the average.

<table>
<thead>
<tr>
<th></th>
<th>Diced Monolithic 5A (square)</th>
<th>CeraNova 5H (circular)</th>
</tr>
</thead>
<tbody>
<tr>
<td>Number Tested</td>
<td>4</td>
<td>14</td>
</tr>
<tr>
<td>From Single Batch of:</td>
<td>1300</td>
<td>1000</td>
</tr>
<tr>
<td>Thickness/Diameter (µm)</td>
<td>114±10</td>
<td>129±1.6</td>
</tr>
<tr>
<td>Avg. Active Length (mm)</td>
<td>2.21 ±0.49</td>
<td>2.46 ±0.44</td>
</tr>
<tr>
<td>Bulk Ec (kV/cm)</td>
<td>11.2</td>
<td>7.2</td>
</tr>
<tr>
<td>Fiber Ec (kV/cm)</td>
<td>11.4±0.8</td>
<td>10.8±3</td>
</tr>
<tr>
<td>Bulk Max Strain (ppm)</td>
<td>4100</td>
<td>3200</td>
</tr>
<tr>
<td>Fiber Max Strain (ppm)</td>
<td>3213±218</td>
<td>2167±493</td>
</tr>
</tbody>
</table>

Results and Discussion

Results of the single fiber testing are shown in Table 2.2 and in Figure 2.8. The table summarizes data from the CeraNova fibers, and ‘fibers’ diced from monolithic wafers by Stavely Sensors. The latter was measured in order to validate the geometry and electrodes of the single fiber test. These fibers were sliced from monolithic wafers with bulk properties known to be of PZT 5A type ceramic, and had a square cross section with dimensions similar to the round fibers. Figure 2.8a shows the strain versus electric field, demonstrating the coercive field and maximum strains for a typical PZT 5A fiber tested. The coercive field of the 5A fibers was the same as for bulk material (~11 kV/cm), indicating that the full field strength was within the ceramic and validating the electrode geometry and electrode material for quasi-static testing. The average maximum strains were lower than expected for bulk ceramic 5A, approximately 78% of bulk. This may be due to the areas in the fibers near the electrodes, where the polarization is distorted and strains occur transverse to the fiber direction. The maximum strains were also seen to be proportional to active length, so that shorter active lengths (with lower aspect ratios) produced lower strains than the average.

The CeraNova 5H fibers showed a greater degree of disparity from bulk ceramic, and a larger variation in the measured properties. Fourteen fibers were tested from a single batch, and the butterfly curve from the best PZT 5H fiber tested is shown in Figure 2.7b. This curve has a shape very similar to the bulk PZT 5H. Once again, the average maximum fiber strain was lower than bulk, although lower in proportion than that

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9 Stavely Sensors
measured for the 5A fibers (approximately 67% of bulk). However, the key result is the much larger than expected coercive field, 10.8 versus 7.2 kV/cm, which indicates that the ceramic is not switching at the correct level. This in turn indicates a lack of bulk properties in the round 5H fibers, exhibiting electromechanical response similar to PZT 5A.

Although all fibers results reported came from a single batch of 1000 fibers, spot testing of other batches revealed a wide variation in actuation strain and coercive fields. To investigate whether the problem could be related to the microstructure, three fibers from a single batch (not from the batch in Table 2.2) were tested using the same procedure as above. Half of each fiber was used in the single fiber test, and the other half was examined using a scanning electron microscope, shown in Figure 2.9. Results showed a correlation between free strain and coercive field (taken at 20 kV/cm voltage input), and the fiber microstructure. Fiber 1 showed the lowest free strain (70 ppm) and highest coercive field (17 kV/cm) of the three fibers. This was due to the lack of complete densification, resulting in small grains and high porosity. This could be caused by firing conditions or the starting composition. Porosity has been shown to affect the coupling factor, dielectric, and remnant polarization, in a manner that is linearly proportional to the fired density (Buchanan, 1986).

Fibers 2 and 3 showed improved microstructure (less porosity, larger grains), which was also reflected in the better performance. Fiber 2 performed the best of the three, and had an average grain size of ~5μm. Fiber 3 performed less well, perhaps due to slight overfiring of the ceramic which caused larger grains (~10μm) and slightly reduced piezoelectric constants (Jaffe et al., 1971). Although not shown here, SEM studies were
Figure 2.9 Scanning Electron Micrograph of commercial PZT 5H fibers, showing correlation between grain size, porosity, and performance. Width of enlarged insert is ~25 μm.

performed on the PZT 5A fibers as well, which indicated typical grain size (2.5 μm) and low levels of porosity. This was expected for the bulk ceramic.

Although this study confirms the link between microstructure and performance, and further validates the usefulness of the single fiber test, it is how the fibers perform in an average sense that is important for use in the models. To this end, the type of results in Table 2.2 are the most valuable. Two other independent studies of CeraNova fibers have
shown similar trends. Measurements of 1-3 PZT/Epoxy rod composites\textsuperscript{10} at MIT showed coercive fields between 10.3 and 11.5 kV/cm, and dielectric constant approximately one third less than the value expected for the disks (Rodgers, 1995). These composite disks were approximately 0.5 mm thick and 4 mm in diameter, with a 50% volume fraction of fibers, oriented through the thickness.

Studies at Penn State using a specially developed charge measuring circuit investigated the remnant polarization and coercive fields of individual PZT 5H fibers and the square PZT 5A fibers (Yoshikawa, 1996). The polarization for the PZT 5H fibers was found to be between 180-220 mC/m\textsuperscript{2}, approximately two thirds of the bulk 5H value of 300 mC/m\textsuperscript{2} shown in section 2.2.1. The coercive fields were also 20-40% higher than bulk. As a consequence of these studies, \textit{the fiber dielectric constant and piezoelectric properties will be reduced by one third in the models when used for comparison to experimental data}. It is unlikely that the stiffness is affected as directly, although fibers with high porosity may show reductions in both stiffness and strength. Since the high porosity shown in Figure 2.9a is likely an exception to the average fiber, it will be assumed that the average fibers have full bulk stiffness. The polarization of the PZT 5A square fibers was found to be that of bulk (~400 mC/m\textsuperscript{2}), as expected.

2.2.4 Ceramic Characterization Summary

Four PZT ceramic types, ranging from very soft (5H) to very hard (8M) were compared for their capabilities in low field and high field regimes. PZT 5H was found to have the largest low field response, and was easiest to polarize. Other ceramics, such as PZT 5A showed better high field capabilities, while hard ceramics like PZT 4S and 8M were better suited for high stress applications. The best ceramic choice for active composites was not clear at this point in time, although PZT 5H was chosen for further characterization and the majority of the experimental work. Poling and aging studies were performed to better prepare for the use of PZT 5H in composites. The poling conditions necessary to maximize piezoelectric properties was determined as a function of the electric field strength and application time, and ceramic temperature. These can be related to the composite conditions through the easily measured coercive field. The aging study verified the aging trend of linear property reduction with time decade, and substantiated the standard 24 hour aging time found in most references.

Finally, to characterize the ceramic to be used in the actual composites, a method was developed to measure the free strain of individual PZT fibers. This work, and others,

\textsuperscript{10} The 1-3 connectivity of PZT rod composite disks offer easy modeling and measurement of longitudinal properties.

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showed a definite cut-down in PZT 5H fiber properties as compared to the bulk ceramic. Remnant polarization, dielectric, and coercive field data all indicate a one third reduction in piezoelectric and electrical performance. All of this information, coupled with the matrix data and the models of the next chapter will provide design capability for current and future actuators.

2.3 Matrix Characterization

2.3.1 Introduction

The matrix material must serve dual roles in an active composite. The matrix provides load sharing among the brittle fibers, and transfers load around fiber breaks when damage occurs. The matrix is also responsible for transfer of electric field from the electrodes to the active ceramic. For this role, the electric properties are of direct importance. First order models show that the electric field cutdown to the ceramic is dependent on the ratio of ceramic fraction along the field lines, and the ratio of dielectric mismatch between the matrix and ceramic. For most polymer based matrix materials, this mismatch will be on the order of 500-1000. Increasing the matrix dielectric can be accomplished by adding fillers, to be discussed shortly. The introduction of fillers, however, can have a number of effects on the matrix properties:

- **Rheology**: fillers can increase the matrix viscosity, causing difficulties in processing
- **Dielectric**: fillers can increase the dielectric, affecting performance
- **Resistivity**: fillers can change the resistivity, affecting poling
- **Toughness**: fillers can increase the brittleness of the matrix, affecting strength
- **Dielectric Strength**: fillers can lower the dielectric strength of the matrix

This study is focused on the matrix properties as they relate to performance - the ability to manufacture, prepare and use the composite for structural actuation applications. Issues of mechanical behavior (stiffness\textsuperscript{11} and strength) are not addressed here, and are left for future work.

Previous work (Bent, 1994) showed some success in increasing the matrix dielectric constant using dielectric fillers. The dielectric constant was increased almost fivefold (from 6 to 27) for an 36% by volume (80% by weight) use of 1 \( \mu \text{m} \) PZT filler.

\textsuperscript{11} The previous work in PFCs showed that even a tripling of matrix stiffness (from added fillers) would have little affect on the actuation capabilities of the composite, as the stiffness mismatch between fiber and matrix is still –6:1. Thus, the matrix stiffness is of little concern, except as it relates to stress recovery lengths in affecting composite strength.
Volumes of these levels have serious impacts on the matrix viscosity, and several dispersants and coupling agents were studied to lower the viscosity. Even at these elevated levels of filler, percolation effects were not apparent. Percolation in this context is the formation of random dielectric pathways between the electrodes, which results in a sudden increase in dielectric constant (or conductivity) at higher volume fractions of filler. For most fillers, this is reported to occur between 16% and 35% volume fractions. In the previous work, the increase in dielectric more closely followed the lower bound models, with no sharp increase in value up to 36% filler.

It may be that the percolation phenomenon may be more pronounced in conductively-filled systems, while in dielectric filler systems it may be more gradual. Conductive fillers are another means for increasing the apparent dielectric in the matrix. Much work has been done in the past on investigating the effect of silver and carbon fillers on polymer conductivity for conductive rubber and anti-static applications, typically at extremely high filler loadings. Percolation has been reported to occur at around 35-37% for silver particles, and 2.5-10% for carbon black which has a very fine (often porous) elongated structure which tends to agglomerate (Yacubowicz et al., 1990a). Percolation in conductively loaded polymers will likely be detrimental, as conductive paths will encourage dielectric breakdown during actuation, or the inability to pole due to the high loss currents.

One example of applying carbon black to active composites was used to increase the ability to pole in 0-3 particulate PZT-polymer composites by decreasing the matrix resistivity (Sa-Gong et al., 1986). The addition of 1.5% carbon black increased the dielectric by 20% and d33 constant by 10%, without causing the aforementioned problems. The challenge is to balance the improvements in matrix properties with the effect of these properties on the composite response. It is possible that the increase in dielectric constant may reduce the needed electric field faster than the dielectric breakdown increases. Another interesting result of tailoring relative material resistivities and dielectrics is the effect on frequency response of the composite.

Several more ‘exotic’ methods have been considered for increasing matrix dielectric. Dielectrophoresis is the alignment of dielectric particles in a chain by using the application of an alternating field. Thus, the particles could be aligned preferentially between the electrode and fibers, and percolation could be artificially created at low filler volume fractions. Dielectrophoresis is an exciting possibility, but represents at best a 2.5 times increase in dielectric (Bowen et al., 1994) in very controlled and stable conditions. Work at MIT has shown that filler chains are easily created in small samples, but that any flow of the matrix during cure destroys the chain formation. Another possibility is semi-interpenetrating networks (IPNs), which controls the polymer cross-linking structure, and
attaches submicron filler to specific sites on the polymer chains (Wang and Rubner, 1992). This creates a percolation threshold at very low filler ratios (2-3%). Again, the difficulty in application to the current problem arises with the maturity of the technology.

The current work will concentrate on applying conductive and dielectric fillers to the problem of improving the matrix properties. The following materials have been chosen for this study:

- **Shell Epon 9405 Resin**: a B-staging two-part epoxy chosen for its prepreg capability (allowing partial cure, storage, and then co-curing with other plies). This material is a structural bisphenol epoxy used for filament winding, RTM, and prepreg operations. The low viscosity and long pot life is ideal for the wet lay-up techniques used in manufacturing PFCs. Previous work used a low viscosity optical epoxy with short pot life (still used for the M series samples).
- **ICI Americas HYpermer KD-2 Dispersant**: a surface modifying agent that improves the wetting of the particles, promoting dispersion of the particles and lowering viscosity. Previous work showed this to be a good choice for an epoxy-PZT system.
- **Morgan Matroc PZT 5H Powder**: a 1 μm average diameter powder, used as the dielectric filler.
- **Penn Color Carbon Black Paste**: a pre-dispersed slurry of Columbian Conductex Ultra high conductivity carbon black in part A of a bisphenol epoxy. Used as the conductive filler.

The influence on the amounts of the above materials on matrix rheology, electrical properties (dielectric, DC resistivity, dissipation), and dielectric breakdown are studied in the following sections. In Chapter 4, the models developed in Chapter 3 are combined with the results from these sections in order to determine the material effects on the composite issues introduced at the beginning of this section.

### 2.3.2 Rheometric Studies

**Introduction**

There are a number of reasons for quantifying the viscosity of the matrix material. By measuring the viscosity of the matrix, it is possible to stipulate a limit on the maximum viscosity that can be easily processed and used in manufacturing of the composites. Then, tradeoffs of filler effectiveness with the ease of processing can be examined. In general, it is advantageous to have a lower viscosity matrix system. Lower viscosity allows better composite compaction under cure pressure, better wetting of the fibers, less damage to
fibers, and reduced void content. Each of these can be related to performance, whether
directly (better actuation with higher compaction), indirectly (higher strength from better
wetting), or in an absolute sense (dielectric breakdown at a void).

Additional filler materials work against the need for lower viscosity. In addition,
untreated fillers can often result in serious embrittlement of the matrix, increased moisture
sensitivity, and a deterioration of strength and electrical properties (Berger and Petty,
1987). As previous work has qualitatively shown, significant improvements can be
achieved using surface modifying agents, or, surfactants. In the previous work, a
particular dispersing agent (Hypermer KD-2, a class of surfactant) was found to improve
the filler dispersion and decrease the viscosity substantially in the epoxy-PZT filler system
used. This chemical additive is in itself a polymer chain, with two functional groups: a
hydrophilic end that readily adsorbs to the surface of the filler (often coated with moisture),
and a hydrophobic tail which orients itself away from the particle and bonds or entangles
with the main polymer chains. Dispersion is facilitated by the similarly oriented chains
extending from each particle, which now appear ‘slippery’ to each other. Interestingly
enough, the use of this dispersant also increased the matrix dielectric, perhaps due to the
highly polar nature of the dispersant.

The first purpose of this section is to quantify the effect of KD-2 on reducing the
viscosity of the particular epoxy/PZT filler system chosen. Approximately a dozen other
surfactants have been previously studied (Bent and Kucera, 1993), with limited success to
PZT fillers. The 1 μm diameter powder was assumed to be optimal for this purpose.
Smaller particles increase surface area, causing higher viscosity for similar volume
loadings, and a reduction in effectiveness due to increased surface resistivities (Ruschau et
al., 1990). Larger particles may begin to interfere with the composite compaction. The 1
μm size is already ~1% of the fiber diameter. The effect of carbon black on viscosity is
ignored, as only small levels of loading will be used, on the order of 2% to 5% by weight
(1.5% to 3.5% by volume).

The second purpose of this section is to obtain data which will allow design
tradeoff decisions in processing with raised temperatures. Higher temperature matrix will
reduce the viscosity, enabling higher particle loadings for the same working viscosity.
This information can be combined with the later composite results such as compaction and
voids.

**Experimental Method**

The viscosity measurements were taken using a Brookfield viscometer, with
temperature capability provided by Neslab components, as shown in Figure 2.10. The
Figure 2.10  Rheology experimental setup, showing viscometer and temperature control equipment (high temperature bath and chiller).

Brookfield viscometer (DV-LVII+) uses the cone/plate measurement technique for determining the viscosity. The viscometer is capable of measuring small sample quantities (0.5 ml) in the measurement cup, and has an accuracy of 1% of the total working range. Viscosity in the range of 300 centipoise to 1 million centipoise could be measured with the available spindle. Temperature stability was provided by circulating silicone oil in a closed loop system consisting of Neslab heater (EX-250HT) and chiller (FTC-350A) components. Sample temperature was monitored through an embedded temperature probe located in the sample cup, which was accurate to within 0.1°C.

Setpoint calibration of the viscometer was performed prior to measuring each sample, done in accordance with the manufacturer’s instructions. The matrix was mixed using the standard technique described in Section 4.2. In order to reduce the effect of possible filler settling, a 0.5 ml portion of matrix was placed in the sample cup immediately after the sonication step, and the spindle started.

The first study involved determining the effect of dispersant amount on viscosity at room temperature. For these measurements, the sample was allowed to stabilize (after sonication) at the test temperature for twenty minutes, prior to taking any readings. All measurements were made at 25.0 °C. Since the measured viscosity can depend on the shear rate at which it was measured (especially for the heavily loaded samples), the reported measurements are compared as close as possible to the spindle speed of 1 rpm\textsuperscript{12}.

\textsuperscript{12} The available shear rates depend on the viscosity of the material. The viscometer permits measurement only when the shear rate requires between 1\% and 100\% of the available motor torque, where accuracy is within the specifications.
Experience in previous work with PZT dispersions dictated the useful range of fillers and dispersant to be studied (Bent, 1994). The PZT filler ratios chosen were 60%, 75%, and 80% by weight. Since the specific gravity of PZT is 7.5 and epoxy is 1.2, this represents PZT volume ratios of approximately 20%, 30%, and 40%. The low end (60%) is driven by achieving noticeable increases in dielectric, while the high end (80%) represents viscosities just measurable with the available equipment. For the dispersant, the theoretical monolayer coverage for 1 μm filler is ~2% by weight. Perfect coverage is unlikely, and the previous work showed advantages for higher levels, so that the range of 0% to 5% by weight was studied. At values higher than 5%, the dispersant becomes a significant portion of the liquid material (especially for 80% PZT), and the resulting cured matrix is poorly polymerized and highly viscoelastic.

The second study involved measuring the viscosity of a matrix sample through a range of temperatures. Three samples were measured, 60%, 75%, and 80% PZT filler loadings, each with 5% KD-2 surfactant (the maximum value used in the first study). As before, the sample was placed in the sample cup immediately after sonication, and allowed to stabilize at 25°C for twenty minutes. Measurements were taken in 5°C increments from 25°C to 50°C. The highest temperature was chosen from composite manufacturing experience, where 50°C was the limit for practical reasons. These included convenient handling of hot components during lay-up and degassing, and avoiding voids created from volatiles that boiled out at raised temperatures. The temperature was raised slowly (~0.5°C/min) to permit the matrix sample to remain in thermal equilibrium as much as possible. At each setpoint temperature, the reading was taken after approximately 2 minutes. Due to the wide range in viscosities over temperature, the shear rate could not be kept the same between PZT samples, although they were held constant within a sample. In this case, 100 rpm, 12 rpm, and 0.6 rpm are used for 60%, 75%, and 80% respectively, as it permits measurement throughout the whole range of temperatures at a single spindle speed. However, this may affect direct comparisons between the first and second study.

Results and Discussion

The results of the first rheology study are shown in Figure 2.11a, as the log of viscosity for varying percent dispersant. Higher levels of PZT filler benefit the greatest from the addition of dispersant to the matrix. A sharp decrease in viscosity accompanies the first 1% of dispersant added, with continuing decreases which eventually level off. This is most apparent in the higher PZT loading levels, where the 75% drops an order of magnitude for the first 1% and the zero dispersant 80% is beyond the range of measurement. The more rapid leveling off of the viscosity for lower PZT loading (60%)

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Figure 2.11 Viscosity of matrix material (a) as a function of Hypermer KD-2 dispersing agent, shown for three PZT 5H filler ratios (by weight), and unloaded epoxy. Data taken at 25.0°C; (b) as a function of temperature for three PZT 5H filler ratios, and 5% KD-2 dispersant.

may indicate more thorough particle coverage from easier processing. The viscosity of pure epoxy (0% PZT) for various amounts of dispersant shows little change over the range of 0% to 5% dispersant. This demonstrates that the effects seen in the loaded matrix materials are not due to any dispersant thinning mechanism, but are due to actual changes in filler surface properties. The viscosity of the pure dispersant is 200 cps at 25°C, as measured by the manufacturer.

The second rheology study is shown in Figure 2.11b, as matrix viscosity versus temperature. Note that the 25°C data for the three PZT loadings do not match the points shown in Figure 2.11a. This is due in part to the different shear rates used, and likely in part from the different batch of PZT filler used. The second batch of PZT particles was thought to have a different particle diameter distribution. For the lower PZT filler ratios, the trend of log viscosity with temperature is very linear. This is not quite the case with 80% PZT, which was also found to be highly sensitive to the shear rate at which the sample was measured. In all cases, the viscosity dropped significantly over the 25°C range. The 80% PZT dropped below the room temperature viscosity of the 60% PZT sample, even though it contained twice as much filler by volume. This information will be used in quantifying tradeoffs between processing and performance in the later chapters.

2.3.3 Electrical Property Studies

Introduction

The purpose of this section is to determine an appropriate electrical material model representation of the bulk materials, and to characterize the parameters of the model for
inputs to the composite models in the next chapter. A typical representation will be some combination of electrical elements (resistors, capacitors, etc.) in series and parallel to model the observed phenomenon of the material. The complexity of the model depends upon the homogeneity and type of material being modeled, the frequency range of interest, and the needed fidelity of the model. A three parameter material model that has been used in the past to model carbon black composites (Yacubowicz et al., 1990) is shown in Figure 2.12a. This model captures the DC (static) and first order AC (dynamic) effects of a dielectric insulating material. This is accomplished through a combination of a resistor and complex capacitance, the latter of which represents the lossiness of the dielectric material.

As it will be shown, this model is quite appropriate for the bulk materials in the current active composite. The design of IDEPFCs is focused on the composite and bulk behavior at DC for poling, and at frequencies between 1 Hz and 10 kHz for operation. The matrix and bulk PZT material will be moderate to high dielectrics, with some loss behavior tied to the polarization of the material. It is expected that any polarization relaxation modes will occur outside the frequency range of interest. Electro-mechanical resonances in coupled materials (i.e. PZT) is geometry dependent, and will also generally be above the modeled frequency range of interest.\(^\text{13}\) Thus, a non-resonant material model will suffice.

The three parameter material model admittance can be written as

\[
Y = \frac{I}{V} = \frac{1}{R} + i\omega C^* \tag{2.6}
\]

where the complex capacitance is given by,

\[
C^* = C' - iC'' \tag{2.7}
\]

\(^\text{13}\) These resonances are also modeled using an electrical equivalent circuit, known as the oscillator circuit, whose parameters can be used to back out the material constants, such as the stiffness, piezoelectric coupling factor, etc. (IRE, 1958).
where $C'$ is the real portion of the capacitance, and $C''$ is the imaginary portion that represents the dielectric loss. The complex capacitance can also be written as a function of the dissipation ($D$), which is the ratio of the imaginary and real capacitance, giving a measure of the relative lossiness of the material:

$$C^* = C'(1 - iD)$$

(2.8)

The electrical equivalent material model can then be represented by the three following parameters: $R$, the intrinsic DC resistance; $C'$, the capacitance; and $D$, the dissipation. All of these elements may be a function of frequency to some degree. Note that the dissipation adds with the real part of the admittance, and appears as a resistance that varies directly with the inverse of frequency. Figure 2.12(b) shows the vectorial combination of electrical components, with $\theta$ representing the phase between the applied voltage and resulting current. The tangent of the phase angle of the equivalent circuit is given by

$$\tan(\theta) = \frac{\omega C' R}{1 + \omega C' R D}$$

(2.9)

Note that as $\omega \to 0$, $\tan(\theta) \to 0$, and the current becomes strictly a function of the DC value for resistance. As the frequency is increased and $\omega \to \infty$, $\tan(\theta) \to 1/D$, giving rise to the familiar definition of dissipation

$$D = \tan(\delta)$$

(2.10)

The angle $\delta$ (known as the loss angle) is usually used in describing capacitors, and represents the AC loss portion of the dielectric. The frequency response that is represented by this material model is illustrated in Figure 2.12(c). At some frequency, $\omega_0$, the lossy portion of the dielectric will dominate the real admittance. For the constituent materials considered here, this frequency will generally be below the bandwidth of AC measuring devices (LCR meters, etc.), and it will not be possible to capture all three parameters with a simple frequency response measurement. Instead, a separate DC resistivity measurement will be necessary.

The above description is useful for describing the physical nature of the materials to be characterized, and represents real material parameters that will be measured. For incorporation into models it is actually more convenient to use a different format. Taking the time integral of the admittance gives the relationship between charge and voltage. Further, multiplying through by the geometry ratio $t/A$ where $t$ is the material thickness and $A$ the electrode area, creates an intrinsic description of the material variables, assuming a parallel plate configuration. The result is an expression that uses the same dependent and
Table 2.3 Typical values for bulk material electrical properties. (*Unpoled to poled values for PZT 5H)

<table>
<thead>
<tr>
<th>Material</th>
<th>Description</th>
<th>Dielectric $\varepsilon'/\varepsilon_\rho$ (1 kHz)</th>
<th>Dissipation $D$ (1 kHz)</th>
<th>Resistivity $\rho$ (ohm cm)</th>
</tr>
</thead>
<tbody>
<tr>
<td>PZT 5H</td>
<td>dielectric</td>
<td>2200-3400*</td>
<td>0.017</td>
<td>2e14</td>
</tr>
<tr>
<td>Carbon Black</td>
<td>conductor</td>
<td>$\infty$</td>
<td>$\infty$</td>
<td>1e-3</td>
</tr>
<tr>
<td>Epoxy</td>
<td>insulator</td>
<td>4-5</td>
<td>0.015</td>
<td>1e15</td>
</tr>
</tbody>
</table>

Independent variables used in the constitutive equations (electrical displacement and electric field):

$$D = \varepsilon E$$

(2.11)

Where the total dielectric constant has AC and DC portions,

$$\varepsilon = (\varepsilon^* - \frac{i}{\omega \rho})$$

(2.12)

And a complex AC component that reflects the lossiness of the dielectric,

$$\varepsilon^* = \varepsilon'(1 - iD)$$

(2.13)

Where $\varepsilon'$ is the real dielectric constant, $\rho$ the resistivity, and $D$ remains the dissipation. Equation 2.12 will replace the simple dielectric in the modeling efforts when examining the complex material behavior for composite design (Chapter 4).

Table 2.3 gives some typical values for each of the major materials used in the composite. PZT 5H has high dielectric constant and high resistivity, while the epoxy can be expected to have low dielectric and high resistivity. The effect of adding PZT filler to the pure epoxy on dielectric is easy to predict, and has been shown to approximately follow lower bound models of two-phase materials (basically, materials in series) up to percolation. This is due to its simple spherical structure, and ease of dispersion. This will generally not be the case for conductive carbon black fillers. Carbon black is a highly structured (porous, non-spherical) particle, which tends to form agglomerated chains even below the system level percolation. Thus, models have not been successful in predicting this phenomenon accurately.

The effects of a second material phase on the resistivity is also difficult to predict, and can depend on phenomenon other than simple ohmic laws (the series lower bound). The DC conductivity in carbon black particulate composite systems depends also on a combination of tunneling and constriction resistances (Ruschau et al., 1992). Tunneling describes the phenomenon of electrons hopping from conductor to conductor, and
introduces the concept of a barrier energy to flow (Buchanan, 1986). Thus, the conductivity becomes voltage potential dependent. Constriction resistance is related to the area of particle-particle contact, and depends on the ratio of contact surface area to diameter of the particle. This difficulty in predicting behavior is further compounded by the morphology of the carbon black particles.

Obviously, this points to the need for empirical results for the matrix material parameters. Knowledge of all three parameters is essential to the design of the composites. The dielectric of the matrix is responsible for determining field in the fiber, and thus composite capability. The resistivity of the matrix may affect the DC poling voltage and current requirements, and the matrix dissipation will determine the operational power requirements. The ability to tailor these material parameters leaves open the possibility for also designing the frequency response of the composite. i.e. for passive damping applications. Obviously, all three parameters cannot be specified independently, so it will also be important to examine the relationships between the various parameters with filler ratios.

**Experimental Method**

**Sample Preparation**

Three series of samples containing 0%, 50%, and 75% PZT filler (by weight) were manufactured. For each series, the carbon black percentage was varied between zero and an upper level, depending on the amount of PZT filler. For the 0% PZT series, up to 9% carbon black (by weight) was mixed into the matrix. The series of 60% and 75% PZT were limited to 4% and 2% carbon black, respectively, due to the concentration of the starting pre-dispersed carbon black. The pre-dispersed carbon black slurry was a 15% by weight mixture of carbon black, which represented a practical limit on content. Higher percentages were definitely possible, but with a sharp decrease in efficiency (dielectric increase with added carbon). This was due to the need for much higher dispersant content which caused a loss in conductivity\(^ {14}\). However, it was expected that these ratios would cover the design regions of interest. The matrix samples were mixed according to the baseline method described in section 5.2.2, with the exception that the pre-dispersed carbon black was added to Part A at the first step. As usual, dispersant was added to the mixture at 5% of the PZT weight fraction, and the matrix was sonicated before use.

Samples were made in disk form, with dimensions 25 mm (1") in diameter and

\(^ {14}\) Based on recommendations from Penn Color. Slurries with increased levels of pre-dispersed carbon black tested at MIT confirmed this, finding nearly twice as much carbon black paste was required to maintain the matrix dielectric.
~0.41 mm (0.016") in thickness, giving an area aspect ratio of approximately 1200 mm²/mm (50 in²/in). High ratios minimize the errors in both the AC and DC measurements. The samples were formed by pouring the matrix into a solid Teflon mold that had been machined in the correct dimensions. The matrix was de-gassed to remove voids, and a flat Teflon top cover was placed on the mold. The matrix samples were cured according to the epoxy manufacturer's guidelines (350°F for 3 hours). Following cure, the samples were polished with 1200 grit sandpaper to improve flatness, and electroded with colloidal silver.

DC Measurements

ASTM D257-93 Standards summarize the issues related to measurement of DC resistivity in insulating materials. Two of the most important test variables are the applied voltage (field) level, and time of electrification. Some variation with electric field level is expected due to the nature of the conduction phenomenon. Thus, all samples were measured in the range of 40 to 100 volts. Time of electrification is important to specify due to the large time constants inherent in these materials. After the potential is applied to the sample, the current generally decreases asymptotically toward a limiting value, following the relationship (ASTM D257-93, 1993),

\[ I(t) = At^{-m} + I_o \] (2.14)

This effect is due to interfacial and volume charging, causing the true leakage current to be indicated only after this charging is complete, which may take from seconds to hours. It was expected that this time would vary with the sample type tested, so that the resistance was taken when the value was changing less than approximately 1% per minute, rather than at a set time.

Measurements were performed with a Keithly 6517 electrometer capable of measuring resistances in the range from 10 ohms to 210 TeraOhms (210e12), using the "direct method" of measurement. This refers to the application of a known voltage, and measuring the resulting current, which has been amplified to increase sensitivity. The device error is specified to be a maximum of 1.15% of the total working range. Error in the measurement setup was a moderate concern, given the range of resistances expected and the sample geometry. The measurement was performed using a two probe technique in a shielded enclosure to reduce signal noise, as shown in Figure 2.13a. Figure 2.13b shows the wiring configuration to the test device. The sample was clamped in a holder designed to minimize leakage charge, both to ground and across the sample through alternate paths. The holder was constructed of a highly resistive material (Lexan). Both the holder and the sample were thoroughly cleaned of oils that might reduce the surface
resistivity, and reduce the apparent resistance of the sample. By monitoring current without a sample in the holder, the leakage charge was found to be insignificant.

**AC Measurements**

Methods used here have been adapted from standard test methods for AC measurements of materials, summarized in ASTM D150-94. Samples were measured using a two probe technique, in the holder described in the previous section. Guard electrodes were not used, as the high aspect ratio creates a relatively high capacitance test sample. This reduces the fringing capacitances and stray capacitances (holder, leads) in relation to the sample.
Measurements were taken using a HP 4194a impedance analyzer, which utilizes an auto balancing bridge measurement technique. This analyzer is capable of measuring impedance functions from 100 Hz to 40 MHz. Output values can be specified directly in equivalent circuit components (such as D, parallel/series capacitance and resistance, etc.). Voltage applied to the sample varied from 0.01 V to 1 Vrms, depending on the frequency and range. Parallel capacitance and dissipation were recorded for frequencies in the range of interest, at 100 Hz, 1 kHz, and 10 kHz. Typically, material capacitances and dissipation are reported at 1 kHz, and this is also done here.

**Results and Discussion**

The results of the different electrical properties are given in Figures 2.15-17. Figure 2.15 shows the matrix dielectric as a function of the carbon black filler, for three different families of PZT filler. Due to the slow task of manufacturing individual samples, the data points generally represent only a single sample, except for the cases of 60% PZT with 0%, 3% and 4% carbon black (designated (60,0) (60,3) and (60,4) respectively). For these cases, several points were measured to determine the range of results.

For the pure carbon black samples (0, X), the dielectric appears to increase gently with added carbon. However, this belies the true difficulty with carbon black and percolative chain formation. Some points (not shown for clarity) include another (0, 4) that had a dielectric of 340, and two (0, 7) points with dielectrics of 103 and 355. These type of results show the randomness of carbon aggregation that can radically affect the electrical properties, even with extremely careful dispersion during manufacture of the samples, and careful measurement techniques.

The samples with 60% PZT show a similar problem, but smaller in magnitude. On

![Figure 2.15 Dielectric constant of matrix samples as a function of carbon black (wt%) loading, shown for three series of PZT loading (wt%). Data taken at 1000 Hz.](image)
average, the dielectric of these samples also increase. However, for the (60,3) and (60,4) samples, these values ranged from no added benefit to a significant increase in dielectric with carbon black. The (60,0) samples had little variation, illustrating that the difficulty resided in the addition of carbon black. Only single data points were measured for the 75% PZT series, and these seemed to indicate an increase in dielectric from added carbon black. It is possible that additional points might have shown a similar range in variation as the 60% PZT.

Figure 2.16 shows the dissipation factor (D), defined in Equations 2.7 and 2.8, as a function of carbon black and PZT fillers. In general, the addition of PZT or carbon black gradually increases the dissipation. At higher levels of carbon black, the dissipation increases more quickly. This is caused by the lower resistance to charge flow, and appears as dielectric loss in the material model. Other researchers have shown that the dissipation maximizes after the percolation threshold, and decreases again (Yacubowicz and Narkis, 1986). For the results shown, it appears that this maximum is at higher carbon black concentrations. Once again, several points are shown for the (60,4) samples. This indicates that the dissipation is also affected by the random formation of chains (as expected).

For both the dielectric and dissipation, the results were found to be essentially flat with frequency in the 100 Hz to 100 kHz range, especially for matrix samples that contained PZT filler. Only the highly loaded carbon black samples, such as (0,7) and (0,9) showed any appreciable dependence on frequency, and this showed a variation of two to four times over the range. This is contrast to the sharp change that would
accompany a polarization relaxation mode in the frequency range of interest. This validated the use of the three parameter model in representing the materials used.

Little experimental data is available in the literature for comparison. Most work is interested in maximal changes in properties with the addition of carbon black filler, and thus deals with much higher filler fraction values. One work shows similar increases in dielectric and dissipation for carbon black (Conductex 40-220, in the same family as the material used here) in polyethylene (Yacubowicz and Narkis, 1986). Another work by the same researchers demonstrated the effect of the interfacial polarization on frequency response of the dielectric (Yacubowicz, 1990a). In the cases presented, all relaxation modes occurred well beyond 100 kHz. Only one work was found for combined PZT and carbon black fillers, which demonstrated an increase with carbon black filler at the same rate as the upper values for the 60% PZT samples (Sa-Gong et al., 1986). i.e. this work appeared to show the full benefits of chain formation. However, the samples were much more heavily loaded with PZT filler (~94% by weight, 70% by volume), so that direct comparison is difficult.

Figure 2.17 shows the DC resistivity as a function of the filler loadings. Reliable measurement of the resistivity was very difficult, in part due to the high values of resistance being measured, and likely in part due to the percolation phenomenon. A number of samples never reached an equilibrium value of resistivity, and are not reported here. The pure carbon black samples (0, X) appeared to follow almost a linear trend with increased filler. As in the other electrical properties, several points showed radically different values. For example, another sample of 4% carbon black (0, 4) which is not shown, had a resistivity of 1e6 (ohm-cm).
The addition of PZT to the pure epoxy increased the resistivity, but by only a small amount. As carbon black was added to the PZT filled matrix, the resistivity decreases, but not with any obvious trends. The range of values for the \((60,3)\) and \((60,4)\) samples are shown. Again, these ranged from almost no visible effect of the carbon black, to a significant decrease in resistivity. Although conclusions were difficult to make on the additive effects for the two fillers, the design chapter will show that adequate information was gained to make the needed conclusions on the composite frequency behavior and poling.

Again, little experimental data was available in the literature for the filler range of interest. For trends in resistivity, one group had shown tremendous variations in the resulting resistivity depending on the arrangement of the carbon black (Yacubowicz et al., 1990a). For very low structure carbon black that was completely dispersed, the resistivity was flat (no change) with increased carbon black up to ~10% by weight, followed by a sharp decrease to 1e4 ohm-cm by 15%. For highly structured and segregated carbon black, the resistivity dropped sharply to 1e4 ohm-cm with the first introduction of carbon black (values less than 1%). The current system is somewhere bounded by these two extremes. No results for resistivity of combined PZT and carbon black fillers were found in the literature.

2.3.4 Dielectric Strength Studies

Introduction

According to the ASTM Standards (ASTM D149-94, 1994), the following definitions are used:

- *Dielectric strength* of a material is ‘the voltage gradient at which dielectric failure of the insulating material occurs under specific conditions of test’

- *Dielectric failure* is ‘an event that is evidenced by an increase in conductance in the dielectric under test limiting the electric field that can be sustained’

The occurrence of dielectric failure is usually made obvious by breakdown which leaves permanent damage (such as a hole) in the material. Single phase materials, both insulators and dielectrics, usually have very high dielectric strength (DS). However, material imperfections and multiphase material mismatches can sharply reduce the strength. In the current composite system, a high dielectric mismatch between fiber and matrix causes field concentrations in the latter. This can cause breakdown prior to reaching poling fields within the fiber. An additional complexity is created when fillers are introduced into the matrix. The filler/matrix interaction length scale is small enough that the effects are
nonlinear and dependent on mechanisms of charge transport (Sichel, 1982), rather than predictable with an equivalent electrical network of conductors and dielectrics.

It is not the intention here to predict the dielectric strength using material models. The real interest is in measuring the relative changes in $DS$ for different filler ratios in the matrix. In Chapter 4, composite models will be used to predict the field concentrations in the matrix as a function of PZT and carbon black filler, given the measurements of the last section. Since the field concentration will decrease with higher filler loadings, it is hoped that this will occur quicker than any decrease in dielectric strength. The primary interest is in the DS of the composite in operation, so that the testing will involve AC breakdown. Several references have reported that DC dielectric strength is 2 to 4 times higher than the AC equivalent at 60 Hz (ASTM D149-94, 1994), and experience has shown that PFCs rarely break down during poling after surviving the same AC field level.

The intrinsic $DS$ of a system is the inherent ultimate dielectric strength that can rarely be obtained in practice (Saums and Pendleton, 1973). The reported $DS$ is usually much lower, and the mechanism of breakdown is related to thermal instability of the material. This is where the heat generated in material by conductive and dielectric losses accumulates faster than it can be removed. Since the $DS$ generally decreases with temperature, the result is a runaway condition. This implies that the dielectric breakdown is a time dependent process, and highly sensitive to the test/sample conditions. In testing materials, it is important to be aware of the factors that affect the $DS$. The following is from discussions in several references (ASTM D149-94, 1994; Saums and Pendleton, 1973; Sichel, 1982):

- **Ion Content**: ion content and mobility are key factors in $DS$, related to the DC resistivity and dissipation
- **Moisture**: wet $DS$ can be less than half of dry $DS$
- **Imperfections/Homogeneity**: voids or cracks result in field concentration, reducing the $DS$ measured for a material
- **Thickness**: $DS$ varies inversely proportional to a fractional power of sample thickness ($DS = C t^{-b}$, where $t$ is thickness, $C$ and $b$ constants). Thus, $DS$ increases with thinner samples, due to reduced statistical likelihood of flaws between the test points.
- **Electrodes**: $DS$ also decreases with increasing electrode area for same reason as above. It is also influenced by electrode geometry and materials, such as less conductive materials (silver epoxy or paint).
- **Frequency/Time**: DS is affected by changes in material losses with frequency, which is generally small except near resonances (such as polarizations).
- **Temperature**: Small variations in temperature have negligible effects on DS, although the general trend is a log(DS) which is linear with temperature.

**Experimental Method**

The matrix samples used in the electrical property studies were also used in the dielectric breakdown study. The colloidal silver electrodes were removed using methanol, and the samples were lapped to a thickness of approximately 6.5 mil (165 microns), and the surface polished to remove imperfections. The thinner samples were necessary due to the limitation of available voltage (10 kV). The samples were cut into four quarters, and all corners were sanded to reduce points where charge build-up could occur. Samples were dried in an oven at 110°C for 30 minutes to remove all moisture. Following drying, samples were stored in sealed containers with moisture-indicating dessicant until tested. Care was taken to avoid contamination of the samples with finger oil, dessicant, or other substances.

The testing was performed in a silicone oil test fixture, shown in Figure 2.18. Silicone oil surrounded the specimen to prevent flashover and partial discharges (ASTM D149-94). The oil was kept at room temperature (23°C). The sample was held between two 0.25” diameter hemispherical electrodes (electrode type #5 in ASTM D149-94), which were connected to the high voltage amplifier. Hemispherical electrodes make contact with a discrete point of the sample, in contrast to electrodeed samples which would result in measurement of the ‘weakest link’ dielectric strength of the entire sample. It is also likely that the DS would have a larger variation using silver epoxy or colloidal silver due to the inhomogenous nature of the material.

The *Method B, Step-by-Step* testing method (ASTM D149-94) was used in the

![Figure 2.18 Test Fixture for dielectric breakdown studies and capacitance measurements.](image)
application of voltage, with modification for the current specimens. Sinusoidally varying voltage was applied to the samples, stepped up in increments of 200 Vpp. This represented steps of approximately 5% of the breakdown voltage for the weakest samples. The frequency of the signal was chosen to be 100 Hz, which is likely typical of low bandwidth applications such as rotorblade control. At each time step, the voltage was held for 5 seconds soak time, and then immediately dialed to the next voltage. The starting voltage for each sample was at least 1000 Vpp below the breakdown voltage to ensure a test duration of at least five voltage steps, or 30 seconds. The recorded breakdown voltage is the highest level reached where the sample survived for the entire 5 second duration. Samples that experienced breakdown prior to 30 seconds were not included in the results.

**Results and Discussion**

The results of the breakdown tests are summarized in Figure 2.19, which shows the dielectric strength (kV/cm) versus the percentage carbon black. Three series of data are shown: 0%, 60%, and 75% PZT filler (by weight). Again, the percent carbon black was limited to 4% and 2% for the 60% and 75% PZT series respectively, due to the starting carbon black dispersion material. Each data point represents the average of between two and four samples.

The most evident result was the rapid drop in breakdown strength for the pure carbon black series. Up to 2% carbon black, the $DS$ was above the amplifier limit (~640 kV/cm), followed by a sharp decrease to 220 kV/cm for 3%. This transition was likely a percolation threshold for this system. Such a result may explain the difficulty with achieving reliable dielectric and resistivity for samples with greater that 2% carbon black (previous section). The large area measurements in those cases reflected the weakest path,

![Figure 2.19 AC dielectric breakdown fields for matrix disk samples at 23 °C, 100 Hz input voltage. Each point represents the average of three or four samples.](Image)

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which was highly susceptible to mixing and agglomeration. The breakdown tests, on the other hand, were likely more reliable since they reflect a typical cross-section. Such a rapid decrease in DS would seem to indicate that pure carbon black matrix would not be suitable for composites at these concentrations.

The addition of PZT filler lowered the initial breakdown strength of the matrix. The high dielectric mismatch between PZT and epoxy causes a field concentration, which may be accentuated by the smaller average gap sizes in the 75% PZT series. However, the addition of PZT seemed to delay or slow the percolation phenomenon by breaking up the otherwise continuous paths of carbon black. Thus, some combination of PZT and carbon black may be useful for application to the composites. Since the thickness of the matrix in the tests do not match those in the composite it is impossible to apply these values directly, but the relative changes in breakdown strength can be used in design.

2.3.5 Matrix Characterization Summary

This section involved the characterization of the matrix material in order to determine the properties as a function of conductive and dielectric fillers. This information will be used to determine the effect of fillers on the processing, performance and reliability of the composites. The range of fillers used for study was determined from previous work on PZT fillers for conventional piezoelectric fiber composites, and from the literature on carbon black filled composites. The first study on matrix rheology demonstrated the significant increases in viscosity with PZT filler, but showed that this viscosity could be reduced with the use of elevated temperatures. PZT loading and the processing temperature can be determined from the measured data once an acceptable manufacturing viscosity is known.

Measurement of the matrix electrical properties was based on a three parameter model description. Dielectric, resistivity, and dissipation factor were measured as functions of the PZT and carbon black fillers. All of these properties showed significant variation for multiple samples of the same filler amounts, illustrating the difficulty of using carbon black at even low concentrations. The effect of the carbon black on PZT filled matrix varied from no additional effect, to a doubling of the apparent matrix dielectric. The electrical properties likely depend on the effectiveness of the carbon black dispersion, which is not easily controlled. However, it is the percolative chain formation in poorly dispersed filler that maximizes the benefit of the conductive particles.

The goal of the fillers is an increase in the composite performance. However, this must eventually be weighed against the danger of dielectric breakdown and the mass penalty of the fillers. The third study of this section measured the dielectric strength of the
matrix samples as functions of filler. The data for this study showed well defined trends, and identified the percolation threshold in the range of 3% carbon black, where the dielectric strength dropped sharply. Added PZT fillers resulted in a stabilizing effect, reducing the severity of the drop in dielectric strength. This may be attributed to the PZT prohibiting the formation of continuous conductive chains.
Chapter 3
Linear Macroscopic Property Models of Coupled Field Multi Phase Materials

3.1 Introduction

A large portion of the effort in developing IDEPFC devices is directed at the modeling of effective composite properties. The utility of a good composite model is demonstrated in the ability to perform design for a variety of constituent materials, rather than relying on experimental trial and error. Models also generate an understanding of the issues involved, particularly the limitations and tradeoffs of a given design. The desired outcome of modeling is to obtain predictions of the composite response in the form of macroscopic constitutive relations. These effective properties then become a way to describe the averaged response of the material for use in applications, such as laminated structures that incorporate active composites.

Models developed in this chapter are combined with the material characterization of the previous chapter for property prediction of the actuators. This is used in the design studies of Chapter 4 which examine issues of performance, and in the subsequent chapters where the models are verified experimentally. To support this goal, the objectives for this chapter are

- to develop linear conservative models for the prediction of effective properties in coupled field, multiphase materials
- to validate the model assumptions through a comparison of the developed models
- to demonstrate the utility of these models in the prediction of composite properties for several cases

The models in this chapter are specifically for the current composite geometry and configuration. However, the methods used and issues examined will allow application to
other designs that incorporate both active and passive materials, such as those types shown in Figure 3.1.

This chapter begins with an overview to the approach of modeling for coupled field materials, discussing previous models in this area and the important elements of a micromechanical model. Two models are developed for the IDEPFC geometry. The first is a Uniform Fields approach, which is a generalization of the Mechanics of Materials method. Insights into the physical nature of the problem, evident in the closed form solution, are discussed. The second model applies a commercial finite element program to modeling the composite behavior. This section identifies the necessary conditions for the correct representation of composite response with a discrete model, including boundary conditions and load cases. The two models are compared to examine the effect of model assumptions and the regimes where the models are most valid. This is done through a comparison of composite properties for different constituent materials, over a range of fiber volume fractions. Finally, the models are used to compare the improvement in performance of the interdigitated approach over the conventionally electroded composite.

3.2 Modeling Approach

Previous Work

Micromechanical modeling began at least a hundred years ago (Rayleigh, 1892), when the technical advantages of multiphase materials became evident. Since that time, thousands of models for the prediction of mechanical and electrical properties have been developed, using dozens of different approaches. A quick review of any book on composite mechanics will reveal methods such as elasticity continuum methods, variational principles, finite elements, averaging techniques, and mechanics of materials approaches, to name the most common. Generally, these models are divided into predicting two classes of properties - transport properties (conduction, dielectric, permeability, thermal, etc.) and mechanical properties (stiffness and strength).
With the use of active materials becoming more common, the extension to micro
electro-mechanical models is necessary to accommodate the coupling between the material
fields. The previous work (Bent, 1994) developed the coupled governing equations for a
piezoelectric continuum, and illustrated the difficulty of exact solution of these problems.
Simplifications in the geometry or the field descriptions are usually made. There are also
certain classes of problems where the mechanical and electrical problems decouple due to
the choice of loading and poling directions. Two such Elasticity models were derived for
the anti-plane piezoelectricity problem (Pak, 1990; Honein et al., 1990). In this problem,
the authors consider a piezoelectric inclusion in an infinite media with far-field applied
electric field and out-of-plane shear. Pak used a series expansion for the displacement and
electric fields, while Honein et al. used complex potentials and conformal mapping to
produce a solution. Self Consistent Scheme (SCS) elasticity solutions have been applied to
piezoelectric fiber composites in two cases. One case applied this method to model the out-
of-plane behavior around the fibers at their ends in rod composites (Li and Sottos, 1994).
The second case applied this approach to the conventionally electroded piezoelectric fiber
composite (Bent, 1994) for determining the complete set of composite properties.

The Mechanics of Materials approach was probably the first method applied to the
modeling of active composites, due to its simple nature. Initial work was performed on
predicting the one dimensional response of piezoelectric particles in a soft matrix
(Newnham, 1980). Although the models ignored material coupling in other directions and
made critical assumptions on the bulk material properties, the authors were able to point out
predominant effects in the composite response. An extension of this work to the three
dimensional spherical inclusion problem also made many of the same assumptions (Banno,
1986). The move to a continuous ceramic phase was made for the prediction of the
effective stiffness, dielectric, and coupling coefficients in rod composites for ultrasonic
transducer applications (Smith and Auld, 1990). A general methodology for this approach
was developed and applied to conventionally electroded piezoelectric fiber composites
(Bent, 1994). This method will be applied to the current problem.

Finite element methods have only recently been used for determining effective
composite properties. In very few instances has this approach been applied to determining
constitutive relations for active materials (Hagood et al., 1993; Gaudenzi and Balhe, 1993).
The finite element method was successfully used for the modeling of conventionally
electroded piezoelectric fiber composites (Bent, 1994). This work will be carried over to
the current problem.
**Modeling Considerations**

According to one researcher, the term *micromechanics* implies 'the use of a model which accounts for explicit interaction at the level of a continuous matrix phase and one or more inclusion phases' (Christensen, 1990). The general purpose in modeling is to obtain a set of constitutive relations that describe the averaged response of the material. The manner in which these averaged properties are found involve understanding the mechanisms at the micro-level. At this level, the individual phases can be distinguished and form a series of repeating elements, often called representative volume elements (RVE). The choice of RVE is made to best approximate the randomness (or orderedness) of the particle/fiber in the composite, while accommodating the assumptions of the particular modeling technique. The macroscopic loads and boundary conditions of the actual composite are used to define those at the microscopic level. Through a particular solution method, the response of these individual elements is found, which can then be related to the response of the composite.

In order to define a complete micromechanics problem, four main elements must be considered in the model, illustrated in Figure 3.2.

1. The individual phases and their properties
2. A method for governing field distributions
3. The microstructure geometry
4. The appropriate boundary conditions

These four items are discussed in reference to the two modeling methods which will be applied to the IDEPFC problem: *Uniform Fields* and *Finite Element*.

The first element refers to the fact that each material phase must be represented in the RVE. That is, the RVE must contain each material, usually in proportion to the volume fraction in the composite. Each material is described by its individual constitutive relations,

---

*Figure 3.2* General approach to micromechanical modeling, illustrating the four necessary elements (1-4) that must be considered for any (physical based) model.
which are assumed to be homogeneous throughout the material, and linear and conservative in behavior. For example, the piezoelectric materials are assumed to be fully poled, and with response that is linear with the magnitude of the applied mechanical or electrical field. This will raise some issues when extending the linear models to the high field regimes of operation. The matrix material will also be assumed homogeneous, due to the length scale of the fillers, and is described by the empirically derived constitutive relations from Chapter 2.

The second element refers to the manner in which the distribution of fields are governed within a particular material phase. The Uniform Fields model, as the name implies, makes the assumption that all the fields are uniform within each material, and the magnitudes of these fields are governed only by material properties and volume fractions. The Finite Element method, on the other hand, allows for fully varying field distributions within each phase. These distributions are governed by the variational form of the continuum equations, applied in a discretized manner.

The third element deals with the manner in which the microstructure is represented in the particular model. The geometry of the IDEPFC problem has symmetries that allow modeling of a quarter section of the composite, as shown in Figure 1.5. Three planes of symmetry cut through the composite along the fiber length, and half way between electrodes. The dimensional variables are those defined in Chapter 1, where \( p, w, \) and \( h \) are electrode spacing and width, and sample height respectively. The RVE for each model is slightly different, and is described in more detail in the individual modeling approaches. Both models, however, assume uniform polarization along the fiber, even though the polarization would tend to follow the electric field lines and be distorted under the electrode surfaces.

The fourth item of modeling deals with boundary conditions, and is invariably tied to the RVE used. In this work, boundary conditions are applied to the RVE that are consistent with an arrangement of volume elements that would be self-compatible. That is, the composite is made of many of these elements, each of which behaves as the composite does. Thus, the boundary conditions at the RVE are the same as those of the macroscopic composite. This is known as the repeating element approach, and has been used extensively for modeling in the past (Hill, 1964; Hashin and Rosen, 1964). This is in contrast to some other methods which apply the boundary conditions at far field, or to an intermediate effective material phase.

A difficulty arises from the application of macroscopic boundary conditions to the RVE from the nonuniformity of the electrodes. In this case, there is a conflict in the length scales of the model. While the RVE is at the microscopic level, the top electrodes that
introduce the electrode field to the RVE are a macroscopic feature of the composite. This should not introduce much error as long as the assumed geometry is observed (p/h>>1, and p/w>>1), and only the global (averaged) response information is used. Local behavior near the electrodes, predicted by the FEM, will be only accurate for a composite that is one fiber diameter thick, and not for the generalized composite structure in Figure 3.2.

3.3 Uniform Fields Models

3.3.1 Introduction

The first model to be presented is the Uniform Fields model. The Uniform Fields approach is a generalization of the well known “Rules of Mixtures” which uses parallel and series (Voight and Reuss) additions to model effective properties of two-phase materials. In the past, effective constants have been found in this manner for transport properties (conduction, thermal coefficient, etc.) and for mechanical properties. More recently, this has been expanded to three dimensions and to accommodate electromechanical coupling (Bent and Hagood, 1993; Newnham et al., 1980; Banno, 1983; Smith and Auld, 1990). As the name implies, the governing assumption is one of entirely uniform mechanical and electrical fields within each material phase. Thus, the Uniform Fields Model (UFM) relies on simple combining rules to determine macroscopic field levels. In fact, the UFM method violates compatibility and equilibrium (and the corresponding equations of electro-statics) at some material interfaces. However, the large material mismatches make this method particularly well suited to modeling these types of composites. Furthermore, it has the advantage of providing a closed form solution that allows physical insight into the problem.

3.3.2 The Uniform Fields Model for IDEPFCs

Previous work (Hagood and Bent, 1993) presented a general methodology for modeling arbitrary multi-phase materials (particulate loaded, fibrous) which can provide a better prediction of transverse response. This method involves a combination of several Uniform Fields cases to obtain the appropriate geometrical description. The key to the current problem will be to bring this methodology to bear on the novel geometry present in the IDEPFC problem.
Figure 3.3 Uniform Fields Model (UFM) geometry: Cases A, B, and C, and combination model. $P$ represents the polarization vector.

The general approach to the IDEPFC is shown in Figure 3.3. The representative volume element consists of three separate cases which are used to capture the major contributions to the overall composite response. Uniform Fields Cases A and B together capture the predominant mechanical response and the electrical response away from the electrodes. Although the polarization will be distorted under the electrode, A and B assume a uniform polarization in the $3$ direction. Local electrical behavior around the electrode area, including the field concentration in the low dielectric matrix, is captured in Case C, where the matrix material represents the matrix that separates the electrode from the fiber directly beneath it. Case C is taken as a purely electrical combination of piezoceramic and matrix material. i.e. the matrix does not contribute to the overall composite mechanical properties, but only to the reduction in field reaching the fiber. In essence, the RVE is an unwrapping of the geometry along a path that follows the electrical field lines.

Each case may be analyzed separately to obtain the macroscopic properties for its geometry. The total combination model represents a combining of the three cases. In each instance, the effective properties of a completed case are substituted for the properties of the piezoceramic ($p$) phase in the next case. For each case (A, B, C), the piezoceramic ($p$) phase is kept fully orthotropic to accommodate the higher degree anisotropy that a composite produces.

The electromechanical shear mode problem is uncoupled from the normal mode of operation. Thus, the constitutive relations for a linear orthotropic active material can be represented by the following, from (IEEE, 1978),

85
\[
\begin{bmatrix}
T_1 \\
T_2 \\
T_3 \\
D_3
\end{bmatrix} =
\begin{bmatrix}
E_{11}^E & E_{12}^E & E_{13}^E & -e_{31} \\
E_{12}^E & E_{22}^E & E_{23}^E & -e_{32} \\
E_{13}^E & E_{23}^E & E_{33}^E & -e_{33} \\
e_{31} & e_{32} & e_{33} & e_{33}^\text{s}
\end{bmatrix}
\begin{bmatrix}
S_1 \\
S_2 \\
S_3 \\
E_3
\end{bmatrix}
\] (3.1)

where, for the pure piezoceramic, \( E_{22}^E = E_{11}^E \), \( e_{32} = e_{31} \), \( E_{23}^E = E_{13}^E \). \( S_i \) are the material strains, \( D_i \) the electrical displacements, \( T_i \) the material stresses, and \( E_i \) the electric fields. Constants \( c_{ij}^E \) are the stiffnesses, relating the conjugate mechanical fields, while \( e_{ij}^E \) are the dielectrics, relating the conjugate electrical fields. Electromechanical coupling is described by the 'clamped' piezoelectric constants, \( e_{ij} \). Note that all \( e_{ij} \) are zero for the matrix phase.

Stiffnesses and dielectrics for the matrix phase can be distinguished from the ceramic phase by their lack of superscripts, which otherwise indicate the mechanical and electrical boundary conditions. For all of the models to be presented, it is assumed that the poling direction is uniform throughout the piezoceramic and aligned with the 3-axis. This will be a good assumption for large p/h ratios, where the percentage of the distorted electric field under the electrodes is small.

Cases A and B can actually be represented by a single case because the lengthwise poling direction causes symmetry about the 3 axis. Thus, the effective properties for Case A will be found, and with a coordinate transformation, may also be used for Case B. The first step in applying the Uniform Fields approach is to recognize those field variables that are equal in the two phases as independent variables, and reorganize the constitutive relations as functions of those variables. In Case A, the fields are,

\[
\begin{bmatrix}
S_1 \\
T_2 \\
S_3 \\
E_3
\end{bmatrix}
\] (3.2)

For each of these field variables, the fields are equal in the two phases by virtue of the Uniform Fields assumption. For example, in the 3 direction the average strain (\( \bar{S}_3 \)) must be equal to the strain in each of the other phases:

\[
\bar{S}_3 = S_3^m = S_3^p
\] (3.3)

Rewriting the constitutive relations (3.1) as a function of these independent variables then gives (for the piezoceramic phase):

\[
T_1 = \frac{C_{33}E - C_{23}^2}{C_{22}^E} S_1 + \frac{C_{13}E}{C_{22}^E} T_2 + \frac{C_{13}E - C_{23}^E}{C_{22}^E} S_3 + \frac{C_{23}e_{32} - C_{22}^E e_{33}}{C_{22}^E} E_3
\]

\[
S_2 = -\frac{C_{33}E}{C_{22}^E} S_1 + \frac{1}{C_{22}^E} T_2 - \frac{C_{12}E}{C_{22}^E} S_3 + \frac{e_{32}}{C_{22}^E} E_3
\] (3.4)
\[ T_3 = \frac{c_{12}^E E}{c_{22}^E} S_1 + \frac{c_{12}^E E}{c_{22}^E} T_2 + \frac{c_{11}^E E}{c_{22}^E} S_3 + \frac{c_{12}^E E_{32}}{c_{22}^E} E_3 \]

\[ D_3 = \frac{c_{22}^E E_{33}}{c_{22}^E} S_1 + \frac{c_{32}^E E_{33}}{c_{22}^E} T_2 + \frac{c_{32}^E E_{33}}{c_{22}^E} S_3 + \frac{c_{33}^E E_{33}}{c_{22}^E} E_3 \]

The above can be written for the matrix phase by setting all \( e_y \) to zero and enforcing elastic isotropy. This may also be written more compactly as,

\[
\begin{bmatrix}
T_1 \\
S_2 \\
T_3 \\
D_3
\end{bmatrix}_{p,m} = \mathbf{A}_{p,m}
\begin{bmatrix}
S_1 \\
T_2 \\
S_3 \\
E_3
\end{bmatrix}_{p,m}
\]  

(3.5)

where \( \mathbf{A}_{p,m} \) is the matrix of constants from the above equations. The average stress, however, is some combination of stresses in the two phases, where the contribution for each is in proportion to the fraction of each phase. This is easily proven through a simple free body diagram. For example, for the ‘3’ direction,

\[
\bar{T}_3 = v_2^p T_3^p + v_2^m T_3^m
\]

(3.6)

where \( v_2^p \) is the fraction of piezoceramic (p) measured across the element in the ‘2’ direction. Thus, for Case A, these dependent field variables are the conjugates of (3.2):

\[
\begin{bmatrix}
T_1 \\
S_2 \\
T_3 \\
D_3
\end{bmatrix}
\]

(3.7)

Carrying out this combination for all directions simultaneously yields,

\[
\begin{bmatrix}
\bar{T}_1 \\
\bar{S}_2 \\
\bar{T}_3 \\
\bar{D}_3
\end{bmatrix}_{p} = v_2^p \begin{bmatrix}
T_1 \\
S_2 \\
T_3 \\
D_3
\end{bmatrix}_{p} + v_2^m \begin{bmatrix}
T_1 \\
S_2 \\
T_3 \\
D_3
\end{bmatrix}_{m}
\]

(3.8)

The dependent variables can then be replaced by Equation (3.5) for each material. Since the independent variables are equal in both phases,

\[
\begin{bmatrix}
\bar{T}_1 \\
\bar{S}_2 \\
\bar{T}_3 \\
\bar{D}_3
\end{bmatrix} = (v_2^p \mathbf{A}_p + v_2^m \mathbf{A}_m) \begin{bmatrix}
\bar{S}_1 \\
\bar{T}_2 \\
\bar{T}_3 \\
\bar{E}_3
\end{bmatrix} = \bar{\mathbf{A}} \begin{bmatrix}
\bar{S}_1 \\
\bar{T}_2 \\
\bar{T}_3 \\
\bar{E}_3
\end{bmatrix}
\]

(3.9)

Rewriting the above to return to the original form yields
\[
\begin{align*}
\begin{bmatrix}
\bar{T}_1 \\
\bar{T}_2 \\
\bar{T}_3 \\
\bar{D}_3
\end{bmatrix}
&=
\begin{bmatrix}
c_{11}^{\text{eff}} & c_{12}^{\text{eff}} & c_{13}^{\text{eff}} & -e_{31}^{\text{eff}} \\
c_{12}^{\text{eff}} & c_{22}^{\text{eff}} & c_{23}^{\text{eff}} & -e_{32}^{\text{eff}} \\
c_{13}^{\text{eff}} & c_{23}^{\text{eff}} & c_{33}^{\text{eff}} & -e_{33}^{\text{eff}} \\
e_{31}^{\text{eff}} & e_{32}^{\text{eff}} & e_{33}^{\text{eff}} & E_3
\end{bmatrix}
\begin{bmatrix}
\bar{S}_1 \\
\bar{S}_2 \\
\bar{S}_3 \\
E_3
\end{bmatrix}
\end{align*}
\]

(3.10)

where

\[
c_{11}^{\text{eff}} = (c_{11}^E v_2^P + c_{11}^m v_2^m) - \frac{v_2^P v_2^m (c_{12}^E - c_{12}^E)^2}{c_{11} v_2^P + c_{22}^E v_2^m}
\]

\[
c_{12}^{\text{eff}} = \frac{c_{22}^E c_{11} v_2^P + c_{12} c_{22}^E v_2^m}{c_{11} v_2^P + c_{22}^E v_2^m}
\]

\[
c_{22}^{\text{eff}} = \frac{c_{22}^E c_{11}}{c_{11} v_2^P + c_{22}^E v_2^m}
\]

\[
c_{13}^{\text{eff}} = (c_{13}^E v_2^P + c_{12} v_2^m) - \frac{v_2^P v_2^m (c_{12} - c_{12}^E)(c_{12} - c_{23})}{c_{11} v_2^P + c_{22}^E v_2^m}
\]

\[
c_{23}^{\text{eff}} = \frac{c_{23}^E c_{11} v_2^P + c_{12} c_{23}^E v_2^m}{c_{11} v_2^P + c_{22}^E v_2^m}
\]

\[
c_{33}^{\text{eff}} = (c_{33}^E v_2^P + c_{11} v_2^m) - \frac{v_2^P v_2^m (c_{12} - c_{23})^2}{c_{11} v_2^P + c_{22}^E v_2^m}
\]

\[
e_{31}^{\text{eff}} = e_{31} v_2^P + \frac{e_{32} v_2^P v_2^m (c_{12} - c_{12}^E)}{c_{11} v_2^P + c_{22}^E v_2^m}
\]

\[
e_{32}^{\text{eff}} = \frac{e_{32} v_2^P c_{11}}{c_{11} v_2^P + c_{22}^E v_2^m}
\]

\[
e_{33}^{\text{eff}} = e_{33} v_2^P + \frac{e_{32} v_2^P v_2^m (c_{12} - c_{23})}{c_{11} v_2^P + c_{22}^E v_2^m}
\]

\[
e_{33}^{\text{eff}} = (e_{33}^S v_2^P + e_{33} v_2^m) + \frac{e_{32}^2 v_2^P v_2^m}{c_{11} v_2^P + c_{22}^E v_2^m}
\]

(3.11)

It is interesting to note the effects of coupling in the above terms. The constants
\(c_{33}^{\text{eff}}, c_{11}^{\text{eff}}, \) and \(c_{11}^{\text{eff}}\) are simple one-dimensional parallel rules of mixtures softened by low stiffness matrix components which reduce the transverse clamping effects on the
piezoceramic. The $c_{22}^{\text{eff}}$, $c_{23}^{\text{eff}}$, and $c_{12}^{\text{eff}}$ stiffnesses are essentially series Rules of Mixtures results, with the latter two incorporating the Poisson’s effects. The clamped piezoelectric terms $e_{31}^{\text{eff}}$ and $e_{33}^{\text{eff}}$ are similar in nature to $c_{31}^{\text{eff}}$, where the basic Rules of Mixtures is accompanied by the transverse mechanical terms.

Since the direction of polarization is assumed to be uniform along the length of the fibers, the composite is axisymmetric about the 3 axis. Thus, Case A and Case B are equivalent representations of matrix and fiber combinations away from the electrodes. The independent variables in Case B are similar to those for Case A except that the axes 1 and 2 are reversed:

$$T_1 \quad S_2 \quad S_3 \quad E_3$$

To represent Case B, it is only necessary to interchange the subscripts 1 and 2 in both the effective and constitutive properties of the previous equations (3.2-3.11):

$$
\begin{align*}
  c_{11} & \rightarrow c_{22} \\
  c_{11}^E & \rightarrow c_{22}^E \\
  c_{11}^{\text{eff}} & \rightarrow c_{22}^{\text{eff}} \\
  c_{13} & \rightarrow c_{23} \\
  c_{13}^E & \rightarrow c_{23}^E \\
  c_{13}^{\text{eff}} & \rightarrow c_{23}^{\text{eff}}
\end{align*}
$$

etc.

and replace $v_2^p$ with $v_1^p$ to represent the fraction of active material measured across the element in the ‘1’ direction.

Only Case C remains to be determined. This case essentially determines the extent to which the electric field reaches the ceramic phase. Since the matrix has already been mechanically accounted for in Cases A and B, this case allows only an electrical contribution from the matrix material. For this case, the independent variables are:

$$S_1 \quad S_2 \quad T_3 \quad D_3 \quad (3.12)$$

Again, rewriting the constitutive relations of (3.1) so that the above variables are on the right hand side gives,

$$
\begin{bmatrix} T_1 \\ T_2 \\ S_3 \\ E_3 \end{bmatrix}_{p,m} = C_{p,m} \begin{bmatrix} S_1 \\ S_2 \\ T_3 \\ D_3 \end{bmatrix}_{p,m} \quad (3.13)
$$

where $C_{p,m}$ is the matrix of material constants in the rewritten form for the piezoceramic (p) or matrix (m) p,case, which are not shown here. The dependent variables are the conjugate fields of the ones given in (3.12). However, unlike the previous cases, the macroscopic
fields for all of these variables are not accurately represented by a weighted combination of the fields in the individual phases. This is true for the electric field, so that,
\[ \bar{E}_3 = \nu_p^3 E_p^3 + \nu_m^3 E_m^3 , \]
(3.14)
where \( \nu_p^3 \) is the fraction of piezoceramic phase across the 3 direction (Note that this is a function of the other ceramic fractions and IDEPFC geometry). The mechanical fields are \textit{not} a combination of the matrix phase and the piezoceramic phase. Rather, the macroscopic mechanical fields are \textit{equal} to the field in the piezoceramic phase. This is consistent with the assumption that the matrix does not contribute to the mechanical response of the Case C combination\(^1\). Thus,
\[ \bar{T}_1 = T_1^n \quad \bar{T}_2 = T_2^p \quad \bar{S}_3 = S_3^p \]
(3.15)

Applying (3.14) and (3.15) to the rewritten constitutive relations (3.13) gives the macroscopic description of the fields:
\[ \begin{pmatrix} \bar{T}_1 \\ \bar{T}_2 \\ \bar{S}_3 \\ \bar{E}_3 \end{pmatrix} = \begin{pmatrix} T_1 \\ T_2 \\ S_3 \\ \nu_p^3 E_3 \end{pmatrix} + \begin{pmatrix} 0 \\ 0 \\ 0 \\ \nu_m^3 E_3 \end{pmatrix} = \mathbf{C} \begin{pmatrix} \bar{S}_1 \\ \bar{S}_2 \\ \bar{T}_3 \\ \bar{D}_3 \end{pmatrix} \]
(3.16)

Finally, rewriting these into the original form gives the effective constants for Case C:
\[ \begin{pmatrix} \bar{T}_1 \\ \bar{T}_2 \\ \bar{S}_3 \\ \bar{D}_3 \end{pmatrix} = \begin{pmatrix} c_{11}^{\text{eff}} & c_{12}^{\text{eff}} & c_{13}^{\text{eff}} & -e_{31}^{\text{eff}} \\ c_{12}^{\text{eff}} & c_{22}^{\text{eff}} & c_{23}^{\text{eff}} & -e_{32}^{\text{eff}} \\ c_{13}^{\text{eff}} & c_{23}^{\text{eff}} & c_{33}^{\text{eff}} & -e_{33}^{\text{eff}} \\ e_{31}^{\text{eff}} & e_{32}^{\text{eff}} & e_{33}^{\text{eff}} & e_{33}^{\text{eff}} \end{pmatrix} \begin{pmatrix} \bar{S}_1 \\ \bar{S}_2 \\ \bar{T}_3 \\ \bar{D}_3 \end{pmatrix} \]
(3.17)

where the effective constants may be succinctly written as,
\[ c_{jk}^{\text{eff}} = c_{jk}^E + \frac{\nu_p^m \epsilon_{3j}^m \epsilon_{3k}^m}{\nu_p^3 \epsilon_{33}^m + \nu_m^m \epsilon_{33}^m} \]
\[ e_{3j}^{\text{eff}} = \frac{\epsilon_{3j}^S}{\nu_p^3 \epsilon_{33}^m + \nu_m^m \epsilon_{33}^m} \]
(3.18)
\[ e_{33}^{\text{eff}} = \frac{\epsilon_{33}^S}{\nu_p^3 \epsilon_{33}^m + \nu_m^m \epsilon_{33}^m} \]

---

\(^1\) This is equivalent to assuming all matrix stiffnesses are zero.
where the material constants \((\varepsilon_{33}^S, c_k^S, \varepsilon_{33})\) represent the piezoceramic material, and \((\varepsilon_{33})\) represents the matrix material.

Again, it is interesting to note the coupling present in the effective constants. The effective stiffness terms are the piezoceramic stiffness with an additional contribution which represents the electrical influence of the matrix material. This additional term is the difference between short-circuit and open circuit stiffness for the piezoceramic phase. For the cases when the dielectric mismatch is low \((\varepsilon_{33}^S = \varepsilon_{33})\), or there is very little matrix phase \((\nu_3^m \rightarrow 0)\), this additional term is small and the majority of the electric field reaches the piezoceramic. Thus, short-circuit conditions are nearly realized. For increasing amounts of matrix or larger dielectric mismatch, the piezoceramic phase is more electrically isolated, and the stiffness represents the open circuit values. The effective dielectric term is simply the series capacitor combination, which is also reflected in the effective piezoelectric terms. This factor

\[
F = \frac{\varepsilon_{33}}{\nu_3^p \varepsilon_{33} + \nu_3^m \varepsilon_{33}^S}
\]  

(3.19)

is the reduction in field reaching the piezoceramic phase.

The combination model is the combining of the three individual uniform fields cases \((A, B, C)\) to yield predictions of effective composite properties. There is more than one order in which these cases may be combined. Previous work (Bent, 1994) has shown that models have different regimes where they best represent actual response. Since uniform fields are assumed, combining more than two cases together will mean that compatibility or equilibrium will be necessarily violated at some interfaces. The best model will be the one that minimizes the violation of compatibility/equilibrium for that regime.

**Combination Model 1 - Uniform Packing (Circular Fibers)**

The order in which the cases are combined in combination model 1 is as follows:

\(A \rightarrow B \rightarrow C\)

Thus, the effective properties from Case A are substituted as the 'piezoceramic' properties of Case B, and the effective properties of Case B are substituted for the 'piezoceramic' properties for Case C, to yield the total effective properties. This combining order provides a combination model that is accurate when the actual composite geometry incorporates fibers that are on average uniformly packed. That is, on the average, there are equal amounts of matrix surrounding the fiber in both directions \((\nu_3^p = \nu_3^m)\). Uniform packing is usually the case for structural composites where the fiber diameter is substantially less than the ply thickness. This will eventually be the case for piezoelectric
Figure 3.4 Representations of composite geometry: (a) Multi-fiber layer composites modeled with uniform square packing (Combination Model 1); (b) Actual IDEPFC geometry modeled with rectangular packing, circular fibers (Combination Model 2); (c) Actual IDEPFC geometry modeled with rectangular packing, square fibers (Combination Model 3).

Fiber composites as fiber manufacturing technology improves. Such a combination forces the electric field to be equal in the fiber and the matrix surrounding it on all sides.

For uniform square packing, the maximum theoretical packing fraction is \( \pi/4 \), and is represented in Figure 3.4a. Since the model fiber is square, it is simply ensured that the area of square fiber in the model is the same as that of a circular fiber with the same total volume fraction, \( v_f \). This is represented by the following equation:

\[
v_f = v_1^p v_2^p = \frac{\pi}{4} X_1 X_2 \quad \left( v_1^p = v_2^p \right)
\]

where \( X_1 \) and \( X_2 \) are called the ceramic line fractions of the actual circular fiber composites. For example, an \( X_1 \) value of 0.5 represents, on average, circular fibers that are spaced apart by one fiber diameter in the \( 1 \) direction. Given the actual composite geometry, the model fiber fractions can be determined from the above.

The piezoceramic volume fraction \( v_3^p \) in Case C represents the portion of ceramic along the electric field path length. It is a function of the ‘2’ direction volume fraction and the geometry of the interdigitated electrode pattern. The fraction of ‘piezoceramic’ phase (which is actually the effective material from Case B) is the piezoceramic length divided by the total path length between the electrode surfaces:

\[
v_3^p = \frac{p/2}{p/2 + h/2v_2^m} = \frac{p/h}{p/h + (1 - v_2^p)} \quad (w \ll p)
\]

Thus, the Case C volume fraction depends on the ratio of the electrode spacing to wafer thickness. This term also serves to illustrate the potential improvement in performance offered by the interdigitated electrode pattern over the conventional electrodes.
For large values of \( p/h \), the volume fraction \( \nu_f \) quickly approaches one. Previous conventionally electroded PFC through-thickness volume fractions (which would be measured using \( \nu_f \)) of 0.9, now produce an effective ceramic volume fraction of 0.98 for the \( p/h \) ratio case of 6 studied here.

**Combination Model 2 - Rectangular Packing (Circular Fibers)**

The above description is appropriate for the averaged composite response of a multi-fiber layer composite. It is also more accurate when \( x_2 \) is not approaching one (i.e. when electrode and fibers are not approaching one another). However, representing the physical geometry of the experimental specimens requires looking at this particular regime in more detail. As \( x_2 \) approaches one, two issues become important.

The first issue is caused by the non-uniform packing of the composite specimens shown in Figure 3.4b. Since the fiber diameters are approximately as thick as the composite itself, \( x_1 \) and \( x_2 \) will not necessarily be equal. In fact, \( x_1 \) can reasonably be any value from 0.1 to 0.9, while \( x_2 \) is approximately 0.92 in the actual composites. As \( x_1 \) and \( x_2 \) become increasingly different, the combining order of Combination model 1 is no longer appropriate to model the composite response. Combination model 1 requires that all of the matrix material must have the same electric field distribution as the fiber it surrounds, regardless of the distance between fibers. However, as \( x_1 \) becomes smaller and the fibers more separated, the influence of the fibers on the matrix material separating them lessens. Thus, the field in the fibers is too heavily weighted by the distant matrix. Furthermore, as \( x_2 \) approaches 1, the matrix material in Case C becomes a very small localized region between the electrode and fiber, and heavily influences the fiber response. Choosing the correct combining order thus becomes a task in minimizing compatibility/equilibrium violations.

\[ C \rightarrow A \rightarrow B \]

This combination order causes the field in the fiber (and hence the composite response) to be more heavily influenced by the matrix between it and the electrode, rather than the matrix along the lengths of the fiber. Note that Cases A and B still accurately represent the mechanical properties, and that as \( x_1 \) and \( x_2 \) approach one another, combination models 1 and 2 give similar results.

The second issue is an artifact of modeling round fibers as square. As \( x_2 \rightarrow 1 \), the actual fibers approach the electrodes on the top and bottom surfaces. The sharp increase in

\[ \text{---} \]

\[ ^2 \text{The reader is reminded that Case C represents the matrix between the fiber and electrode and is a purely electrical combination.} \]
composite response in this limit is not captured by the Case C volume fraction as defined in Equation (3.21). In equation (3.21), \( v_2^p \) represents the average distance between the fiber and electrode, and is appropriate for uniformly packed fibers with diameters much less than the composite thickness. For the case when the individual fibers approach the electrode, it is necessary to replace \( v_2^p \) with \( x_2 \), the actual closest distance between the fibers and electrode. Thus,

\[
v_2^p = \frac{p/h}{p/h + (1 - X_2)} \quad (w \ll p)
\]

(3.22)

**Combination Model 3 - Rectangular Packing (Square Fibers)**

Combination Model 3 (Figure 3.4c) is also proposed for representation of actual composite geometry, but in the case where the fibers are square, not circular. These could be fibers that have been tapecast and diced (as used in Section 2.2.3), extruded, or cast into a mold. The advantage of square fibers is the increased level of ceramic. For this case, the ceramic line fractions (\( x_1, x_2 \)) are now equal to the fractions used in the combination model (\( v_1^p, v_2^p \)). The same arguments for Combination Model 2 hold here, and the combining order and Case C volume fraction definitions (Equation 3.22) are still appropriate.

### 3.3.3 Extension to Rate Dependent Material Behavior

The Uniform Fields model cannot accommodate nonlinear or nonconservative material properties without significant difficulty. More importantly however, the model can be easily adapted to account for rate dependent effects, such as damping from intrinsic material losses or piezoelectric shunting. It is incorporated into the above models (Combinations 1, 2, or 3) simply by substituting the material properties in the constitutive relations with their complex equivalents. Two examples are considered:

1. **Complex Electrical Properties:** The Uniform Fields can accommodate the three parameter electrical material model developed in section 2.3.2,

\[
\varepsilon^* = \varepsilon'(1 - iD) - \frac{i}{\omega \rho}
\]

(3.23)

where \( C'' \) is the capacitive loss from both DC resistivity and the basic dielectric lossiness, and is a function of frequency.

2. **Complex Mechanical Properties:** It is possible to generate an elastic equivalent network to represent the viscoelastic behavior of a material,

\[
E^* = E' - iE''(\omega)
\]

(3.24)
where \( E'' \) is the measure of basic mechanical lossiness, and is also a function of frequency.

In each case, these complex representations can be used for both the ceramic fiber or matrix. The difficulty is in characterizing the materials throughout the frequency range of interest, and as a function of each material variable that can be altered. This has been done for the electrical properties of the matrix material in section 2.3.2., but has not been carried out for the mechanical properties.

Of the two effects, the electrical lossiness is expected to contribute more significantly to material damping, especially by tailoring the property mismatch of the fiber and matrix. This is of interest in aerospace applications, where the reduction of structure borne noise and vibrations are of particular interest (see Chapter 1). For short circuit composite boundary conditions, the active material is passively shunted to a network of lossy material (the matrix) through several parallel and series paths, as modeled in the Uniform Fields approach.

The figure of merit is the mechanical loss factor, which is the ratio of imaginary and real parts of the composite stiffness (shown here in a one dimensional sense),

\[
\eta(\omega) = \tan \delta_m = \frac{\text{Im}(1/s_{33}^*)}{\text{Re}(1/s_{33}^*)} \tag{3.25}
\]

where \( \tan \delta_m \) is the mechanical loss angle, and \( s_{33}^* \) is the effective complex compliance in the fiber direction predicted by the Uniform Fields model. Many electrical and mechanical coupling effects contribute to this parameter, so that the solution is not easily expressed in closed form. The loss factor is a function of frequency, with a maximum at some \( \omega_0 \),

\[
\eta_{\text{max}} \rightarrow \frac{\partial (\tan \delta_m)}{\partial \omega} = 0 \tag{3.26}
\]

An example of the ability to tailor material properties for optimizing the material loss factor is shown in Figure 3.5. These predictions are made using the Uniform Fields model of this section (combination Model 2 for experimental geometry), augmented by the complex dielectric in Equation 3.23. The material property inputs are taken from the measured matrix properties of Chapter 2 for three levels of carbon black (0%, 5%, and 9% by weight) and no PZT filler. PZT 5H properties are assumed for the fiber, which has line fractions of 0.8 and 0.92 in the 1 and 2 directions, respectively (typical of experimental actuators).
Figure 3.5 Short circuit composite stiffness and material loss factor as a function of frequency for three levels of matrix carbon black loading. Predictions made with Uniform Fields model, using material properties in Table 2.3 and Figures 2.15-2.17.

The figure shows the real component of the stiffness and the material loss factor for short circuit electrical boundary conditions (leads joined), as a function of frequency. These curves are the mechanical analog of the relaxation phenomenon evident in dielectric polarization, where a peak in the dielectric loss occurs at the relaxation frequency for each polarization mechanism (see Buchanan, 1986). In this case, the stiffness increases sharply at the frequency where the real portion of the admittance (i.e. the apparent dielectric) drops, and the piezoceramic becomes more ‘electrically clamped’. As carbon black is added, the matrix dielectric is increased, and charges from the fibers can flow more freely, reducing the short circuit stiffness. At the point where the stiffness is changing maximally, the material loss factor peaks. Note that as the level of carbon black is changed, both the frequency of the peak and its magnitude are shifted. The reduction in the peak is due to a higher resistivity mismatch with additional carbon black. Maximizing the loss factor requires an impedance matched design.

As the figures show, a high level of carbon black is needed in order to introduce significant damping in the likely frequency range of interest. This may cause difficulties for practical operation - it may be impossible to pole the composites due to the high level of loss current needed to sustain the poling voltage. The next chapter will investigate these issues in the design tradeoffs involved in material selection.

3.3.4 A Comparison to Classical Theory

Modeling of fibrous composites is not an easy task. The level of anisotropy is at least transversely isotropic, even for composites with isotropic constituents. This results in five independent material properties which must be determined. For constituents of higher
orthotropy, this becomes even more difficult. Longitudinal effective properties were the first to be predicted, easily captured by simple combining rules for even moderately mismatched Poisson’s Ratios. Higher order models (elasticity methods, variational bounding) did not greatly improve on these predictions. Transverse properties such as the transverse stiffness (\(E_T\)) and shear modulus (\(G_T\)), on the other hand, are extremely difficult to predict accurately. The prediction of the transverse properties of a particular method became the basis on which the method was judged, and quickly became the focus of modeling efforts in the mid 1960’s.

This section shows a comparison between several of the most well known models and the Uniform Fields model, using the transverse stiffness as a metric. The most simple model for the transverse stiffness are the 1st order bounds derived from a one dimensional consideration of the two materials - the Mechanics of Materials approach. This method imposes absolute limits on the lower and upper values of the stiffness, based on the simple parallel and series combination of matrix and fiber. This method does not take into account composite microstructure. The Uniform Fields model is a higher order version of this approach, applying the combining rules to three dimensions. This captures the effects of induced stresses caused by Poisson’s Ratio mismatches, and matrix clamping of the fiber on all sides. This causes a violation of compatibility, but this is minimized for highly mismatched materials.

More accurate methods require consideration of the microstructure. The well known Composite Cylinders Assemblage (CCA) assumes a distribution of fiber sizes in order to fill out the composite, each with a corresponding ratio of matrix surrounding it, as shown in Figure 3.6a (Hashin and Rosen, 1964; Hashin, 1986). Hashin reports that this is the only analysis for which an exact solution is available. The solution is a combination of direct and variational approaches to determine the bounds on the transverse stiffness for the geometry assumed. Another well known solution is provided by Whitney and Riley, who used a Repeating Element approach (Whitney and Riley, 1966). This was similar to the composite element of Hashin and Rosen, but with less mathematical rigor. In this work, an Airy stress function was applied to the elasticity solution of a modified plane strain problem. A number of approximations were made in order to allow a closed form equation for the transverse stiffness.

The results for the four methods are shown in Figure 3.6b, which is a comparison of the transverse stiffness (normalized by the fiber stiffness) as a function of fiber fraction. The comparison is shown for PZT fibers in a matrix with 5% of the fiber stiffness, and unequal Poisson’s Ratios. For a more accurate comparison, the PZT is assumed to be
Figure 3.6 A comparison of models for the prediction of the transverse modulus, for a composite with a stiffness mismatch of 20 between fiber and matrix. Transverse modulus shown normalized by the fiber stiffness. Electrical coupling ignored for the Uniform Fields model.

isotropic. The Uniform Fields model is the full Combination Model #1, where the electrical coupling can be ignored by using an infinitely high dielectric so that the open and closed circuit PZT stiffnesses are identical. The 1st order bounds representing the Mechanics of Materials approach are extremely far apart. This is understood by recognizing the upper bound is the parallel combination usually used for approximating the longitudinal stiffness.

The higher order models tend to start near the lower bound, but digress as more fiber is included, and the stresses in the matrix and fiber are no longer equal. The upper and lower bounds of the Hashin and Rosen model are the closest possible bounds on the transverse stiffness, given the assumption on the model geometry. It is not expected that the Uniform Fields necessarily fall within these bounds. In fact, the Whitney and Riley prediction more closely compares to the Uniform Fields model. This is understandable, since the boundary conditions and geometry of the U.F. are more similar to Whitney and Riley than to the CCA. In any case, all three of these models provide very close predictions for the transverse stiffness. This is a significant finding for the Uniform Fields model, and indicates a degree of reliability for this approach. One assumption for all of these models, is the requirement that no fibers are intimately connected, i.e. the fibers are all separated by an equal amount of matrix. One would then expect that these models may underpredict the stiffness at high fiber fractions, where random contacts between fibers occur.
3.4 Finite Element Method

3.4.1 Introduction

The Finite Element Method (FEM) is a type of elasticity solution because it approximately satisfies the coupled electrical-mechanical governing equations. The problem is formulated in terms of displacement and voltage degrees of freedom, so that the compatibility and potential distribution equations are exactly satisfied. The energy formulation, however, only approximately satisfies the equilibrium and charge distribution field equations through a minimization of the total potential energy. The Finite Element, although an implicit approach to modeling effective material properties, is a convenient method for verifying the assumptions utilized in the simpler model (Uniform Fields approach).

The finite element approach was used successfully for the prediction of composite properties of the conventionally electroded PZT Fiber Composites (PFCs) in the previous work (Bent, 1994). This method demonstrated the ability to capture physical phenomenon that the Uniform Fields could not because it was limited to non-varying fields within each material. The first was a concentration of electric field, due to the high dielectric mismatch between the materials which drew the field lines into the fiber. The second was the appearance of a ‘dead zone’ of fiber near the outer edges that were furthest away from the electrode. Although the former effect was stronger than the second and increased the predicted response over the Uniform Fields slightly, these two effects tended to cancel out (pointing to the ability of the Uniform Fields to capture the response in an average sense). This effect is expected to be minimized in the present geometry, outside of the non-uniform behavior near the electrodes. A greater benefit of the finite element approach is its ability to model exact geometry, such as round fibers. This becomes important at high volume fractions.

3.4.2 Problem Definition and Solution

A commercially available FEM program (ANSYS) with coupled, multi-field elements was used to model the composite. The general approach to modeling followed the four important items discussed in the introduction of this chapter. Homogeneous material properties were input into the element libraries, with the PZT assumed uniformly poled in the 3 direction. Fully coupled field elements permit accurate linear modeling of the piezoelectric effect, unlike the use of thermal elements which are not sensitive to internal electric field variations caused by stresses. The full formulation of the electro-elastic finite element can be found in Allik and Hughes (Allik and Hughes, 1970).
The representative volume used in the model is shown in Figure 3.7. Like the Uniform Fields, the volume element is taken to be a representative (quarter) single fiber with surrounding matrix material in proportion to the volume fraction being modeled. In accordance with the discussion in previous sections, the amount of matrix surrounding the fiber is taken to be equal in the 1 and 2 axis directions for the purpose of model comparison (Combination Model 1 for the UFM). However, for the experimental comparison with manufactured composites, the actual geometry would necessitate a different thickness and width fraction.

The volume element comprises six faces which each have appropriate mechanical and electrical boundary conditions. The 1a, 2a, and 3a faces are the faces with positive (outward) direction normals in the 1, 2, and 3 directions, respectively. These faces are free to strain, restricted only by coupled displacements in that face’s respective direction. In the 3 direction, this represents the assumption that the plane perpendicular to the fiber must remain plane after deformation. This is true away from the ends of the fibers. Since the fibers are ‘long’ this is a good assumption. In the 1 and 2 directions, this represents the requirement that the repeating element must have uniform deformations that satisfy those of the composite macroscopic response.

The faces 1b, 2b, and 3b are the faces with negative axis orientation. These represent the planes of symmetry for the quarter model, and have symmetric electrical and mechanical conditions. The electrical symmetry is enforced through equating the normal electrical displacement to zero. This is done automatically for any interface for which no electric potential is specified. The mechanical symmetry is enforced by setting all normal mechanical displacements to zero. Although they are not modeled explicitly, the electrode surfaces shown as shaded areas in the Figure are generated by electrically coupling the nodes on face 3a together and the nodes on face 2a together (in the proper electrode region). The boundary conditions for all faces are summarized in Table 3.1. These are invariant with load case.

<table>
<thead>
<tr>
<th>Face</th>
<th>Mechanical B.C.s</th>
<th>Electrical B.C.s</th>
</tr>
</thead>
<tbody>
<tr>
<td>1a</td>
<td>All $u_1$ coupled</td>
<td>$D_1 = 0$</td>
</tr>
<tr>
<td>2a</td>
<td>All $u_2$ coupled</td>
<td>$V$ coupled (on electrode) $D_2 = 0$ elsewhere</td>
</tr>
<tr>
<td>3a</td>
<td>All $u_3$ coupled</td>
<td>$D_3 = 0$</td>
</tr>
<tr>
<td>1b</td>
<td>(Symm) $u_1 = 0$</td>
<td>$D_1 = 0$</td>
</tr>
<tr>
<td>2b</td>
<td>(Symm) $u_2 = 0$</td>
<td>$D_2 = 0$</td>
</tr>
<tr>
<td>3b</td>
<td>(Symm) $u_3 = 0$</td>
<td>$V$ coupled</td>
</tr>
</tbody>
</table>
Figure 3.7 Representative Volume Element for FEM model. Faces (1a-3b) defined for boundary conditions; voltage coupled DOF shown shaded. Not to scale.

Figure 3.8 Exploded view of RVE, illustrating tetrahedral coupled field elements. Shown for case with $X_1=X_2=0.8$. Fiber shown light gray, and electrode shown dark gray. Note that RVE is rotated from figures above and below (3.7 and 3.9).

Figure 3.9 Voltage potential lines for case of $X_1=X_2=0.8$, and dielectric mismatch of 10. Figure illustrates that electric field quickly becomes uniform after electrode.
### Table 3.2 FEM load cases to determine effective material properties.

<table>
<thead>
<tr>
<th>Case</th>
<th>Mechanical Loading</th>
<th>Electrical Loading</th>
<th>Properties Obtained</th>
</tr>
</thead>
<tbody>
<tr>
<td>1</td>
<td>$u_i = 0$ on faces 1a,2a,3a (zero strain)</td>
<td>Apply voltage $V$ on top electrode</td>
<td>$e_{33}^{\text{eff}}$ $e_{31}^{\text{eff}}$ $e_{32}^{\text{eff}}$ $e_{33}^{\text{eff}}$</td>
</tr>
<tr>
<td>2</td>
<td>Apply displacement $u_1$ on face 1a</td>
<td>$V = 0$ on electrodes (short circuit)</td>
<td>$c_{11}^{\text{eff}}$ $c_{12}^{\text{eff}}$ $c_{13}^{\text{eff}}$ $e_{31}^{\text{eff}}$</td>
</tr>
<tr>
<td>3</td>
<td>Apply displacement $u_2$ on face 2a</td>
<td>$V = 0$ on electrodes (short circuit)</td>
<td>$c_{22}^{\text{eff}}$ $c_{12}^{\text{eff}}$ $c_{23}^{\text{eff}}$ $e_{32}^{\text{eff}}$</td>
</tr>
<tr>
<td>4</td>
<td>Apply displacement $u_3$ on face 3a</td>
<td>$V = 0$ on electrodes (short circuit)</td>
<td>$c_{33}^{\text{eff}}$ $c_{13}^{\text{eff}}$ $c_{23}^{\text{eff}}$ $e_{33}^{\text{eff}}$</td>
</tr>
</tbody>
</table>

The composite properties are found by posing a series of hypothetical experiments. In the finite element method, a series of separate load cases are applied to the RVE, and are summarized in Table 3.2. The first load case (#1) applies an electric field to the element, while all faces (1a-3a) are clamped. This is accomplished by setting the potential on the top electrode to some arbitrary voltage $V$, while the other electrode (face 3a) is held at ground. Given this set of loading conditions, the solution to the problem returns values of force and surface charge on the electrode. From a summation of these nodal forces and charges, it is possible to calculate the effective dielectric and clamped piezoelectric constants. Similar tests produce the stiffness constants by applying displacements to the various faces. In these load cases, the electrical boundary conditions are short-circuit.

Figure 3.8 illustrates the actual model RVE in the finite element program, shown as an exploded view. Tetrahedral elements were used to allow automatic mesh generation for the complex geometry. These elements had four nodes, each having three displacement and one voltage degree of freedom. The number of elements required depended on the ‘severity’ of the geometry, and ranged from 500 for lower volume fractions, to 2400 for high volume fractions. 580 elements were used in the case shown in the figures. Figure 3.9 shows the voltage potential lines resulting from the solution of a problem where $X_1=X_2=0.8$, and the dielectric mismatch between fiber and matrix was 10. This mismatch was chosen in order to illustrate the uniformity of the electric field. Distortions of the potential occurred only near the electrode for the electrode geometry considered (p/h=6, w/h=1), illustrating the applicability of the uniform field assumption in the previous model. The results for all cases are shown in the next section in graphical form.
3.5 Model Comparison

3.5.1 Low Field Composite Properties

Figure 3.10 shows the comparison of the two model approaches. Combination model 1 is used for comparison with the FEM model, and assumes that \( v_f^p = v_f^p \). This is usually the basis upon which micro-mechanical models are compared. All plots have been nondimensionalized by the respective values of the monolithic ceramic, so that a value of one represents pure ceramic. The effective properties are shown as a function of total fiber volume fraction. Since a square packing arrangement is assumed, the maximum theoretical volume fraction that can be achieved is \( \pi/4 \). Since the Uniform Fields model has square fibers, it is simply ensured that the area of fiber is the same as that of a circular fiber with the given volume fraction.

A family of three curves is shown in each plot, which represent different possible material combinations. For the effective piezoelectric properties, the material dielectric ratios \( (e_{33}^p/e_{33}) \) are varied for the values of 500, 100, and 10 (shown as curves 1, 2, 3), to illustrate the effect of dielectric mismatch on actuation capability. For these curves, the stiffness is held constant \( (e_{11}^E/c_{11} = 20) \). Some small change of the effective piezoelectric constants was seen with stiffness, but only as the matrix becomes very stiff and starts to clamp the fibers. For the effective stiffness constants, the dielectric is held constant \( (e_{33}^p/e_{33} = 500) \) and stiffness mismatch ratios \( (e_{11}^E/c_{11}) \) of 20, 5, 2 are shown for curves 1, 2, 3. A small change of these effective constants was seen with dielectric, due to the difference in short circuit conditions for higher dielectric matrix.

The material \( e_{ij}^{p} \) and \( c_{ij}^{p} \) constants are found directly from the models, while the \( d_{ij}^{p} \) and \( s_{ij}^{p} \) constants are easily found from the constitutive relationships (described in Appendix A). Excellent agreement is seen between both models in all of the effective material constants. This indicates that the geometry and field assumptions of the simpler technique are adequate for predicting the response of the quarter model, within the assumptions prescribed on the model. Naturally, a portion of this good agreement is due to the common assumptions of uniform polarization and common boundary conditions in both models. However, this agreement indicates that the assumption of field uniformity is a good one when considering the averaged properties. It also indicates that round fibers are adequately modeled as square, again by using the effect of averaging. Whether the actual composite response is well represented by these assumptions will be a true test of the models’ validity.
Stiffness constants for $\varepsilon_{33}^T/\varepsilon_{33} = 500$ with ratios $c_{11}^F/c_{11}$ of 20, 5, 2 for curves 1, 2, 3.

Free strain constants for $c_{11}^F/c_{11} = 20$ and ratios $\varepsilon_{33}^T/\varepsilon_{33}$ of 500, 100, 10 for curves 1, 2, 3.

Clamped constants for $c_{11}^F/c_{11} = 20$ and ratios $\varepsilon_{33}^T/\varepsilon_{33}$ of 500, 100, 10 for curves 1, 2, 3.

Figure 3.10 Effective composite material properties compared for two models, where composite ratios are $w/h = 1$ and $p/h = 6$.
It is interesting to note the composite orthotropy. Both the piezoelectric free strain constants \((d_{33}^{\text{eff}}, d_{31}^{\text{eff}})\) have approximately the same magnitude when normalized by the monolithic ceramic values. Thus, the piezoelectric free strain constants have the same ratio \((d_{33}:d_{31})\) in the IDEPFC composite as they do in monolithic ceramic. However, the \textit{induced stress} piezoelectric constants \((e_{33}^{\text{eff}}, e_{31}^{\text{eff}})\) are largely different, reflecting the softer material in the 1 direction, due to the soft matrix separating the fibers.

### 3.5.2 Improved Actuation Capability

Figure 3.11 shows the increase in actuator performance over the conventional piezoelectric fiber composite (from Bent, 1994). The improvement is shown in two ways: an increase in piezoelectric induced stress capability, and an increase in piezoelectric anisotropy. Both plots are for a single material system, which corresponds to the middle of the family of the three curves in the previous figures (Figure 3.10). This corresponds to \(c_{11}^E/c_{11} = 20\) and \(e_{33}^T/e_{33} = 100\), ratios which are close to the actual material system. Like the previous comparison between the two models, this one assumes square packing.

The first plot (Figure 3.11a) shows the increase in the piezoelectric induced stress constants \(e_{\text{long}}\) and \(e_{\text{trans}}\), by plotting the capability ratio of those constants (IDEPFC:PFC). The nomenclature refers to the \textit{longitudinal} and \textit{transverse} planar directions, where longitudinal is along the fibers. The convention of ‘33’ and ‘31’ is not appropriate due to the different poling directions of the two composites. Note that the absolute value is used, as \(e_{\text{long}}\) for the two actuators is actually opposite in sign. A substantial improvement using

![Figure 3.11](image)

**Figure 3.11** Comparison of IDEPFC actuator to conventionally electroded PFC: (a) IDEPFC/PFC ratio of stress constants \(e_{\text{long}}\) and \(e_{\text{trans}}\), (b) Comparison of maximum induced shear stress constant, \(e_{36}^{\text{eff}}\).
the interdigital electrodes is apparent, especially in $e_{\text{long}}$. In the actual actuator geometry which uses a mono-fiber layer, the ratios will be most closely approximated by the values at maximum volume fraction (\(\pi/4\)).

The second figure, Figure 3.11b, shows the increase in piezoelectric anisotropy, illustrated through the induced shear stress parameter, $e_{36}$. When used in laminated structures, it is possible to orient the fibers at angles other than along the laminate axes. This will induce a shear stress in the structure which may be used to actuate in twist. This parameter is maximized at a 45° orientation from the ply axis$^3$:

$$e_{36} = (e_{\text{long}} - e_{\text{trans}}) \cos \theta \sin \theta$$  \hspace{1cm} (3.27)

A large increase in twist capability is available with the new IDEPFC actuator, perhaps as much as 25 times greater than the conventional PFC actuator. This increase is a result of the improved actuation capability (from the electrodes), and the two piezoelectric terms which are now opposite in sign, which maximizes the induced shear. Again, comparison values for the experimental composites may be taken as the values at maximum packing fraction.

3.6 Summary

In this chapter, two models were developed to predict the effective composite properties of the interdigitated electrode piezoelectric fiber composite. The first of these, the Uniform Fields model, used a closed form approach based on simple combining rules that allowed insight to the mechanisms that govern the composite response. The second, a Finite Element model, validated the simpler approach to modeling including assumptions on field uniformity away from the electrode region. These models can now be used for the design of active composite actuators, presented in the next chapter, and for comparison with experimental results in Chapter 6.

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$^3$ See (Bent et al., 1995) for a discussion of twist actuation
Chapter 4

Design of Active Fiber Composites

4.1 Introduction

Chapter 2 characterized constituent materials for use in the design and manufacture of active fiber composites. These materials represented a wide range of ceramic types and matrix compositions which can act as realistic material inputs to models. The next chapter (Chapter 3) developed the ability to predict the effective composite properties using linear modeling techniques. This chapter combines these efforts in order to advance toward the main goal of this thesis - the combination of active and passive materials for the development of a composite actuator which meets the performance requirements of aerospace applications. This is accomplished by examining the change in composite properties with different material combinations in order to map a design space for composite selection. Optimizing the choice of composite from this design space depends on the particular application, and is based on metrics of performance and reliability.

The overall purpose of this chapter is the illustration of the design choices and issues that were encountered in this work. Specifically, the objectives are

- to explore the issues and tradeoffs in the design of active fiber composites for structural actuation, and
- to determine a recommended set of material combinations to be used in manufacturing experimental samples.

The decisions in this chapter represent a single design iteration through the decision making process. True closed loop design would entail returning to the design tradeoffs after experimental work to adapt models and improve the design. Also, as new materials become available, more optimal solutions to the active fiber composite problem will be possible. However, the important issues will have been identified and the general approach will remain in place for future designs.

The chapter is divided into two sections: fiber design selection, and matrix design selection. These two material choices are obviously not independent, but the design steps
can be best illustrated by considering the effects separately. From the information given, it is simple to scale the effects for different fiber and matrix combinations. The fiber selection investigates tradeoffs with the different ceramics and the effects of bulk properties when combined with a low dielectric matrix. This section also examines the potential advantages and disadvantages of square fibers, versus the conventional round geometry. The matrix section investigates the tradeoffs of conductive and dielectric fillers on the processing, performance, and operational risk of composite design. The chapter is summarized by suggesting a series of composites for manufacture and experimental verification.

4.2 Design Issues

The following two subsections describe the various issues of fiber and matrix properties on the overall composite response. Predictions for the design studies are made with the Uniform Fields model, using combination models #2 and #3 for the round and square fibers, respectively. These are best suited for the geometry of the composites considered. In keeping with the planar actuation focus, plane stress conditions are assumed in all property predictions.

4.2.1 Fiber Design Issues

There are a number of issues in composite design related to fibers. Several of these involve choices of the fiber geometry, and the amount and shape of the ceramic. Another issue is the selection of different PZT ceramic fiber materials. Bulk properties are assumed for all ceramic materials in this section, to represent properties that may eventually be available (even though the current PZT 5H fibers have been shown to have properties less than bulk). The matrix material composition is held constant throughout this subsection, and is assumed to be an intermediate PZT filler fraction of 60% (no carbon).

Ceramic Content

An obvious choice in the design of composite actuators is the ceramic content. One basic objective is to maximize actuator authority, captured by the piezoelectric induced stress constants \((e_{33}, e_{31})\). Maximizing the induced stress requires the highest possible line fractions, \(X_1\) and \(X_2\). A second objective is to maximize the anisotropy (or, directionality) of the actuator, captured by the difference in the two induced stress constants. This is desired for optimizing induced shear stress for twist applications, and other anisotropic actuation designs. Fortunately, maximizing the disparity between the two planar actuation constants also requires the highest possible \(X_1\). This is true for the IDEPFC configuration, because \(e_{33}\) and \(e_{31}\) are opposite in sign. This was not the case in the conventionally
electroded composite, where both planar constants operated in phase, so that maximizing the directional nature of the PFC required a soft transverse direction (i.e. minimizing the $e_{31}$). Figure 3.8 showed the advantage of high directionality by comparing the two composite types using the piezoelectric induced shear stress constant ($e_{46}$) as a metric.

It is also desired that the thickness fraction ($X_2$) be as high as possible, which is controlled by the compaction of the composite during cure. Small differences in thickness greatly affect performance. On the other hand, practical limits on $X_1$ are set by manufacturing and strength related issues. If the ceramic content is too high in the width direction, there may be difficulty with matrix wetting of the fibers, and excessive fiber damage and cross-overs. Poor wetting might result in increased void content. A reduction in strength may also accompany high ceramic fractions from inadequate matrix coverage. Matrix coverage of the fibers is necessary for shear transfer around damage in the brittle fibers. The value of 0.8 is chosen for the nominal $X_1$ fraction.

**Composite Thickness**

Another choice is the thickness of the composite ply, with respect to the fiber diameter. Practical limitations on available voltage between 2 kV to 4 kV set the electrode finger spacing on the order of 1 mm in order to maintain significant fields in the composite. Since the performance is dependent on maximizing the uniformity of the field along the fiber, it is necessary to have composites 20% or less as thick as the electrode spacing (p/h ratio discussed in section 1.3.2). This limits the composite to thicknesses of 200 microns or less. Available fibers are in the range of 100-130 microns, restricting manufacture to composites that are only one fiber diameter thick until piezoceramic fibers in the range of 5-10 microns become available. Smaller fibers would permit hexagonal packing (if circular), which would increase the maximum ceramic fraction. However, it is unclear whether smaller fibers will actually decrease the performance due to the increased number of low dielectric matrix gaps between the ceramic.

**Fiber Cross Section**

With the availability of both square and round fibers, it is interesting to consider the differences that fiber cross section makes in performance. Figure 4.1 shows the predicted piezoelectric induced stress constant in the fiber direction, as a function of the ceramic width fraction ($X_w$). It is assumed that the diameter of the round fibers is equal to the thickness of the square fibers. The results are shown for two values of ceramic thickness fraction, 0.90 and 0.96, which are typical of the range expected for the composites. These predictions are based on PZT 5H fibers in a 60% PZT doped matrix.
Figure 4.1 Comparison of predicted induced stress constants for square and round fibers of the same cross section dimension. Predictions with Uniform Fields model.

The results are very straightforward. The change in $e_{33}$ is strictly linear with increases in the ceramic content ($X_1$) for a given thickness fraction ($X_2$). The difference in performance between the square and round cross sections is $\pi/4$ - the difference in cross sectional area. This is the only difference predicted, as both the square and round fibers are assumed to have the same $X_2$, i.e. both approach the same distance to the electrode. It is this thickness fraction that predominantly controls the piezoelectric response (see equation 3.22). Naturally, a higher composite $X_2$ value increases the sensitivity, as shown in the figure, but this $\pi/4$ ratio remains constant. If the performance was normalized by the mass of the composite (not shown), this would reduce the apparent advantage of the square fibers. Square fibers would still show a slight advantage due to the more efficient use of the matrix material, which contributes significant mass when doped with PZT filler. i.e. less non-active matrix material is required for the same amount of fiber.

The choice of square versus round fiber then depends on application needs. For a design with restrictions on volume, square fibers would be desired; for mass efficiency, the difference between square and round is slight. Additional advantages of square fibers include more easily controlled properties, including performance and possibly strength. However, some disadvantages may be inherent in using square fibers. Difficulty with processing and sufficient matrix coverage are two such issues which may only be determined with experimental work.

Fiber Materials

A number of PZT ceramic types exist, which exhibit a range of properties and offering different potential benefits to the active composite. Section 2.2.1 characterized
four of these PZT types that represent the spectrum of possible ceramics from very soft to very hard (5H, 5A, 4S, and 8M in increasing hardness). The soft ceramics exhibited the highest electromechanical coupling, while the hard ceramics allow operation at high compressive stress. Although not all of these ceramics are currently available in fiber form, this design study considers all of the above, to examine the range of materials that may eventually be available.

Using the Uniform Fields model, the properties of composites with each of these ceramics was predicted. All other variables were held constant (fibers assumed round, ceramic fractions of $X_2 = 0.92$ and $X_1 = 0.8$, and 60% PZT doped matrix). Full bulk properties were assumed for all ceramics. The results are summarized in Table 4.1.

The most pronounced effect of compositing the different ceramics is the effect on composite actuation capability, exhibited in the piezoelectric free strain constants ($d_{33}$). The harder ceramics have comparable composite values due to their lower dielectric. This lowers the dielectric mismatch and compensates for their generally lower bulk electromechanical coupling. The harder ceramics also have higher bulk stiffness, resulting in high clamped (or, induced) stress constants ($e_{33}$). Interestingly, the higher dielectric mismatch in the softer ceramics cause composite stiffness that is closer to the open circuit values. This, in effect, compensates for the dielectric mismatch, and results in a high $e_{33}$ for the soft ceramics as well. Note that for the degraded PZT 5H properties, the modulus and induced stress properties are greatly reduced, approximately 38 GPa and 4.1 N/C, respectively.

Generally, PZT 5H and 5A are the most often used in structural actuation. However, PZT 4S appears to have at least 15% higher actuation authority in composite form. Another comparison can be made based on the depolarization stress. This is the value of compressive stress at which 25% of the original actuation capability has been

<table>
<thead>
<tr>
<th>Plane Stress Properties</th>
<th>PZT 5H</th>
<th>PZT 5A</th>
<th>PZT 4S</th>
<th>PZT 8M</th>
</tr>
</thead>
<tbody>
<tr>
<td>Dielectric, $e_{33}^T$</td>
<td>455</td>
<td>370</td>
<td>352</td>
<td>264</td>
</tr>
<tr>
<td>Free Strain, $d_{33}$ (pm/V)</td>
<td>132</td>
<td>135</td>
<td>141</td>
<td>116</td>
</tr>
<tr>
<td>Clamped Stress, $e_{33}$ (N/C)</td>
<td>6.7</td>
<td>6.2</td>
<td>7.7</td>
<td>6.8</td>
</tr>
<tr>
<td>Modulus, $1/s_{33}^E$ (GPa)</td>
<td>51.5</td>
<td>46.6</td>
<td>55.5</td>
<td>58.8</td>
</tr>
<tr>
<td>Depole Stress, $T_3$ (MPa)</td>
<td>25</td>
<td>42</td>
<td>&gt;87</td>
<td>&gt;87</td>
</tr>
</tbody>
</table>

Table 4.1 Predicted composite properties using Uniform Fields model, for different piezoceramic fibers, and shown for plane stress conditions along fiber direction. Bulk properties assumed for all ceramic.
removed due to dipole realignment. PZT 4S and 8M have substantially higher stress capability than either of the two soft ceramics. From these linear considerations, it appears as though the harder ceramics are the best choice for active fiber composites.

The linear property predictions are important for understanding the material tradeoffs. However as section 2.2.1 showed, the PZT ceramics had very different high and low field response. High field response was characterized using the Representative Work Cycle, which was a measure of free strain at high electric fields. The size of the Rep Cycle is nominally dependent on the same linear material property tradeoffs, but at high fields, two additional factors become important:

- Degree of nonlinearity of bulk ceramic, i.e. how the response changes with the size of the applied field,
- and, the level of the saturation strain.

PZT 4S is a much harder piezoceramic, which reduces the nonlinearity in the strain response, particularly for field levels at which the Rep Cycle is measured. As the section on ceramic characterization showed (Section 2.2.1), PZT 4S has a much lower saturation strain than either PZT 5H or 5A. Both of these would likely contribute to a lowered Rep Cycle strain.

For design of active composites, this concept can be applied in the form of a scaled Representative Work Cycle. This metric is an attempt to extend the linear Uniform Fields model to prediction of the expected composite response at high electric fields. This is based on using the Uniform Fields model in conjunction with high field characterization of the different ceramics.

The concept is based on the premise that the actuation strain of the composite is the same as the strain of the bulk ceramic in the composite (a logical assumption), which leads to the following relation

$$S_{\text{bulk}} = S_{\text{comp}} \rightarrow d_{33}^{\text{bulk}} E_{\text{bulk}} = d_{33}^{\text{comp}} E_{\text{comp}}$$  \hspace{1cm} (4.1)

This can be rewritten as,

$$E_{\text{bulk}} = (d_{33}^{\text{comp}} / d_{33}^{\text{bulk}}) E_{\text{comp}}$$  \hspace{1cm} (4.2)

The ratio $d_{33}^{\text{comp}} / d_{33}^{\text{bulk}}$ is the ratio of composite to bulk ceramic free strain constants, which is an indicator of strain sensitivity reduction in the composite form. It combines the reduced field to the fiber and any mechanical clamping or Poisson's effects in the composite. The sensitivity ratio is easily found from the Uniform Fields results in Table 4.1. Values of this ratio for the four ceramics are shown in the first row of Table 4.2, and
Table 4.2 Prediction of relative high field actuation capability for different piezoceramic fibers, shown for plane stress conditions along fiber direction. Bulk properties assumed for all ceramic, and predictions found using Uniform Fields model.

<table>
<thead>
<tr>
<th>Plane Stress Properties</th>
<th>PZT 5H</th>
<th>PZT 5A</th>
<th>PZT 4S</th>
<th>PZT 8M</th>
</tr>
</thead>
<tbody>
<tr>
<td>Sensitivity Ratio, $d_{33}^{comp}/d_{33}^{bulk}$</td>
<td>0.21</td>
<td>0.35</td>
<td>0.38</td>
<td>0.52</td>
</tr>
<tr>
<td>Bulk PZT Cycle, $E_{bulk}^{bulk}$ (kV/cm)</td>
<td>0 to 3.8</td>
<td>0 to 6.3</td>
<td>0 to 6.8</td>
<td>0 to 9.4</td>
</tr>
<tr>
<td>Scaled Rep Cycle (ppm)</td>
<td>275</td>
<td>350</td>
<td>210</td>
<td>160</td>
</tr>
</tbody>
</table>

range from 0.21 for the high dielectric mismatch of PZT 5H to 0.52 for PZT 8M. It is assumed that the high field composite actuation is an extension of this low field sensitivity\(^1\).

To predict the strain response of the composite at high field, a high field Rep Cycle ($E_{comp}^{comp}$) was chosen for the composite (0 to 18 kV/cm was chosen arbitrarily), and the sensitivity ratio was used to determine the field that the bulk ceramic in the composite would experience ($E_{bulk}^{bulk}$), given by equation 4.2. Each of the four bulk ceramics was then characterized at this reduced Rep Cycle field level. The resulting bulk strain is a prediction of the composite strain at the high field. As the final row in the table shows, the high field response of the harder ceramics is significantly less than for the softer ceramics. This is largely due to the lower bulk Rep Cycle strains, measured in Chapter 2. Considering the high field behavior of the composites, the softer ceramics appear to be the better choice for actuation capability in these regimes.

4.2.2 Matrix Design Issues

This section investigates different composite designs based on a range of different fillers, both conductive and dielectric. Experimental data from Chapter 2 provides inputs to the Uniform Fields model, which is used once again to determine the predicted composite response. Design issues such as tailored frequency response, processing, and performance are discussed as functions of the fillers.

In all of the designs shown in this section, only the matrix composition is varied. All other materials are held constant. The fibers are assumed to be round, with the degraded PZT 5H properties, which will actually be used in the experimental work. Unless otherwise noted, the ceramic fractions are assumed to be the typical values of $X_1=0.8$, and $X_2=0.92$.

\(^1\) This assumption only requires that the electrical and mechanical properties of the materials do not change substantially with the size of the applied field.
Frequency Response

One of the most interesting effects of fillers on composite properties is the change in frequency response. Figure 4.2 shows the predicted response for a series of composites using the field in the fiber as a metric. This ratio is key to determining both the poling and operational capabilities of the composites. The predominant term in the Uniform Fields model that governs this cutdown ratio was given in Equation 3.19. If the dielectric in this equation is replaced with the complex dielectric from Equation 3.23, this factor describes the cutdown as a function of frequency. Taking the frequency limits gives the AC and DC asymptotes (ignoring dissipation, D),

\[
AC \quad \omega \to \infty \quad F \to \frac{\varepsilon_{33}^m}{\varepsilon_{33}^m \nu_3^p + \varepsilon_{33}^p \nu_3^m}
\]

\[
DC \quad \omega \to 0 \quad F \to \frac{1/\rho^m}{\nu_3^p/\rho^m + \nu_3^m/\rho^p}
\]

(4.3)

where \( \nu_3^p \) is the Case C volume fraction (it has a value of 0.986 for an \( X_2 \) of 0.92). As expected, the DC response is dependent on the relative resistivities, while the AC response is a function of the dielectric mismatch. These asymptotes approximately correspond to the field in the fiber determined by the full Uniform Fields model, as shown in the figure.

A number of important conclusions can be drawn from these results. The first is with regard to the DC component of the composite response. The DC resistivity mismatch between the fiber and matrix is much less than the dielectric mismatch. In fact, with added carbon black, the mismatch works in favor of the fiber, allowing full field in the ceramic.

**Figure 4.2** Uniform Fields prediction of frequency response for composites with varying amounts of fillers, based on measured properties for matrix samples (Chapter 2). (a) As a function of carbon black weight % (no PZT); (b) As a function of PZT weight % (no carbon).
For all cases considered, the field in the fiber is higher at DC than it is at AC. This is fortunate, as it guarantees full poling. Had the ceramic/matrix mismatch been the opposite, effective poling would require an AC field. Since soft ceramics such as PZT 5H require extended time to reach full polarization, this would have reduced the capabilities.

The transition from DC to AC response occurs primarily below the frequency range of interest (1 Hz - 10 kHz), except for the composites with very high percentages of carbon black filler in the matrix (i.e. 9%). Thus, the operational performance is dominated by the dielectric mismatch alone. Even in the case of 9% carbon black, conservative design would require assuming the lower field in the fiber. In order to get a resistively dominated design, it would require levels of carbon black far above the percolation threshold, resulting in almost assured dielectric breakdown.

The material models used for the matrix and fiber contained three components: the resistivity, the dielectric, and the dielectric loss (or, dissipation, D). Before the corner frequency, the dissipation is swamped by the resistivities of the materials. After the corner frequency, the dissipation becomes the only resistive component of the material. As section 2.3.3 described, the dissipation is relatively constant with frequency. This dissipation acts to increase the field in the fiber, in the same way that the resistive mismatch did before the corner frequency. However, this increase is slight except for very large dissipation, such as those in the 9% carbon black matrix (D=0.5). Even in these cases, the increase of field in the fiber is on the order of 15%, so that the effect of dissipation can generally be ignored.

**Processing and Viscosity**

Section 2.3.4 demonstrated the significant increase in viscosity of the uncured matrix as a function of the PZT filler percentages\(^2\). Higher matrix viscosity can have adverse effects on the ease of processing, causing higher void content and increased difficulty with wetting the fibers and avoiding damage. However, by processing at higher temperatures, the viscosity was shown to greatly decrease, thereby minimizing the effects of the filler.

A maximum temperature for processing was determined to be 50 °C. Beyond 50°C, additional bubbles created from evaporation of solvents were introduced into the matrix, and the hot components were difficult to work with. Figure 2.9b shows the viscosity of matrix with 60%, 75%, and 80% PZT fillers as a function of temperature. An acceptable viscosity for processing was qualitatively determined, by working with fiber and matrix

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\(^2\) Carbon black filler had only a small affect on the viscosity, due to the small amounts used. 5% by weight is only 3% by volume.
materials at room temperature. The undoped matrix (0% PZT) was sufficiently inviscid to allow manufacture of composites, while the 60% PZT was slightly too viscous for easy application to fibers. From Figure 2.9b, this placed the acceptable viscosity at approximately 3000 cps (log viscosity of ~3.5). For processing at 50°C, this sets the maximum PZT filler to between 75-78 weight percent.

**Performance and Composite Thickness**

The addition of fillers to the matrix obviously increases the matrix dielectric, resulting in a lower mismatch with the fibers, and improving the expected performance. However, the addition of fillers may also decrease the performance, depending on the effect of fillers on composite compaction. The increased viscosity of the PZT filled matrix, and the finite size of the filler particles may actually act to offset the benefits of higher dielectric.

Figure 4.3 shows the predicted induced stress constant ($e_{33}$) as a function of thickness line fraction ($X_2$), and PZT filler percentages. Again, the effect of carbon black is not included because it does not affect the viscosity significantly, and the particles are orders of magnitude smaller than the PZT.

The figure shows that the performance improves proportionally to the increase in matrix dielectric, and increases nonlinearly with the thickness fraction. For the range of $X_2$ from 0.90 to 0.96, which represents a change of only 4 microns above and below the fiber, the composite performance increases significantly. It is possible that the increased viscosity of the doped matrix might cause a reduction in compaction of this size. Even

![Figure 4.3 Uniform Fields model prediction of composite clamped stress piezoelectric constant, illustrating the variation in magnitude with composite thickness line fraction and PZT filler.](image)

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without increased viscosity, the PZT filler size is on the order of the change in gap size. Given that the mean PZT particle size is 1 micron, with a distribution that likely contains larger particles, it is very possible that the composite thickness may increase by 4 microns on either side. In such cases, the undoped matrix composites would have better performance than their doped counterparts.

Smaller particle sizes may not avoid such a problem. Smaller particles have a higher surface to volume ratio, which would result in higher matrix viscosity and potentially lower effectiveness caused by increased surface resistance (Ruschau et al., 1990). Smaller particles are also more difficult to disperse effectively, which might result in particle aggregates that are as large as the filler size in the current system.

One potential benefit of the PZT fillers, however, is the decreased sensitivity to the compaction on performance. i.e. the compaction becomes less important for materials with a lower dielectric mismatch. This is also illustrated in Figure 4.3, by the greater relative reduction in performance of the undoped matrix composites as $X_2$ is decreased.

**Field Distributions**

Optimal selection of the matrix and fiber combination is a balance between performance and reliability (i.e. operational risk). Electric fields in the fiber and matrix materials define this balance. As previously discussed, performance can be represented by the induced stress capability of the composite. The induced stress is a direct function of the proportion of applied electric field reaching the fiber. Reliability is related to dielectric breakdown, which is controlled by several factors. Some factors, such as voids in the matrix or porosity in the fibers, are not easily accounted for in design. Field concentrations due to geometry and material property mismatches, however, are possible to predict. The weak point in the composite is the matrix material located between the fiber and electrode. Field concentrations in the matrix there can be as high as 100 times the applied field to the composite.

The Uniform Fields model was used to predict the field in the fiber and matrix, using the experimental data of Chapter 2. Continuous functions were fit to the data, in order to obtain continuous descriptions of matrix properties for varying PZT and carbon black. This is shown in Figure 4.4. Since there was such large variation for the effect of carbon black on dielectric constant (Figure 4.4a), it was assumed that the combined behavior of carbon black and PZT was multiplicative: the dielectric constant as a function of pure carbon black fit by a quadratic least squares approximation; the curves for the series with PZT loading were taken as the first curve, multiplied by the ratio of dielectric at zero carbon black. Although this is not necessarily accurate, the somewhat random effect of
carbon black makes it difficult to pinpoint the 'best' values. For the sake of illustrating the design process, the multiplicative fit will suffice. For the 60% PZT series, this falls along the high end in the range of measured dielectrics. For the 75% PZT series, this is a conservative estimate. Figure 4.4b shows the curve fits for the dielectric strength of the matrix as a function of filler loading. In this case, the assumed fits were both quadratic and splines that appeared to 'sensibly' fit the data.

Figure 4.5 demonstrates the effect of these field distributions as a function of both carbon black and PZT fillers: Figure 4.5a shows the field in the fiber, and Figure 4.5b shows the value of maximum field in the matrix. The solid lines represent the range of manufacturable matrix compositions with the given materials, while the dotted lines are extrapolations of the current data. For this comparison, it was assumed that the composite thickness remained constant with added fillers, with $X_1=0.8$ and $X_2=0.92$.

The field in the fiber increases with both PZT and carbon black, while decreasing the field concentration in the matrix considerably. Pure epoxy matrix (no fillers) shows the largest field concentration, approximately 86 times the applied field for the case considered. For an 18 kV/cm field applied to the composite (2 kV), the matrix will experience a 1550 kV/cm field. Although this appears excessively high (compared to the measured breakdown fields shown in Figure 2.19), the matrix has extremely high dielectric strength, especially at a thickness of 6 microns (for $X_2=0.92$). Interestingly, increasing compaction (higher $X_2$ values) increases the field in the fiber and the concentration in the matrix by equal amounts.
Figure 4.5 Uniform Fields prediction of field in fiber and matrix, as a function of carbon black and PZT fillers. Shown as a fraction of field applied to composite. (a) Field throughout fiber, (b) Field in matrix between fiber and electrode, illustrating concentration of field due to small gap and dielectric mismatch. Designs of equivalent field ratios shown as open circles 'o'.

Alternate designs for the same performance/reliability can be seen by taking horizontal lines across the figures. For example, either 75% PZT doping or 5% carbon black filled matrix will result in a 37% fraction of field in the fiber, and a 65 times concentration in the matrix. These alternate designs are shown as ‘o’ in the figure. Although these curves illustrate the effects of the fillers, design with fillers will involve relating these results to the two metrics discussed above - performance and reliability. For the performance, this will involve examining the composite induced stress as a function of the fillers. For the reliability, this will necessitate weighing the field concentrations by the dielectric breakdown of the particular matrix composition. These two design metrics follow.

Design for Performance

The metric for comparing performance of the various designs is given by the induced stress constant of the composite (e_{33}). The trends are very similar to the field in the fiber, but account for the ceramic content (i.e. effect of X_1), as well as the fiber/electrode gap (X_2). In this comparison of performance, the continuous functions fitted from Chapter 2 data are again used as model inputs. The same case is examined as above, using a constant X_1=0.8 and X_2=0.92. Note that this assumes a constant composite thickness, even though fillers may increase X_2.

Figure 4.6a shows the piezoelectric induced stress as a function of the two filler ratios, for the plane stress assumptions. The induced stress constant increases from ~2 with no fillers, to 4 for a 75% PZT filled matrix. The use of carbon black increases the
matrix dielectric constant to 4.8 at 5% weight fraction. The combination of the two fillers is as expected. In all cases, the increases are proportional to the increases in matrix dielectric constant. Note that the actual design space is only a portion of the surface shown: carbon content is limited to 2% and 4% with 75% and 60% carbon black, respectively.

Figure 4.6b shows the same performance, normalized by the composite density (grams/cm$^3$). This normalization effectively levels the improvement of performance with added PZT fillers, showing little added benefit to these fillers. Note that if the fillers also increase the composite thickness, the use of PZT filler will be detrimental to the mass efficiency. The use of carbon black, on the other hand, does not significantly affect the mass normalized performance. This indicates that using small amounts of carbon black in the matrix is likely the best method for improving performance. However, the effect of carbon black on operational risk must still be determined.

**Design for Risk**

Risk-based design centers on choosing matrix compositions depending on the predicted risk for composite electrical damage during operation$^3$. Increased levels of filler increase the matrix dielectric, which in turn reduces the field concentration in the matrix, and increases the performance by allowing a larger portion of the field in the fiber. However, risk of dielectric breakdown also increases with the introduction of fillers. The

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$^3$ Poling can also be considered a risk. However, the DC breakdown strength is 2-3 times higher than the AC equivalent, and the field concentration in the matrix is lower in the resistively dominated DC regime.
question is whether the reduction in dielectric strength is outpaced by the benefits of the lower field concentration.

To answer that question, a new metric was developed. The premise for the metric is based on the assumption that all composite designs must perform to the same level of actuation, which is governed by the portion of field in the fiber ($E_{\text{fiber}} / E_{\text{comp}}$). The lower the ratio, the higher the field on the composite must be to maintain the same field in the fiber. If this ratio is divided into the ratio of field in the matrix ($E_{\text{matrix}} / E_{\text{comp}}$), this gives a measure of the field concentration in the matrix to maintain a set level of performance in the composite. This is shown in Figure 4.7a. For example, if a field of 7 kV/cm (~Ec for PZT 5H) was required in the fiber, this would correspond to a field in the matrix of ~190 times higher, for a composite with 75% PZT filler.

For design, this value still needs to be compared to the dielectric strength. The measured dielectric strength of 75% PZT filled matrix was 420 kV/cm (from Figure 2.19), while the above field corresponds to 1330 kV/cm. Since this was for a test sample of approximately 160 microns, while the actual matrix thickness in the concentration area is ~6 microns, the dielectric strength is not directly comparable. However, the relative degrees of risk for different designs can be assessed using the relative Risk Factor,

$$R.F.(%\text{PZT}, %\text{carbon}) = C \cdot \frac{E_{\text{matrix}} / E_{\text{fiber}}}{D.S.}$$

where $D.S.$ is the dielectric strength, and $C$ is a constant that accounts for the difference in sample thickness. Since the relationship between $D.S.$ and thickness is not known, $C$ is

![Figure 4.7](image.jpg)

*Figure 4.7* Design curves for operational risk as a function of PZT and carbon black fillers. (a) Field concentration in matrix for equal field levels in fiber; (b) Operational Risk Factor based on curve (a) and dielectric strength of matrix compositions, shown in Figure 2.19.
set to one. The resulting Risk Factor for the current designs is shown in Figure 4.7b, for PZT fractions up to 75%, and carbon black fractions up to 5%. Although the absolute value of the Risk Factor is meaningless, the relative difference in the factor for different designs is a direct measure of the increased likelihood of dielectric breakdown.

This figure shows some interesting results for the different matrix compositions. For 0% carbon black, the addition of PZT fillers slightly increases the risk. However, for the addition of carbon black to the epoxy (without PZT), the Risk Factor increases dramatically, due to the sharp drop in dielectric strength seen in the characterization tests for carbon black concentrations above 2%. The addition of PZT filler to the carbon black designs stabilized the dielectric strength, which is reflected in the sharp drop in risk for the combined filler cases. These results would seem to indicate that matrix compositions with only carbon black fillers would be a high risk design. This is in contrast to the design for performance. In that case, the ‘optimal’ choice for performance was to use pure carbon black fillers to maximize the mass normalized induced stress. Clearly, it is important to consider both the effects of performance and risk in designing active composites.

This study was shown as a function of matrix composition only. The effects of different geometry and fiber types are the following:

- **Fiber Composition**: Ceramics other than PZT 5H (used in this section) will have lower dielectric mismatch over the entire range of fillers, shifting the Risk Factor proportionally downward.

- **Composite Thickness**: Increasing the ceramic thickness line fraction ($X_2$) increases both the field in the fiber and matrix equally; there is no change in the Risk Factor for a different composite thickness.

- **Fiber Cross Section**: Square and round fibers that have the same $X_2$ will have equal field in the fiber and matrix, resulting in no change of Risk Factor. This also means that R.F. is independent of $X_1$.

As discussed in the opening of the chapter, these effects can be considered independent of the matrix design studies, and the combined effects are easily scaled.

### 4.3 Selected Composite Designs

The previous section outlined the issues and tradeoffs of using different fibers and matrix compositions. In order to validate the models, determine the remaining design inputs (such as composite thickness), and to iterate on the design process, a number of selected designs must be tested experimentally. The following describes the designs selected, summarizing the information learned from this design process.
4.3.1 Fiber Materials

A number of different designs based on fiber material and geometry were selected for manufacture, while holding constant the matrix composition. This was done to isolate the effects of the fiber on composite properties and performance. Based on the available fibers, only composites with one fiber diameter thickness were manufactured. Except for the study of ceramic fraction, all composites were manufactured with a ceramic width fraction of \( X_1 = 0.80 \) to facilitate easy manufacture, while maximizing ceramic content for high performance. The following designs were selected:

- In order to evaluate the Uniform Field model’s ability to predict composite properties with different volume fraction, manufacture composites with varying amounts of ceramic fiber. Since the composite thickness is fixed, the width ceramic fraction \( (X_1) \) will be varied to change volume fraction.

- Select designs with both square and round fibers to determine whether there are any unpredicted differences in performance, or differences in ease of manufacturing. Model predictions show that no benefit is gained from square fibers, except in mass efficiency of design.

- Manufacture with all available PZT ceramic types - PZT 5H, PZT 5A, and PZT 4 in this case. Each type offers different benefits to the composite performance. This will allow comparison of the different materials for both low field and high field behavior. Low field results will allow comparison with model predictions, while high field response will allow evaluation of the different ceramics for structural actuation control applications. This is important, as low field predictions cannot directly predict this regime.

4.3.2 Matrix Materials

A number of different matrix materials were selected for manufacture, while holding constant the fiber material and composite geometry. This was done to isolate the effect of matrix on performance and reliability. The nominal materials for manufacturing were round PZT 5H fibers, with width ceramic fraction as above \( (X_1 = 0.80) \).

- Select matrix materials with 0%, 60%, and 75% PZT filler in order to investigate the effect of PZT filler on composite thickness, and to determine whether this offsets the benefits of lowered dielectric mismatch. The PZT filled matrix compositions are at moderate to low risk for breakdown.

- Select matrix compositions with pure carbon black (5%, 7%, and 9%), which offer the highest performance benefit in terms of actuation capability.
Figure 4.8 Contour plot of the Risk Factor metric shown as a surface in Figure 4.7b. Open circle ("o") points refer to selected matrix designs for experimental validation.

According to the Risk Factor, however, these design are at high risk for breakdown. (see Figure 4.8).

- Select a number of low-risk designs combining both carbon black and PZT fillers to assess the stabilizing effect of PZT filler on the carbon percolation. These designs show approximately the same Risk Factor, as shown in Figure 4.8, and represent the outer limit of the design space with the given materials.

4.4 Summary

This chapter discussed a number of issues in the design of active fiber composites using different constituent materials. The design studies demonstrated the tradeoffs with material selection, which often involve conflicting choices for the optimal solution. Such a conflict exists with fiber material selection, where hard ceramics seem best suited for low field response and high stress designs, but soft ceramics are the material of choice for high field actuation. Choices in matrix composition also involve tradeoffs. Carbon black filled matrix is the optimal method for maximizing performance, whereas PZT filled matrix materials offer the lowest operational risk. Taking mass efficiency of the design into consideration may result in the best composition as one that contains no fillers at all. A number of these designs have been selected for experimental verification. The next two chapters involve manufacturing and experimental measurement of the properties, which will determine whether the model predictions in this chapter are realized in the actual composites.
Chapter 5

Manufacture of Active Fiber Composites

5.1 Introduction

The development of manufacturing techniques is an integral step in the development of active fiber composites. Manufacturing brings to light practical limitations in composite performance, often governed by the ability to produce particular geometry or work with specific materials. For example, the matrix viscosity and fiber geometry may set the maximum compaction and maximum composite size, respectively. In developing the techniques for manufacturing, it is also possible to come across innovations that could improve the design. Perhaps the most obvious reason for manufacturing, however, is to produce experimental data with which to compare the models. Comparisons often suggest improvements in the model, which when validated, can be used for more accurate design of future actuators.

Previous work in PFCs developed rudimentary methods for manufacturing (Bent, 1994), which lacked geometrical uniformity and repeatability in composite properties. Much of the difficulty was related to the low performance of the conventional electrodes, necessitating sanding of the composites prior to electroding, causing difficulty in these areas. However, this work identified many of the issues with manufacturing methods, and laid the groundwork for the methods presented in this chapter. This chapter describes new manufacturing techniques, which have been extended to accommodate interdigitated electrodes.

The previous chapter on actuator design defined the choices of materials and geometry that should result in the best composite actuators. Techniques for manufacturing and preparation of active composites are now described, using this information. The objectives for this chapter are,

- to develop standard methods for repeatable batch manufacturing of active fiber composites with interdigital electrodes,
• to identify, through the manufacturing process, the practical limitations in composite parameters
• and, to explore techniques for incorporating IDEPFC actuators into laminated structures.

The second of these objectives is really a task that is part of the iterative design process. Information of this type has already been used throughout the chapters in modeling and design to establish bounds on the regions of interest. While this chapter focuses on the actual manufacturing, the next chapter deals with poling and measurement of the composite response, and making comparison to the models of Chapter 3.

The first half of this chapter focuses on the manufacturing of the individual composite actuators. The baseline method is presented, including a discussion of the key issues of manufacturing and the materials used throughout the process. A step by step lay-up procedure is given, correlating descriptions with photographs of the process. Post-cure preparation of the composite for testing is also described. An alternate manufacturing method is described for manufacturing composites with different ceramic fractions, which are used to compare with model trends. Only the differences in manufacturing are highlighted. The second half of the chapter examines lamination issues when incorporating IDEPFC actuators into active structures. This section deals with lamination procedures, materials, and difficulties in embedded actuators. Two laminated rotorblade applications that apply IDEPFC actuators to structural actuation are summarized: a discrete flap actuator, and an integral blade actuator. Application-relevant issues are described.

5.2 IDEPFC Manufacturing Methods

The manufacturing section is divided into two parts. The first part (5.2.1) describes the baseline manufacturing procedure that is used in the manufacture of the majority of composites used in this work. It explains the techniques for manufacturing that were developed to permit repeatable performance in the composite actuators. The second part (5.2.2) describes the deviations from the baseline techniques which were used for manufacturing actuators that examine the trends of composite properties with increasing ceramic fraction. In each case, the materials and geometry are specific to the objectives of the particular method.

The first step in manufacturing was to identify the key requirements of the manufacture. i.e. the requirements that were used to drive the manufacturing process. The most important were the following:
**Voids:** The presence of voids (region where air or vacuum is trapped in the matrix) creates a concentration of electric field in this area. Small voids can be tolerated in matrix, but voids that extend substantially between the electrode fingers will cause dielectric breakdown. Thus, it is important to develop methods that minimize the introduction of voids, whether from mixing the matrix or curing, etc. Based on preliminary manufacturing of composites, it was decided that a vacuum would not be applied to the mold during cure. Only voids around the mold edges were successfully removed using a vacuum. Any small void trapped under the top electrode remained in place, and was greatly expanded in size by the vacuum, leaving the composite with many potential breakdown sites. To remove these trapped voids would require a much longer degas time, higher temperatures, or a porous top electrode (not tried in this work).

**Electrode Pattern Alignment:** It is important to have good alignment of the features between the bottom and top electrodes. Although no models have been created to predict the effect of misalignment, it is estimated that as little as one finger width of shift along the fiber direction will cause a significant change in field path lines and the planes of symmetry. This would reduce the composite actuation capability and make correlation with model more difficult. To ensure proper registration of electrodes, locating pins in the cure plate can be used to align the electrodes which have holes punched in the same spacing (outside of the active electrode area).

**Compaction:** The previous chapter showed that the improvement of composite actuation is extremely dependent on the thickness line fraction ($X_2$), illustrating the need for high compaction during cure. The limitation on $X_2$ ultimately depends on such items as the fiber diameter variability, and the finite PZT filler size which will act to separate the electrode and fiber. To reach the best possible compaction, it is necessary to develop techniques that ensure easy matrix flow, such as low viscosity matrix and removal of excess epoxy, and minimize the occurrence of fiber overlaps.

These ideas are incorporated in the following manufacturing methods.

### 5.2.1 Baseline Methods

This section describes the baseline method for composite manufacture. This procedure has evolved over the past three years with the input of Mr. John Rodgers and Mr. Alex Pizzochero. The majority of composite specimens (over 100) have been manufactured using this method, and it represents a standard, repeatable process that allowed reliable comparison between different material systems. The purpose of the baseline manufacturing was to develop a database of composite properties for a nominal material combination, and compare this with results for other fiber types and matrix
compositions. A large number of the nominal material composites were manufactured to create a statistical sampling of results for comparison with the model.

The individual materials used are described in the next section. The nominal materials were PZT 5H fibers and a 60 wt% PZT loaded matrix (no carbon black). Other combinations were manufactured to explore the design space defined in the last chapter. Variations on the fiber materials included circular PZT 5A fibers, and square PZT 5A and PZT 4 fibers. Variations on the matrix composition included 0% and 75% PZT loading, several levels of carbon black filled matrix, and some combinations of the two fillers. Only one variable (fiber or matrix) was varied in any one composite. The ceramic fraction was held at an $X_1$ of 0.8, a relatively high level of ceramic that would give substantial actuator authority, but avoid problems with fiber packing at increased ceramic fractions (such as 0.9 and higher). All other variables were held constant.

The geometry of the baseline actuator was chosen to facilitate several objectives. The composite ‘active dimensions’ had an aspect ratio of 5, with an active area length of 63.5 mm (2.5”) and width of 12.7 mm (0.5”). This conforms to ASTM Standards on tensile specimen testing (ASTM D3039, 1993), so that the composites could be used for mechanical fatigue and stress-strain data collection\(^1\) as well as for model comparison. The total composite length was actually 89 mm (3.5”), allowing a non-active area for gripping during testing. This aspect ratio maximized the longitudinal displacement output for easier measurement, while minimizing the number of fibers used in each composite.

An overview of the materials and assembly of the manufacturing process is shown in Figure 5.1. The entire lay-up was assembled on an aluminum cure plate, with vacuum fittings for degassing the matrix. Pins (0.125” dia) were set into the plate for accurately locating the top and bottom electrodes. The manufacture was a wet lay-up process, done completely by hand, and built up the composite in layers. This starts with the cure plate base, building the mold using cure materials (Teflon, flash tape, Kapton tape), and laying up the fibers and introducing the matrix. The top electrode was put in place, followed by more cure materials to protect and seal the composite, and top covers to which the clamping pressure was applied. More detail of this process is given below.

Prior to the manufacture, several preparation steps were needed. The vacuum plate was prepared by applying a thin coat of mold release to ensure easy removal of the samples and any excess epoxy that flows during cure. The aluminum top covers were also treated in the same manner. The top and bottom electrodes were aligned by eye, and the locator

\(^1\) Mechanical data is not directly collected in this work, although clamped piezoelectric strain is indirectly a measure of composite stiffness (see section 6.2.3).
pin holes were punched in the pair of electrodes simultaneously, using a 0.125” punch and die set created for this purpose.

*Composite Materials*

The following materials are used in the manufacture of IDEPFC actuators in this chapter. Several of these were described in Chapter 2, but are repeated here:

- **CeraNova PZT 5H/5A Fibers**: 130 μm diameter circular fibers, used in continuous lengths of 3.5” (9 cm).
- **Stavely Sensors PZT 5A/4 Fibers**: ~116 μm square fibers with known bulk PZT properties, used in continuous lengths of 3.5” (9 cm).
- **Shell Epon 9405 Resin**: a B-staging two-part epoxy chosen for its prepreg capability (allowing partial cure, storage, and then co-curing with other plies). This material is a structural bisphenol epoxy used for filament winding, RTM, and prepreg operations. The low viscosity and long pot life is ideal for the wet lay-up techniques used in manufacturing PFCs.
- **ICI Americas Hypermer KD-2 Dispersant**: a surface modifying agent that improves the wetting of the particles, promoting dispersion of the particles and lowering viscosity. Used as 5% by weight of PZT filler.
- **Morgan Matroc PZT 5H Powder**: a 1 μm average diameter powder, used as the dielectric filler.
• **Penn Color Carbon Black Paste:** a pre-dispersed slurry of Columbian Conductex Ultra high conductivity carbon black in part A of a bisphenol epoxy. Used as the conductive filler.

• **Etched Copper/Kapton Electrodes:** a 0.0005" Kapton substrate with a 2500 Å layer of sputtered copper (Southwall Technologies). Electrodes were etched in-house using photolithography techniques. The electrode pattern can be seen in Figure 5.3, and had the same electrode features as those used throughout the modeling chapter. The electrode fingers were 190 microns (0.0075") wide and spaced 1143 microns (0.045") apart. Side feeder rails ran the length of the active area, and were connected to terminal pads at each end.

Obviously, not all of these materials were used in any one composite. As described in the previous section, the nominal material set consisted of PZT 5H fibers and a PZT 5H doped (60%) matrix. Other composites investigated the effect of the fiber material and the matrix composition.

**Step-by-Step Lay-up Procedure**

The following is a step by step description of the manufacturing procedure. The numbers are correlated to the photographs in Figure 5.2, three pages ahead.

*Photograph 1 & 2: Mold Preparation*

Guaranteed non-porous Teflon (GNPT) was the first layer laid down, and ensured easy removal of the composite from the mold. The GNPT was cut larger than the bottom electrode, and to fit around the pins. The GNPT was taped down with flash tape (blue) on all sides to maintain a flat bottom surface. The top electrode was placed over the pins. The side and end mold pieces were cut from a double layer of Kapton tape (yellow). The mold pieces kept the fibers in place during manufacture. The material was soft enough that it would not restrict the extension of the fibers, and its height was slightly less than the fiber diameter so that full compaction would be possible. The placement of the side mold pieces was approximately 0.5 mm outside the ends of the electrode fingers. A small layer of air usually collected at the edge of the mold, and this positioning avoided voids in the active area which would lead to breakdown. The end mold pieces were positioned to accommodate 3.5" fibers, which placed them 0.5" from the ends of the active area. The bottom electrode was then taped down along all edges with flash tape to ensure no excess matrix would flow under the electrode. Small slits were made through the electrode and GNPT to avoid ballooning during vacuum degassing. Four composite samples were laid up in a single manufacturing batch.
Photograph 3&4: Fiber Preparation

Fiber preparation was a recent addition to the lay-up procedure, and was started when contaminants and/or moisture was suspected on the fibers. Contaminants were never found on any circular fibers (CeraNova), but it was known that the square fibers had residues from the dicing step of production. A single lot of fibers (for one sample) were washed with acetone for approximately 30 seconds, and allowed to air dry. The fibers were then dried at elevated temperatures (120°C) for 15 minutes to remove moisture. After the drying step, the fibers were immediately placed in the mold. The fibers were arranged so that they lay flat within the prepared mold, with a minimum of cross-overs. This facilitated later arranging after the matrix was applied. The fibers were covered with matrix within 10 minutes of being removed from the drying step.

Photograph 5&6: Matrix Preparation

This step was carried out concurrent to the fiber preparation. The matrix materials were placed into a mixing dish, and weighed on a scale to within 1% of the calculated amount. The matrix components were added in the following order: Epoxy Part A, Epoxy Part B, dispersant, and PZT. Following general mixing to homogenize the material, the matrix was mixed in a two step process. This was done to break up the large particle agglomerates into smaller primary particles and fully distribute the dispersant among the particles to minimize viscosity. First, the matrix was transferred to a mortar and pestle and mixed for 5 minutes. This acted as a coarse mixing step, breaking up the larger secondary particles. The matrix was then poured into a vial for ultrasonication. The ultrasonication step introduced high frequency energy to disperse the smaller aggregates. The vial was held in a warm water bath to maintain the temperature below ~70°C, to minimize boiling off any matrix components (solvents or other organics). The matrix was sonicated for 5 min.

Photograph 7&8: Applying Matrix to Fibers

The plate was heated to 50°C on a hot plate during this step to keep the matrix viscosity low. A very small amount of matrix material was applied to the fibers, allowing capillary action to move the matrix between the fibers. The capillary forces were relied upon to keep the fibers together and in place while arranging the fibers. Too much matrix material at this time caused the fibers to ‘float’, and cross over one another. The fibers were arranged using a small flat piece of Teflon or metal to gently push the fibers into a single layer, without overlaps which would reduce compaction. Once the fibers were arranged, additional matrix material was applied to the fibers, and distributed evenly along the composite length. Excess matrix was important to facilitate the removal of voids during application of the top electrode.
Photograph 9: Vacuum De-gassing

To remove voids introduced during matrix preparation and application, the entire assembly was vacuum de-gassed. A Plexiglas top cover was used to keep the vacuum bag off the uncovered samples. Standard composite vacuum tape and bagging were used to create the seal around the cure plate. Air was evacuated with a vacuum pump, which was used to apply a vacuum until air no longer appeared in the matrix (from 5 to 20 minutes, as necessary). Following the de-gas step, the bagging, tape, and cover were removed.

Photograph 10: Top Electrode

For each sample, the matching top electrode was placed on the open mold. This was accomplished by securing the electrode over two pins at one end, and rolling the electrode on from one end to the other. During this, a soft rubber material was used to push along the top electrode, keeping any trapped air moving ahead of the electrode as it was being placed. This largely eliminated voids caused by air entrapment. The top electrodes were taped down along the sides with flash tape to avoid matrix flow onto the outside of the top electrode, and to hold the top electrode in place during the next steps (to maintain good alignment). Any remaining air was gently pushed out using the rubber material, along the direction of the fibers.

Photograph 11: Mold Covers

To protect the composite from bonding with the top cover during cure, a layer of GNPT was placed on the top electrode, and taped down at the ends. A hard top cover was found to work best for uniformity and maximum compaction. Soft top covers (such as silicone rubber) tended to push the electrode between fibers, and reduce compaction due to the force distribution. Top covers were fabricated from aluminum cut exactly to the size of the composite. These covers were taped in place on top of the GNPT. Silicone rubber was subsequently taped on top of the aluminum covers, allowing more even distribution of clamping pressure among the four samples.

Photograph 12: Clamping of Samples

Pressure was conveniently applied using a large top plate and four clamps positioned at the corners, above each sample lay-up. The pressure was estimated to be 20-40 psi, using a force transducer. An autoclave was used (allowing more accurate pressure application) for preliminary manufacturing investigations, but the additional time and cost needed to operate the equipment was not worthwhile. Sufficient repeatability and compaction could be achieved with the clamping technique for the given geometry and ceramic fraction. The entire assembly was cured for 3 hours at 360°F (175°C).
Figure 3.2  Step by step layup procedure for piezoelectric fiber composites
Figure 3.2 Step by step layup procedure for piezoelectric fiber composites (Cont’d)
Post Cure Sample Preparation

Following the cure, the specimen was allowed to cool slowly to room temperature. After removing the top covers and GNPT, all flash tape was removed and the composite and mold was lifted off of the pins in one piece. The composite was trimmed around the side electrode rails and terminal pads, as shown in Figure 5.3. Some portion of the Kapton tape which acted as the side mold remained under the electrodes, but this would not significantly clamp the actuation and could easily be accounted for in models. One set of terminal pads were selected for connection to lead wires, while the other set was kept for backup in case of damage. Lead wires were soldered to the pads, and the top and bottom pads were soldered together at this time. To protect the copper on the pads from fatigue during poling and testing, Kapton tape was wrapped around the pads and lead wires, as seen in the figure.

Figure 5.3 Photograph of baseline IDEPFC specimen, slightly larger than actual size (3.5" by 0.5"), showing lead wire attachment and protective tape.

5.2.2 Manufacture for Model Verification (M Series)

A second manufacturing technique with slightly different procedures was developed for manufacturing composites with different ceramic fractions. These composites were used to compare trends of models over a wide range of ceramic fraction, from 10% $X_1$ to 90% $X_1$. This necessitated a different approach to manufacturing. Since the specimens varied so widely, it was important to control the uniformity of the specimens to a greater extent.

The composites were manufactured in a square configuration, as shown in Figure 5.5, so that longitudinal and transverse properties could both be measured equally well. The electrodes were prepared in the same manner as in the baseline approach (photo-etching), although the dimensions were now 25 mm by 25 mm (1" by 1"). The electrode
Figure 5.4 Manufacturing method for IDEPFC actuators that were used for examining trends with ceramic fraction.

finger width and spacing were maintained at 190 microns and 1143 microns, respectively. The same nominal material set was used in these composites, which consisted of CeraNova PZT 5H fibers, and 60 wt% PZT 5H filler particles in the matrix (and 5% KD-2 dispersant). A different low viscosity epoxy was used, although it was also a bisphenol epoxy that used an amine curing agent. The properties of this matrix were characterized in the previous work (Bent, 1994). No other material combinations were used.

A similar lay-up was used as the baseline method. The specimen was again assembled on an aluminum cure plate equipped with vacuum fixtures (Figure 5.4). The plate was treated with mold release to ensure easy specimen removal from the plate after cure. The bottom electrode was put in place over the pins and held down with two side Teflon mold pieces (15 mil thick), which completely restricted matrix flow to occur along the fibers. Fibers were evenly spaced by hand one at a time, according to the ceramic fraction, and held in place using a thin piece of double sided tape placed at each end of the bottom electrode. The tape was necessary to maintain the placement of the fibers, and the high Teflon side mold pieces ensured that no matrix flow would damage the fibers. The fibers were much more susceptible to damage, as there was no lateral support along their lengths from adjacent fibers (for lower ceramic fractions). Porous Teflon lead away from the fiber ends in both directions to facilitate removal of excess matrix.
Figure 5.5 Photograph of IDEPFC actuator manufactured for examining trends with ceramic fraction. Actuator shown twice actual size (1" by 1").

The matrix was prepared in the ratio previously specified. After applying the matrix, the top electrode was placed on the specimen, followed by a thin GNPT top (0.001") and an aluminum cover, similar to the baseline methods. The composites were cured in an autoclave system, so that an exact pressure could be applied to the composite. This was necessary due to the range of ceramic fractions, and the desire for obtaining composite thickness as uniform as possible between the samples. The clamping technique of the baseline method would have applied higher forces to the fibers in the composites with lower ceramic fractions. The entire assembly was enclosed in a vacuum bag during cure. No vacuum was applied (for the reasons stated earlier), but the bag provided the pressure differential for the compaction. The composite was cured for one hour at 65°C and 80 psi pressure. Following cure, the composite was cut from the mold, and prepared for testing in the same method as the baseline actuators.

5.3 IDEPFC Lamination and Integration Issues

This section deals with issues related to using IDEPFC actuators in laminated structures, particularly for incorporation and operation without damage to the active material. It begins with a description of simple lamination techniques, consisting of actuators between two host plies for the evaluation of clamped actuation capability. Three types of host structural materials were used: glass, stainless steel, and 0°/90° E-glass fabric. Two applications of composite actuators are also described, the discrete trailing edge flap actuator, and the integral blade actuator. The applications were based around proof of concept demonstrations for active composites in rotor blade control. In each case, the application is briefly described, and the experience gained is summarized. For the discrete actuator, this involved procedures for lamination of IDEPFCs, and operational issues for bender configurations. For the integral blade actuator, the experience was primarily in
developing manufacturing techniques for 45° actuators, and the repair of electrical damage in composites.

5.3.1 Lamination Procedures

For the evaluation of clamped actuation behavior, the baseline (T series) and M series actuators were laminated to several materials. The baseline actuators were laminated to fiberglass fabric to evaluate the high field performance of the actuators, and evaluate the effect of the highly nonlinear portion of the strain on performing useful work. The M series actuators (which were developed for model comparison) were laminated to isotropic host materials to determine the piezoelectric clamped stress constants, which can be indirectly used to evaluate the fiber stiffness. In both cases, the lamination procedures were similar.

Figure 5.6 shows the materials and lay-up used in the lamination of the M series (1” by 1”) actuators to the isotropic host materials. The actuator is surrounded by one ply of host material on either side, which was either glass or stainless steel. The glass substrate was 0.15 mm (6.0 mil) thick, cut from microscope cover slides. The stainless steel was 0.079 mm (3.1 mil) thick, cut from a roll of shim stock. Both were sized to exactly cover the active area of the composite. All surfaces were prepared by cleaning with acetone prior to applying the epoxy. The same low viscosity epoxy (Epo-Tek 301) used for manufacturing the actuators was used for bonding as well. Surrounding the host materials, GNPT (guaranteed nonporous Teflon) was used to avoid adhesion to other materials. Silicone rubber is used to distribute the clamping force as evenly as possible over the sample for even bond thickness, and to avoid breaking the glass. A hard backing material (aluminum), placed surrounding the rubber material, supports the pressure applied to the

![Diagram of lamination process](image)

**Figure 5.6** Materials and approach to laminating 1" by 1" actuators to isotropic host material for piezoelectric clamped stress testing.
assembly. The epoxy was cured according to the manufacturer’s instruction of 60 minutes at 65°C.

For the baseline actuator lamination, a very similar approach was used. In this case, lamination was to adjacent E-glass fiberglass fabric plies that were impregnated with resin. The fiberglass was cut from a continuous roll of material which was kept at low temperatures to minimize pre-polymerization of the resin, and warmed to room temperature prior to lamination. The only other difference was an additional thickness of aluminum was placed next to the sample (with the GNPT in between), in the same manner as in the baseline actuator manufacture. This was due to the softness of the host material, which needed a hard backing in order to compact evenly over the length of the actuator. The material was cured according to the manufacturer’s instruction of 90 minutes at 120°C.

Several issues arose from working with the laminated actuators, the most important of which are summarized here:

**Thermal Depoling:** thermal depolarization from the lamination cure was initially a consideration in the lamination of actuator materials. The composites were always poled *before* lamination. Poling after lamination was not studied, although the compressive stresses caused by restraining the poling strains, in addition to the thermal compressive strains of lamination, would likely inhibit the full polarization of the soft PZT 5H fibers. A second reason for poling prior to lamination are the oil environment and temperature used during poling which may not be desired after lamination. It was found that temperatures up to 175°C could be tolerated for extended periods of time without reducing the degree of polarization in PZT 5H (Trapa, 1996). Thus, common graphite/epoxy and other such cures would fall within acceptable temperature limits, as long as temperature overshoot during cure does not happen (which can often occur with poorly designed controllers).

**Host Materials:** it was found that metal host materials were not sufficiently isolated from the electrodes to allow operation at typical high voltage levels of 2.5 kV (~20 kV/cm field between the electrodes). At these voltage levels, the composites routinely shorted to the steel. Thus, the 12 micron Kapton substrate plus ~12 micron bond layer was not sufficient for insulating. This implied that there would also be difficulties with materials such as graphite/epoxy and other PMCs (polymer matrix composites) with graphite fibers, and all MMCs (metal matrix composites). Use with these materials would necessitate using another insulating structural material (such as fiberglass) separating the two plies.
Strain Measuring Techniques: it was found that strain gages were a poor method for measuring the strains of the composite actuators, especially un laminated samples. For one reason, the use of strain gages provided a local response of the actuator in one location, rather than an averaged global response. In some cases, measurement with laser interferometry provided significantly different strain levels (up to 15% higher). This was especially important in the some of the low ceramic fraction samples (10% and 20% X_1), where measured local response was heavily dependent on composite uniformity. Any eccentricities that caused bending in the composite would also cause errors in the measured response. These effects were largely eliminated in the laminated actuators.

Another difficulty with using strain gages in the un laminated composites was the phenomenon of capacitive coupling at moderate frequencies. Conventionally electroded composites allowed placement of the gages on the ground side, which avoided the large potential difference between the gage foil and electrode. With the interdigital pattern, however, only the Kapton of the electrode and gage, and the bond layer (about 0.0015” in total), separated the sensor from the applied field. This created a capacitor in parallel with the gage resistance, which increased in magnitude with frequency. As the frequency approached 100 Hz, this caused a large apparent hysteresis and a roll upwards in strain response as the phase angle was increased. The effect was significantly stronger in the transverse measurements, where the electrode fingers and gage foil were aligned in parallel. At high voltages, the field was sufficient to cause shorting to the gage, a problem similar to the conductive host plies. Lamination with glass and the E-glass plies solved these problems.

5.3.2 Application - Discrete Flap Actuator

The Discrete Flap Actuator was an application that used IDEPFC actuators to actuate a flap which could be used in the trailing edge of an airfoil. Deflection of the flap would cause aerodynamic forces to twist the blade, permitting aeroelastic control for reducing vibrations caused by Blade Vortex Interactions (see Chapter 1). This application was significantly different in nature from the simple laminated composites of the last section. This is due to the multiple layers of actuators that were stacked together and operated in a bender configuration. This introduced several interesting issues surrounding the operation of the material and considerations for laminating the actuators.

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2 This work was conducted as part of an MIT senior design project (Ilvedson, 1996; Lutz, 1996).
**Figure 5.7** Photograph of discrete flap that incorporates 10 plies of IDEPFC actuators laminated with tapered sandwich core. Actuators operated in bending configuration with motion of laminate amplified through leveraged flexures in compression molded flap.

A photograph of the complete flap actuator is shown in Figure 5.7. The structure shown represents several distinct parts. The bender portion (Figure 5.8), which consisted of the actuators, would be housed within the trailing edge fairing of the blade. A foam shim was used to separate the IDEPFCs in order to maximize the bender efficiency, as was the use of stepped actuators. In fact, the design was driven by impedance matching the structural stiffness to the aerodynamics expected on the flap at a particular airspeed. The actuators were bonded at the thick end to an aluminum bar which simulated the clamped end condition of the main spar, and allowed attachment to the Lexan test stand. The flap mechanism itself (white in the photograph), was manufactured from a single piece of Delrin. Two offset attachment points, operating through flexures, caused a leveraging action which magnified the bender motion by a factor of ten. This design was used with monolithic piezoceramics (Hall and Prechtl, 1996), but the advantage of the IDEPFC actuators was the lower weight, which would provide a much better weight distribution (less weight aft of the quarter chord).

Several important issues were recognized for successful use of these actuators in this application:

**Lamination:** Placement of adjacent layers was extremely crucial. For stacked actuators, it is necessary that electrode features bonded closest together have the same voltage potential. It was also important in this bender configuration to have all actuators on one side operate in phase. These can be accomplished by
Figure 5.8 Bender portion of flap actuator, illustrating IDEPFC ply arrangement. Adapted from (Ilvedson, 1996).

- Marking the poling direction for each actuator, and grouping all ground leads of the actuators to one side.
- Positioning adjacent plies so that the side rails are exactly on top of one another, and the electrode fingers are aligned with the same polarity.

To ensure the alignment was not lost during the lamination cure, a two step process was used. A fast cure adhesive (cyan acrylate) was placed at the corners of the actuators to be bonded, while the usual bonding epoxy was applied to the rest of the surface. The fast cure adhesive did not inhibit the thin bond layer of the epoxy, but ensured the actuators were held in place while the bonding epoxy cured. The adhesive was not used for the entire surface due to its inability to flow (and cover all areas), and its brittle nature.

**Operation:** Several operational issues needed to be solved during the use of the flap actuator. The representative work cycles used for driving the composites required DC offsets on a sinusoidal input in order to maximize the available strain (see section 2.2.1). It was important to ensure exactly 180° out of phase operation of the two actuator groupings, which required either a two output signal function generator (which would require two amplifiers), or use of the same AC signal for both actuator groupings if the components were wisely arranged. In this case, one DC high voltage output, one high voltage amplifier, and one function generator were used, with the arrangement shown in Figure 5.9. This arrangement utilized a floating ground on one grouping of actuators, and two voltage offsets on opposed poling directions to correctly drive the bender. Limiting circuits were also designed to limit the peak voltages to +2100 volts with the direction of poling and -900 against the direction of poling (first described in Hall and Precht, 1996). This was implemented to avoid depolarization (or damage) due to incorrectly applied fields.
Figure 5.9 Wiring and drive components for actuation of bender configuration flap actuator. Adapted from (Lutz, 1995).

Interestingly enough, depolarization due to voltage was not experienced, but the actuators were partially depoled in one grouping during operation near resonance, due to compressive stress. The low structural damping in the system caused the depole stress to be reached when operating at a frequency near the first bending mode of the system (with flap). This was evidenced by a sharp drop in flap deflection and drop in capacitance in the depoled grouping. This is a typical problem in soft piezoceramics, and illustrates the difficulty of using PZT 5H in applications. From section 2.2.2, it is known that full polarization is not reached at room temperature poling, so that re-polarization in service may not be an option.

5.3.3 Application - Integral Blade Actuator

An alternative method of active rotor blade control is to apply actuators integral to the blade structure. A 1/16th Froude scale blade modeled on a Chinook CH-47D helicopter was manufactured to demonstrate the concept of integral actuation through the use of IDEPFC actuators (Hagood and duPlessis, 1995). Figure 5.10 shows the planform view of the model blade, which was 483 mm (19") in length and 50 mm (2") wide, with a NACA 0012 profile.

The blade was manufactured using wet lay-up procedures, using 2 plies of E-glass fabric wrapped around a foam core. A total of six IDEPFC actuators were embedded between the two plies, three on each side. These actuators were non-standard in design, as the fibers were oriented at ±45°. The three top actuators were oriented at +45° with respect to the blade axis, while the bottom three were oriented at -45°. This arrangement induced shear flow around the closed box beam cell, causing a net twist in the blade structure.
Figure 5.10  Photograph of 1/16th scale Chinook helicopter blade model, manufactured using wet lay-up techniques. Three IDEPFC actuators (±45°) laminated within blade per side.

Figure 5.11  Photograph of 45° actuator mold, illustrating fibers placed in separate bays formed by Kapton mold sides and spacers.

Figure 5.12  Photograph of 45° IDEPFC actuator.
The majority of new experience that came from this application surrounded the adaptation of manufacturing techniques to the 45° angle. Although 45° actuators could theoretically be cut from 0° plies, this would create large waste. More importantly, the electrodes must be designed for the particular fiber orientation and cannot be generalized. Thus, the 45° actuators must be made uniquely. The second important experience came from dealing with electrical damage in embedded actuators.

**Manufacture of 45° Actuators:** The manufacture of 45° IDEPFC actuators depended heavily on the knowledge gained from manufacture of baseline actuators. Only six actuators were manufactured, all of which were used in the model blade, illustrating the importance of an established, repeatable manufacturing process. Exactly the same bulk materials and cure materials used in the baseline actuators were used here, with one exception. The matrix material used 75% (wt) PZT filler in epoxy, instead of the nominal 60%, to increase the composite response.

Figure 5.11 is a photograph of the mold for the 45° actuators. Locator pins were not used to match the top and bottom electrodes. Instead, the bottom electrode was taped down on all sides with flash tape, and the top electrode was positioned over it using alignment marks at the far ends of the electrode. The top electrode was taped down along one of the long sides, and then rolled back during the lay-up process. This was successful in demonstrating that good alignment of the electrodes in cured composite (at least large ones) could be achieved without locator pins. However, the pins were much easier to use for the smaller composites, and provided a more repeatable process for the dozens that were manufactured.

The Kapton tape molds were modified to accommodate the 45° fibers. The side mold pieces were positioned along the side rails, as usual. The end mold pieces were now oriented at 45°, parallel to the placement of the fibers (but perpendicular to the electrode fingers). In addition to the end mold pieces, 1 mm thin mold spacers were positioned at 25 mm (1") intervals, dividing the composite mold into five separate bays. Previous experience in manufacturing 45° conventionally electroded composites showed difficulty with fiber alignment (Rodgers, 1995a). In this case, fibers often rotated and slid along the side mold pieces, causing large errors in the effective fiber angle. These separate bays were a means for limiting misalignment of the fibers. The fiber ceramic fraction was held at 80% X 2 (as usual), giving 111 fibers per bay. All other steps were kept the same, including placement of the top GNPT, silicone rubber, and top aluminum covers over the composite. This resulted in successful manufacture of the first 45° IDEPFC plies (Figure 5.12).
**Figure 5.13** Typical damage sites and damage characteristics for dielectric breakdown in baseline actuators (and the blade).

**Electrical Damage Repair:** Electrical damage was a common occurrence in baseline actuators when pushing at high electric fields, or even at low fields in actuators with flaws. Often flaws could be identified as void related, although some otherwise 'perfect' specimens experienced dielectric breakdown. This might be attributed to intermittent porosity in the fibers, air or moisture at the fiber/matrix interface, or contaminants in the matrix or on the fiber surface. These flaws provide a path of lowered 'resistance' to breakdown. Over the testing duration of the model blade, several breakdowns occurred in the actuators, causing the epoxy to carbonize in an area which would then act as an electric short.

Typical damage found in the baseline actuators (similar in the blade) is illustrated in Figure 5.13. Virtually always, the damage starts with a line between two electrode fingers. Then, depending on the time until the voltage was removed, destruction of the electrode fingers along a path leading away from the original breakdown site. One of the most common sites of initiation was at the end of a finger, near the edge of the side mold (site A), where air commonly collected during the application of the matrix. This air was not a problem when the Kapton mold pieces did not move into the active region during cure. However, since the Kapton tape was used as spacers in the 45° plies, there were potential locations for trapped air in the active area.

A method was developed for repairing damaged actuators. This was done by drilling out the damage, and backfilling the hole with epoxy. In applying this repair, it was important to remove all of the carbon from the area to prevent future breakdown in the same location. Sometimes portions of electrode fingers left electrically floating (unconnected to side rails) were the site of future breakdowns, probably due to charge which collected on the copper, and then discharged when a high enough potential was reached. This repair was successfully used on the blade actuators to allow a second round of testing after multiple shorts. However, as
would be expected, the results (section 6.4.3) showed reduced actuation capability. These repairs were also successful with the laminated baseline actuators, although this was rarely done, as the reduced actuation was not useful for comparison with models. It does, however, demonstrate that active materials can be repaired in laminated structures, if the damage area can be identified (a common problem in composites). In the conventionally electroded composites laminated with graphite/epoxy (Rodgers, 1995a), delamination was evident at the site of dielectric breakdown, and the damage was easily identified.

5.4 Summary

In this chapter, techniques for manufacturing IDEPFC actuators were described in detail. These included methods for manufacturing baseline composites for exploring different combinations of fiber and matrix materials, composites to examine the trend of properties with ceramic fraction, and lamination of these composites for evaluating the stress inducing capability. These methods were the culmination of three years evolution in the manufacturing process, the result of which was a repeatable procedure for batch manufacturing. Two applications that incorporated IDEPFC actuators for structural actuation were introduced in this chapter. The experience in the laminating and operating of these applications were summarized, providing information for the next step in structural control. The next chapter will describe testing methods, and show the results of testing for the composites and the applications.
Chapter 6

Experimental Determination of Composite Properties

6.1 Introduction

The previous chapters dealt with the modeling, design, and manufacture of active fiber composites. In this chapter, the results from experimental measurements of composite performance and properties will be compared with the predictions, and the actuator design will be evaluated. This comparison will close the design loop, creating a continuous path from bulk characterization to the laminated active structure. This completes the upward flow of capabilities in the vertical technology integration philosophy, and now permits the downward flow of requirements to redefine the metrics and materials. This results in an update of the modeling assumptions for more accurate predictions, and points to changes or improvements in manufacturing to achieve better actuators.

The question of model accuracy arises when discussing model-data correlation. In this case, the accuracy of the model can be expected to vary depending on the composite geometry and the regime of operation. Geometry refers to the question of whether the actual manufactured geometry is well represented by the model assumptions, and whether the assumptions are accurate throughout the range of composites likely to be used. This is addressed by examining a full range of comparisons for a variety of materials and fiber fractions. Regime of operation refers to the extension of assumptions made for low field modeling and low field bulk material characterization to predicting composite operation at high field levels. Both low field and high field properties are measured and compared to models.

The objectives for this chapter are,

\footnote{For example, comparison of model and experiment illustrated the model assumptions of Combination Model 1 were inappropriate for the geometry, and lead to the development of Combination Model 2 which captures the trends at high ceramic fraction.}
• to develop methods for characterizing the active composite electrical and electromechanical properties,
• to validate the Uniform Fields model's ability to predict low field linear composite properties,
• to examine the high field performance of the IDEPFC actuator, including laminated actuation capabilities, and
• to evaluate the actuator designs of Chapter 4 in order to select the most promising active composite for structural actuation.

This chapter begins by examining the geometry of the resulting composites, and describing methods for characterizing the low field properties, including poling the actuators and measuring the dielectric properties. The piezoelectric properties are also determined, using a quasi-static approach to determine electromechanical coupling, of both free and clamped actuators. Classical laminated plate theory is introduced for the purpose of determining the induced stress piezoelectric constants. Comparisons are made with models in a summary of experimental results, divided into two sections. The first section examines the model's ability to predict trends in composite properties with varying ceramic fraction. The second section further evaluates the accuracy of the model with the baseline actuator, for several combinations of fiber and matrix materials. The high field performance of the actuators is measured using the Representative Work Cycle (introduced in Chapter 2) as a metric. Finally, the two IDEPFC laminated applications are revisited, where the results of the testing are tied to the lessons learned in developing the actuators.

6.2 Sample Geometry and Poling

6.2.1 Geometry

Figure 6.1 shows the microstructure of three of the manufactured baseline actuators, with different fiber types and matrix compositions: circular PZT 5A doped with filler, square PZT 5A doped with filler, and square PZT 4 undoped. The white matrix indicates PZT filler at 60% by weight. The M series (1" by 1") composites are not shown, but are very similar to the other circular fiber composites in geometry. The samples were embedded in potting epoxy, and polished using a series of polishing diamond pastes, down to a 1 micron abrasive. Although this reduces scratches in the photomicrograph, it eliminates much of the fiber and filler microstructural detail. Photographs of the PZT 5H fiber microstructure can be seen in Figure 2.9.
Figure 6.1 Cross section photographs of IDEPFC composites, shown approximately 300 times magnification. Composite thickness ~ 160 µm.
Table 6.1 Comparison of geometry for five types of the manufactured composites. Doped matrix refers to nominal matrix material (60% wt PZT, no carbon black).

<table>
<thead>
<tr>
<th>Fiber Type</th>
<th>Matrix Type</th>
<th>Number Tested</th>
<th>Avg. Fiber Size (μm)</th>
<th>Aspect Ratio</th>
<th>Composite (X_1)</th>
<th>Composite (X_2)</th>
</tr>
</thead>
<tbody>
<tr>
<td>circular 5H</td>
<td>doped</td>
<td>40</td>
<td>129 ±2</td>
<td>1</td>
<td>0.80</td>
<td>0.920±0.024</td>
</tr>
<tr>
<td>circular 5H</td>
<td>undoped</td>
<td>4</td>
<td>129 ±2</td>
<td>1</td>
<td>0.80</td>
<td>0.955±0.004</td>
</tr>
<tr>
<td>circular 5A</td>
<td>doped</td>
<td>4</td>
<td>128±4</td>
<td>1</td>
<td>0.80</td>
<td>0.916±0.007</td>
</tr>
<tr>
<td>square 5A</td>
<td>doped</td>
<td>4</td>
<td>114±10</td>
<td>1.17</td>
<td>0.90</td>
<td>0.865±0.009</td>
</tr>
<tr>
<td>square 4</td>
<td>doped</td>
<td>4</td>
<td>117±8</td>
<td>1.13</td>
<td>0.92</td>
<td>0.858±0.011</td>
</tr>
</tbody>
</table>

In general, the relatively high ceramic fraction \(X_1\) ensured uniform packing throughout the composites, although differences can be seen among the fibers in size and geometry. Table 6.1 summarizes the measured geometry of both fibers and composites for five manufactured composites\(^2\). The circular fibers (5H and 5A) were relatively uniform in diameter, and this is reflected in the composite geometry. The fibers were consistent in diameter, approximately 129 μm (5.09 mil) and 128 μm (5.04 mil) for the PZT 5H and 5A respectively, with a small standard deviation. An occasional large diameter fiber is seen in the composite (perhaps 140 μm), which increased the composite thickness locally. This general uniformity in size permitted good compaction, which resulted in high values of the thickness line fraction, \(X_2\). The fibers were uniformly packed across the thickness, and had an \(X_1\) of 0.80.

The square fibers varied significantly in size, which caused variations in the composite geometry. The fibers had a nominal size of 100 μm (4 mil), and the composites were manufactured to maintain the same ceramic content as the circular fiber composites. This was done to account for the decrease in thickness and the increase in ceramic content due to the square shape. However, the square fibers were actually larger than 100 μm, and had a significant difference in lengths of the two fiber sides. i.e. the square fibers were rectangular. The square PZT 5A had an average minimum thickness of 105 μm, and an average maximum thickness of 123 microns, giving an average aspect ratio of 1.17. The square PZT 4 fibers had an average minimum thickness of 110 microns, and an average maximum thickness of 124 μm, giving an average aspect ratio of 1.13. The average of the minimum and maximum thickness is used to determine the line fractions \(X_1\) and \(X_2\) (as shown), used as inputs to the models, although this assumes that the long side is oriented

\(^2\) This is a sampling of manufactured composites that illustrate the geometry differences. Other composite combinations were
equally often in each direction. Examination of the composite microstructure appeared to confirm that this was true, on average.

The effect that the aspect ratio problem will have on the performance is evidenced by the difference in $X_2$ values between the circular and square fiber composites. The high aspect ratio in the square fibers caused a significant drop in the average line fraction. This means, on average, that the square fiber was further away from the electrode than the circular fiber. This is also evidenced in Figure 6.1. Thus, the square fibers were much less effective than they could be if the aspect ratio was closer to one. This may be compounded by the fact that the square fibers may also reduce compaction due to their flat sides. The excess matrix may not be able to flow around the square fibers as effectively, to be removed at the ends of the mold.

Table 6.1 also shows the effect of the PZT filler on the composite thickness. Comparing the undoped and doped PZT 5H fiber composites shows that the thickness line fraction was reduced from 0.955 to 0.920 (which corresponds to a thickness increase of 5 microns (0.2 mil)) when the matrix contained 60% PZT filler. This increase could be caused by the increase in matrix viscosity or the finite size of the particles (Chapter 2), both of which impedes composite compaction. As the design chapter demonstrated (Figure 4.3), the lower thickness of the undoped composite may offset the reduced matrix dielectric, maintaining the performance of the composite while also decreasing the weight.

### 6.2.2 Poling and the Coercive Field

The fibers arrived from the manufacturer in an unpoled state, meaning that the composites had to be poled before any electromechanical coupling would be present. However, the coercive field of the composite first needed to be measured before the proper poling conditions could be determined.

The poling conditions for the composites were based on the study in Chapter 2 on bulk PZT 5H. The degree of poling was established as a function of time, temperature, and applied field, where the electric field was based on percentages of the coercive field of the material. In order to measure the coercive field of the composite samples, the actuators were tested in the planar strain measuring device (Figure 2.6) to determine the voltage level where repolarization occurred. The sample was clamped in the same manner as the cover slides for the single fiber testing, with weights placed on the sample to eliminate any bending component that might cause errors in the movement of the sliding retroreflector.

---

manufactured (such as with carbon black matrix), and are discussed in section 6.3.3.
The current on the high voltage amplifier was also recorded, so that the electrical displacement could be determined, and compared to bulk. Electrical displacement (D) is simply the time integral of the current, divided by the electrode area,

\[ D = \frac{1}{A} \int I dt \]  \hspace{1cm} (6.2)

In this case, D was determined numerically from a forward Euler summation of the current data. The area is defined by the electrode in the active area of the composite, where charge collects (see Figure 6.7).

Typical butterfly curves and hysteresis loops are shown in Figure 6.2 for the baseline actuator configuration with the four fiber types. As in the bulk materials, the curves are shown for an applied voltage that is approximately twice the coercive field. The coercive field for the material was determined either from the repolarization voltage in the strain versus electric field curve, or the point where the electrical displacement was zero. From the strain (butterfly) curve, the repolarization appeared much more rounded than for bulk ceramic materials. This suggested a wider distribution of repolarization fields for the various families of piezoelectric domains. For the circular fibers, this was probably caused by the fact that some portions of the circular fiber were closer to the electrode than others. For the square fibers, this could be attributed to the aspect ratio of the fiber cross section, where orientation of the fiber long axis was distributed between the 1 and 2 axis directions.

This distribution of repolarization fields may also explain the apparent coercive fields of the composites. Table 6.2 summarizes the coercive fields for the bulk ceramic and composite samples. The table also shows the model prediction for the ratio of field in the fiber to the applied field (\(E/E_{comp}\)), which varies from 27% to 36%. According to this ratio, the coercive fields of the composites should have been significantly higher than that for bulk - three to four times higher. However, this was not the case. The composite coercive fields were at most 1.5 times higher than bulk. In fact, for PZT 5H fibers, the composite coercive field was less than the fiber value (10 kV/cm versus ~10.8 kV/cm for fiber). These results pointed to the likelihood that some dipoles were reversing direction early, causing a low apparent coercive field, while others required higher fields before reorienting by 180°. It was also possible that the repolarization was progressive, starting near the electrode where the field concentration was highest, and moving along the fiber length. The reorientation of the dipoles would be assisted by adjacent dipoles which have already switched, providing a favorable stress/energy state for the new position. This highly nonlinear phenomenon was suggested by recent work in nonlinear repolarizing finite elements, applied to the IDEPFC problem (Ghandi and Hagood, 1997). This illustrated the difficulty of using linear models to describe piezoceramic materials at high fields.
Active Composite with PZT 5H Fibers (Circular)

Active Composite with PZT 5A Fibers (Circular)

Active Composite with PZT 5A Fibers (Square)

Active Composite with PZT 4 Fibers (Square)

Figure 6.2 Typical hysteresis and butterfly curves for a baseline actuator with nominal matrix (60 wt% PZT filler), and different fibers. Voltage applied at 1 Hz.
Table 6.2  Comparison of coercive field and remnant polarization values between bulk and composite. *Bulk values for the PZT 5H dielectric and piezoelectric properties have been reduced by 1/3 from true bulk values, as measured in Section 2.2.3. Uniform Fields model prediction of field in fiber also shown for comparison. ± denotes standard deviation on data.

<table>
<thead>
<tr>
<th>Fiber Type</th>
<th>Matrix Type</th>
<th>Model E/E&lt;sub&gt;comp&lt;/sub&gt;</th>
<th>E&lt;sub&gt;c&lt;/sub&gt; (bulk*) (kV/cm)</th>
<th>E&lt;sub&gt;c&lt;/sub&gt; (comp) (kV/cm)</th>
<th>P&lt;sub&gt;r&lt;/sub&gt; (bulk*) (mC/m&lt;sup&gt;2&lt;/sup&gt;)</th>
<th>P&lt;sub&gt;r&lt;/sub&gt; (comp) (mC/m&lt;sup&gt;2&lt;/sup&gt;)</th>
</tr>
</thead>
<tbody>
<tr>
<td>circular 5H</td>
<td>doped</td>
<td>0.31</td>
<td>10.8</td>
<td>~10</td>
<td>200</td>
<td>~10</td>
</tr>
<tr>
<td>circular 5H</td>
<td>undoped</td>
<td>0.27</td>
<td>10.8</td>
<td>~10</td>
<td>200</td>
<td>~10</td>
</tr>
<tr>
<td>circular 5A</td>
<td>doped</td>
<td>0.36</td>
<td>11.2</td>
<td>~11</td>
<td>375</td>
<td>~11</td>
</tr>
<tr>
<td>square 5A</td>
<td>doped</td>
<td>0.27</td>
<td>11.2</td>
<td>~11</td>
<td>375</td>
<td>~11</td>
</tr>
<tr>
<td>square 4</td>
<td>doped</td>
<td>0.31</td>
<td>~8</td>
<td>~8</td>
<td>~110</td>
<td>~110</td>
</tr>
</tbody>
</table>

A final interesting feature of the butterfly curves was the unsymmetric nature of the response. The butterfly curve was lop-sided in all cases, so that the strain was higher for negative fields than for positive fields. This was consistent with all composites, regardless of fiber type or geometry. Eventually the curves became symmetric with even higher electric field. It appeared the same as a ceramic that had some degree of poling already present. This cannot be attributed to any pre-stresses induced during cure, because this would tend to orient the dipoles at 90° to the poling axis, nor can this be attributed to processing that would preferentially orient the lattice structure because both the extruded and diced fibers exhibit this asymmetry. It is likely related to the composite processing or geometry in some way, but the exact cause is unclear.

The hysteresis curves reflected the asymmetry of the butterfly curves, but in a much more subtle manner. These curves also reflected the broadening of the coercive field, in the slope of the curve during switching. The transition from positive to negative electrical displacement (and vice versa) was much less sharp, which indicated a more gradual flow of charge as the dipoles aligned. Also, the ‘flat’ portion of the curve after switching was much less flat than bulk materials. The coercive field values matched those found from the butterfly curves.

The composite remnant polarization (P<sub>r</sub>) is summarized in the last column of Table 6.2. The remnant polarization is another quantity not easily predicted using the linear models. The values are not dependent on the ratio of field in the fiber, but are a direct function of ceramic volume fraction. A complication is the fact that some portions of the fiber may not be fully polarized near the electrode. However, relative differences in these values provide additional information about the various composites. The P<sub>r</sub> for the undoped and doped PZT 5H composites was virtually identical, which was expected
because the fiber PZT type and quantity was similar in both cases. Passive materials such as the matrix cannot possess residual spontaneous polarization, and will not contribute to the composite value. The value for the circular PZT 5A was much less than that for the square, which may indicate lower than bulk properties in the circular fibers.

The poling conditions for the PZT 5H fiber composites were selected from the poling study conducted in Chapter 2, and the measured composite coercive field. This consisted of poling for 20 minutes at 60°C and 2 kV. Since the coercive field may have varied according to the position in the fiber, higher poling fields were tried. This did not change the response, indicating that full poling was achieved in the composite at this level\(^3\). The PZT 5A and PZT 4 fiber composites were also poled for 20 minutes, but the applied field levels and temperature were different to reflect the increased hardness of these ceramics. These composites were poled at 3 kV, approximately twice the measured room temperature coercive field. The temperature used in the PZT 5A and PZT 4 fiber composites was 80°C, as reported in the literature for these materials. Following poling, the composites were allowed to age for 24 hours, as suggested in the aging study of section 2.2.2.

6.3 Model Verification - Low Field Properties

This section describes the methods and results for low field testing of the active composites. The composites tested in this section were selected as points in the design space of potential actuators, determined in Chapter 4. Low field testing was used to determine the effective properties of the composite, which could be compared with the Uniform Fields predictions. The measured properties provided a measure of effectiveness of the different material combinations, so that the matrix and fiber selections made in the design chapter could be evaluated. The next section (6.4) describes testing at high field, which is focused on determining the performance in structural actuation applications.

6.3.1 Measurement Methods

Dielectric

The capacitance was measured using the same equipment as used for the matrix characterization, described in section 2.3.3. The capacitance values were taken at 400 Hz. The composite dielectric could be calculated from the capacitance using the following equation,

---

\(^3\) This was largely due to the higher field in the fiber at DC, caused by the lower fiber/matrix resistive mismatch
where L was the spacing between electrode fingers (the equivalent parallel plate capacitor), and A was the electrode area, both of which were set by the electrode pattern. L is 1.143 mm (0.045") and A depended on the composite series, and was calculated from the number of electrode pairs, and the length and width of the fingers in the active area (illustrated in Figure 6.7). For the baseline composites, this area was 111 mm², and for the M series (1" by 1" samples) it was 89 mm².

**Electromechanical Properties - Piezoelectric Free Strain**

Quasi-static methods were used for the measurement of the piezoelectric free strain constants \( d_{33} \), \( d_{31} \). The quasi-static methods were validated on bulk PZT 5H, which showed that this method resulted in the same free strain constants as resonance methods conducted by the manufacturer for the same samples (Bent, 1995). The quasi-static technique involved measuring the strain for an applied voltage (the converse piezoelectric effect).

Composite samples were tested in the planar displacement device again, where a 1 Hz sinusoidal voltage was applied to the specimen (Figure 6.3a). The effective d constant for that voltage level was found from a linear least squares fit of the slope of strain versus electric field for a single cycle (1 sec). However, since piezoceramics exhibit a significant nonlinearity, this value of d constant was dependent on the applied field level. Thus, the d constants were measured at several field levels: ranging from 5% to 40% of the composite.

![Figure 6.3](image-url)  
**Figure 6.3** Illustration of quasi-static technique for determining piezoelectric d constants. Shown for an M series actuator with nominal materials. (a) Measuring the d constant at a given field. Slope taken as linear least squares fit. (b) Extrapolating the d constants measured at different fields to the low field value. Quadratic and linear least squares fits used for free and clamped response, respectively.
coercive field, as shown in Figure 6.3b. An extrapolation of the $d$ constants to the zero field level provided the low field linear $d$ constant, which is typically reported. This best fit for the unclamped free strain constants was found to be a quadratic (least squares) with voltage, as shown in the figure. However, values for the strain constants of laminated actuators were sufficiently captured with a linear (least squares) fit. This is due to the fact that the piezoelectric nonlinearity is strain based (Chan, 1994), and the clamping drastically reduces the strain output. This is discussed in more detail in the section on high field testing (6.4.1). This technique was applied to measuring the longitudinal $d$ constant ($d_{33}$), and the transverse $d$ constant ($d_{31}$).

**Electromechanical Properties - Piezoelectric Induced Stress**

The goal of the composites is successful use in a laminated application. Thus, it is important to know how the actuator performs under these conditions in order to determine the actuator authority. The metric representing actuator authority at low field is the induced stress capability of the material, which is a combination of the material stiffness and free strain capability. A technique for assessing the induced stress capability in planar materials involves bonding the active material to host materials with well known properties. Using laminate plate theory, it is possible to predict the residual strain, for comparison with the experimental results. The analysis can also be used with the measured residual strain to estimate the piezoelectric induced stress constants ($e_{33}$, $e_{31}$).

As described in the previous chapter, the composites were laminated to various host materials. The M series (1" by 1") composites were laminated to isotropic materials, glass and stainless steel, to determine the low field induced stress constant, for comparison with model predictions. A high level of clamping was used to investigate the effect of a stiff substrate material. The baseline composites were bonded to E-glass fiberglass fabric, a structural material often used in the manufacturing of rotor blades. The residual actuation

![Figure 6.4 Axis convention for laminated actuators.](image)

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Table 6.3 Comparison of the baseline actuator and the materials used in the lamination. Baseline and E-glass mechanical properties taken from Rodgers et al., 1996. Properties for other materials taken from Cook and Young, 1985.

<table>
<thead>
<tr>
<th>Material</th>
<th>Thickness (mm)</th>
<th>Stiffness (GPa)</th>
<th>Poisson’s Ratio</th>
<th>Stiffness Ratio</th>
</tr>
</thead>
<tbody>
<tr>
<td>Baseline</td>
<td>0.140</td>
<td>34</td>
<td>0.3</td>
<td>1</td>
</tr>
<tr>
<td>E-Glass</td>
<td>0.083</td>
<td>21</td>
<td>0.15</td>
<td>0.73</td>
</tr>
<tr>
<td>Stainless Steel</td>
<td>0.078</td>
<td>190</td>
<td>0.30</td>
<td>6.2</td>
</tr>
<tr>
<td>Glass</td>
<td>0.149</td>
<td>70</td>
<td>0.17</td>
<td>4.4</td>
</tr>
<tr>
<td>Bond Layer</td>
<td>~0.0125</td>
<td>3</td>
<td>0.33</td>
<td>0.02</td>
</tr>
</tbody>
</table>

Capability in these actuators was used to examine the performance of different material systems at high field levels. The materials used are summarized in Table 6.3. The stiffness ratio is a measure of the relative stiffness of the host material to the actuator,

$$\psi = \frac{2E_{h}t_{h}}{E_{a}t_{a}}$$  \hspace{1cm} (6.4)

where E is the material stiffness, and t is the thickness. The factor of 2 comes from the two plies of host material around the actuator. The stiffness ratios are given with respect to the baseline actuator with $X_{i}=0.8$, although the M series actuators (which are bonded to glass and steel) start at approximately these same values, but increase for the lower ceramic fraction composites.

The laminated composite plate model follows the standard analysis found in composite mechanics books (Jones, 1975), augmented with piezoelectric terms (see Bent et al., 1993, for more details). The model assumes linear behavior, allows no variation in geometry within the plane, and ignores shear lag. Since the laminated structures here are symmetric, the bending and twisting components can be ignored.

The standard form for the laminate response is found by a piecewise integration of the constitutive relations (Equation 2.3) over that thickness of the laminate,

$$\begin{bmatrix} N_{1}^{a} \\ N_{3}^{a} \end{bmatrix} = \begin{bmatrix} A_{11} & A_{13} \\ A_{13} & A_{33} \end{bmatrix} \begin{bmatrix} S_{1} \\ S_{3} \end{bmatrix}$$

where $S_{1}$ and $S_{3}$ are the planar strains (Figure 6.4 for axis convention), and $A_{ij}$ are the laminate stiffnesses, found by summing the plane stress lamina stiffnesses over the k plies,

$$A_{ij} = \sum_{k} (c_{ij} t_{k})$$

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The plane stress stiffnesses are used as a standard assumption in thin laminated plate theory. The \( N_1^a \) and \( N_3^a \) are the in-plane applied forces per unit length in the 1 and 3 direction. Since no mechanical forces are applied to the laminate in this experiment, these forces are due only to the active materials, where

\[
N_1^a = \sum_k (e_{31} E_3 t)_k \\
N_3^a = \sum_k (e_{33} E_3 t)_k
\]  

(6.7)

where the \( e_{33} \) and \( e_{31} \) constants are the (plane stress) piezoelectric induced stress constants, which are related to the free strain and stiffness through the matrix relation

\[
e = d c^E
\]  

(6.8)
as discussed in Appendix A. Since only one ply is active (the IDEPFC), these reduce to

\[
N_1^a = e_{31} E_3 t_{idepfc} \\
N_3^a = e_{33} E_3 t_{idepfc}
\]  

(6.9)

By inverting equation (6.5), the residual actuation capability of the laminate becomes a function of the applied electric field. This can be used to predict the response of the laminates to actuation of the IDEPFC layer,

\[
\begin{bmatrix}
S_1 \\
S_3
\end{bmatrix} = \begin{bmatrix}
A_{11} & A_{13} \\
A_{13} & A_{33}
\end{bmatrix}^{-1} \begin{bmatrix}
N_1^a \\
N_3^a
\end{bmatrix}
\]  

(6.10)

Of course, prediction is based on assuming the IDEPFC stiffness, which has not been measured. The measured free strain constants (\( d_{33} \) and \( d_{31} \)) can be incorporated into the prediction through the relationship in equation (6.8).

So far, the data from the clamped tests has not been used. By comparing the predictions with the measured values for residual actuation, additional information can be gained. The stiffness constants still cannot be explicitly determined, as there are 3 unknowns (\( c_{11}^E, c_{13}^E, c_{33}^E \)), and only two available measurements (\( S_1, S_3 \)). However, it is possible to implicitly determine the clamped stress constants (\( e_{33}, e_{31} \)), using a variation of the above equations and a simple technique.

Since the free strain constants are known, the unknown stiffness of the composite is varied until the residual strain in equation (6.10) matches the measured value. The clamped constants are then found by substituting equation (6.9) into equation (6.5), and dividing by the actuator thickness and the applied field,

\[
\begin{bmatrix}
e_{31} \\
e_{33}
\end{bmatrix} = \begin{bmatrix}
A_{11} & A_{13} \\
A_{13} & A_{33}
\end{bmatrix} \begin{bmatrix}
S_1 \\
S_3
\end{bmatrix} \frac{1}{E_3 t_{idepfc}}
\]  

(6.11)
In matching the residual strains \((S_1, S_3)\), only the longitudinal stiffness of the composite was varied. Varying the transverse stiffness \((c_{11}^E)\) within 50% of the nominal (i.e. model predicted) value had little effect on the output strains, since this direction was much softer than the longitudinal value, and the stiffness ratio of the host materials was so large. Note that the non-active plies considered in this analysis included not only the host material, but also the bond layer, electrode, and IDEPFC dead area. The results of this analysis are summarized in the next section.

6.3.2 Results for Varying Ceramic Fraction

This section evaluates the Uniform Fields model for a range of composite geometry, while holding the constituent material properties constant. The M series actuators (1” by 1”) were manufactured for this purpose: to providing model-data correlation over a range of ceramic fractions, in order to see whether trends with geometry were correctly captured by the model. This validation of the model is important in order to carry out accurate design of actuators.

The actuators used the nominal material set: PZT 5H circular fibers, and a 60 PZT wt% doped matrix (no carbon black). As discussed in the manufacturing chapter, a different base resin was used, resulting in a higher matrix dielectric constant for this series (11.3, as compared to 9.8 for the baseline actuators and the nominal materials). This resulted in higher \(d\) and \(e\) constants than for the baseline series. However, this resin was not suitable for structural systems. The thickness line fraction \((X_2)\) was manufactured to be as high as possible, using techniques identical in all actuators. The width line fraction \((X_1)\) was varied from 0.10 to a maximum of 0.88. The manufacturing of these actuators was discussed in section 5.2.2.

The results from the testing of these actuators was compared to the Uniform Fields model. The finite element model could not be used for comparison, because the high thickness line fractions caused difficulty with element aspect ratios. A further problem was the large number of elements needed to capture the response accurately in this geometry regime. This demonstrated the utility of the Uniform Fields model in design. The results are compared specifically to Combination Model #2 for the round fibers, which is appropriate for the given geometry.

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Figure 6.5a Comparison of measured piezoelectric free strain with Uniform Fields model along the fibers (3 direction). Single sample data with measurement error indicated.

Figure 6.5b Comparison of measured piezoelectric free strain with Uniform Fields model transverse to fibers (1 direction). Single sample data with measurement error indicated.
**Figure 6.5c** Comparison of measured piezoelectric stress constant with Uniform Fields model along the fibers (3 direction). Single sample data with measurement error indicated.

**Figure 6.5d** Comparison of measured dielectric (relative to vacuum) with Uniform Fields model (3 direction). Single sample data with measurement error indicated.
The results are shown in Figure 6.5. Shown are the piezoelectric free strain in both planar directions ($d_{33}$ and $d_{31}$), the zero stress dielectric $\varepsilon_{33}^T$, and the piezoelectric clamped stress (or induced stress) in the fiber direction ($e_{33}$). All of these predicted constants had to be manipulated from the model outputs in order to get the plane stress constants. Appendix A describes the different forms of the constitutive relations. Two lines generated by the model are shown for comparison. These represent the variation in specimen thickness, which varied between 0.137 mm and 0.142 mm without electrodes (an $X$, from 0.909 to 0.942), a difference of only 5 microns that has a large effect on the resulting properties. The circles indicate data from a single experimental specimen. The error bars are a conservative estimate of measurement error only, and cannot account for the variations in manufacturing or bulk materials processing.

All four measured properties show excellent agreement with model. The model appears to capture both the trend and the magnitude of the properties. The free strain constants ($d_{33}$ and $d_{31}$) start at zero for pure matrix, but increase quickly with additional ceramic and level off as the fibers overcome the matrix clamping effect. The constants eventually asymptote to the final free strain value, which is dependent primarily on the reduction in field reaching the fibers due to the low dielectric matrix. In fact, the $d$ constant is a direct measure of the portion of the field reaching the ceramic fiber. The clamped stress constant ($e_{33}$) is linearly varying, but continues to increase with $X_1$ because the added stiffness linearly increases the actuator authority. The one extremely high value is most likely due to poor bonding to the host material, which would appear as a higher stress inducing capability. Finally, the relative dielectric begins at a value of 11.3 for the pure matrix and increases linearly with ceramic fraction, as expected in the parallel addition of capacitors.

The values of free strain constants indicate a substantial improvement over the conventionally electroded PFC, showing a fivefold increase from approximately 30 to 150 pm/V for the piezoelectric constant along the fiber direction. Such an increase is expected, given the improvements that the interdigital electrode provides, as described in section 3.2.2. This would also tend to increase the induced shear stress by an order of magnitude or more for twist applications.

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4 This comparison is made for specimens made with a separate electrode material (such as copper/Kapton), not for specimens that have been sanded prior to electroding with silver paint or deposited metals.
6.3.3 Baseline Actuator Design Results

The previous section examined the trend in composite properties while varying the fiber ceramic fraction. This section evaluates the Uniform Fields model, and the improvement in composite properties when the geometry is fixed, and the bulk materials are varied. The bulk materials were varied in such a way as to cover the most promising choices identified in the composite design space (see Chapter 4), as well as choices that represented a higher risk, but helped to define the acceptable design boundary. These results, when used in conjunction with the high field tests of the next section, permitted comparisons among the available material combinations in order to determine the best potential actuator for applications. This testing also allowed further exploration of the effectiveness of the Uniform Fields method for a variety of different cases. Baseline actuators were used exclusively in this section (actuators with 5:1 geometrical aspect ratio).

Fiber Compositions

Different fiber materials were evaluated to determine the performance tradeoffs in composites with different PZT compositions. Chapter 4 highlighted these performance tradeoffs, including the piezoelectric free strain, clamped residual actuation, and stress depolarization to name a few. The different fibers used in the experimental work were those available at the time of manufacturing: PZT 5H, PZT 5A, and PZT 4. Since each fiber type offered a particular benefit, composites of each were manufactured. In particular, the expected benefits were:

- **PZT 5H** - high bulk electromechanical coupling
- **PZT 5A** - lower dielectric to increase composite response, high saturation strains
- **PZT 4** - high stiffness to improve clamped capability, and high compressive depolarization stress

The PZT 5H and 5A were available in circular form, while the PZT 5A and 4 were available in square (rectangular) form. As section 6.2.1 showed, the fiber geometry should have an effect on the performance, due to the maximum thickness line fraction. The matrix was held constant at the nominal composition (60 wt% PZT filler) to isolate the effect of the four fiber types. The results of the low field testing is summarized in Table 6.4. The results for undoped PZT 5H composites are also shown for comparison.
Table 6.4 Model-data comparison of the low field constants for baseline composites manufactured with different piezoceramic fiber materials. Basic measurement error for capacitance and induced strain constant of ±8% and ±7%, respectively (see Appendix B). Standard deviations for measured data shown as ± values after the mean.

<table>
<thead>
<tr>
<th>Fiber PZT Type</th>
<th>Matrix Type</th>
<th>Predicted Dielectric</th>
<th>Measured Dielectric</th>
<th>Predicted $d_{33}$ (e-12 m/V)</th>
<th>Measured $d_{33}$ (e-12 m/V)</th>
</tr>
</thead>
<tbody>
<tr>
<td>circular 5H</td>
<td>doped</td>
<td>411</td>
<td>577±54</td>
<td>108</td>
<td>106±22</td>
</tr>
<tr>
<td>circular 5H</td>
<td>undoped</td>
<td>366</td>
<td>508±69</td>
<td>95</td>
<td>98±23</td>
</tr>
<tr>
<td>circular 5A</td>
<td>doped</td>
<td>360</td>
<td>461±15</td>
<td>130</td>
<td>92±16</td>
</tr>
<tr>
<td>square 5A</td>
<td>doped</td>
<td>364</td>
<td>542±7</td>
<td>99</td>
<td>111±10</td>
</tr>
<tr>
<td>square 4</td>
<td>doped</td>
<td>324</td>
<td>496±36</td>
<td>87</td>
<td>91±15</td>
</tr>
</tbody>
</table>

A large discrepancy existed between the measured and predicted composite dielectric constants. Since the agreement was very good on the M series composites, this was unexpected, but the explanation possibly rests in the calculation of the electrode area. This electrode area was used to find the composite dielectric constant from the measured capacitance. In the models (both the FEM and Uniform Fields), all of the electrode material consisted of electrode finger in the active area (see Figure 6.6). For practical reasons, electrode side rails and finger extensions are needed. However, this adds to the conductive material where charge can collect, increasing the apparent dielectric of the composite due to the underestimation of the electrode area. In the baseline actuators, this area was ~30% of the composite width. Unfortunately, there was no easy way to determine the effective electrode area, and its value would depend on the material system being used. In the M series actuators, this area was a much smaller portion of the width, so that the error was less.

The piezoelectric free strain constant $d_{33}$, however, compared very well between model and experiment. In all cases except the circular PZT 5A, the measured values fell

![Figure 6.6 Illustration of the dead area of the IDEPFC actuator.](image-url)
within 12% of the model predictions. This provided additional validation of the model for
different fiber geometry and ceramic compositions. For the circular PZT 5A, it was likely
that the fiber properties were not equal to the bulk properties (as in the circular PZT 5H).
This was also evidenced by the composite remnant polarization, discussed in section 6.2.
Overall, there was not a significant difference between the measured free strain capability of
the different fiber types. Thus, there is no clear conclusion on the best fiber type to select
based on this data, but this was largely due to the high geometrical aspect ratio of the
'square' fibers, and the non-bulk properties of the extruded fibers. Using the Uniform
Fields models, square fibers with lower aspect ratio closer to one would likely show a
better performance for the PZT 5A and 4 square fiber composites.

The above results showed that the low field performance was well captured by the
linear Uniform Fields model. It was also expected that using the Uniform Fields model in
conjunction with the laminated plate theory should capture the low field \textit{laminated}
response of the actuators. Section 6.3.1 described the method for predicting the response of the
laminated composites, where Equation 6.10 gives the predicted residual strain of the
system. For comparison with data, this equation was modified by dividing through by the
applied electric field to determine the laminated/residual piezoelectric $d_{33}$ constant. The
inputs to the model were the measured free strain constant, the material properties
(including the E-glass fabric, bond material, electrodes, etc.), and the Uniform Fields
predicted composite stiffness.

The results are shown in Table 6.5. As expected, the models adequately predicted
the laminated response. All of the measured laminated piezoelectric constants are within
12% of the predicted values. The values were closest for the circular fibers, while the
square fibers perform less well than predicted under clamping. The last column in the table
shows the ratio between the laminated and free $d_{33}$ constants, as measured. Approximately

\begin{table}[h]
\centering
\caption{Model-data comparison of the low field laminated piezoelectric constants for
baseline composites manufactured with different piezoceramic fiber materials. Basic
measurement error of ±7% assumed for induced strain (see Appendix B).}
\begin{tabular}{|l|l|l|l|l|}
\hline
Fiber Type & Matrix Type & Predicted Lam $d_{33}$ (e-12 m/V) & Measured Lam $d_{33}$ & $d_{33}$,Lam/$d_{33}$,Free
\hline
Circular 5H & doped & 53 & 49 & 0.49
Circular 5H & undoped & 48 & 45 & 0.44
Circular 5A & doped & 49 & 48 & 0.52
Square 5A & doped & 67 & 59 & 0.53
Square 4 & doped & 59 & 58 & 0.59
\hline
\end{tabular}
\end{table}
50% of the actuation is retained, with the values varying slightly between ceramic types. The undoped composites retain less than the doped counterparts, perhaps due to the less stiff matrix. The PZT 5A composites have higher residual strains than the PZT 5H. In the square fibers, this may have been partly due to the slightly higher ceramic fraction. The PZT 4 fibers retained a significantly higher portion of the actuation than the others, which can be attributed to the higher stiffness (and thus, induced stress capability).

From these results, it would appear as though PZT 4 fibers are best suited to actuation of structures. The low dielectric constant offsets the lower bulk d constant, and the higher stiffness gives high actuator authority. However, the performance at high field is also important to quantify for structural actuation, and is not simply an extension of the low field capabilities. This will be dealt with in the upcoming section (6.4.1).

**Matrix Compositions**

Different matrix materials were used to evaluate the effect of dielectric mismatch on composite performance, and to determine whether conductive fillers were a viable means for improving the composite response. A number of promising matrix compositions were identified in the design chapter, based on several criteria. These criteria included considerations such as processing viscosity, dielectric constant, and dielectric strength. This resulted in the selection of the following compositions:

- **(0,0)** - Pure epoxy; for low viscosity processing to maximize compaction; low weight.
- **(60,0)** and **(75,0)** - 60% PZT and 75% PZT, without carbon black; as a method for increasing the matrix dielectric.
- **(60,3), (60,4),** and **(75,2)** - PZT filler with low amounts (2-4%) of carbon black; for increasing the apparent matrix dielectric using conductive fillers; PZT increases 'stability' of matrix by reducing continuous chain formation.

In addition to these compositions, several higher risk, pure carbon black/epoxy compositions were attempted. Although the design chapter identified these as candidates for dielectric breakdown, they were included to better define the design boundary. The compositions tried were **(0,5), (0,7),** and **(0,9)**. For all of these composites, only PZT 5H circular fibers were used in order to isolate the effect of the matrix on performance.

The results of the composites are summarized in Table 6.6. The first two columns define the matrix composition, as weight percentage of PZT and carbon black. The next two columns show the composite dielectric constant, as a percentage of the pure epoxy case **(0,0)**. This was done to illustrate the effect of added fillers, and avoid the difficulty with
### Table 6.6 Model-data comparison of the low field constants for baseline composites manufactured with different matrix materials. Basic measurement error for capacitance and induced strain constant of ±8% and ±7%, respectively (see Appendix B). Except for 60% PZT series, only two samples were made for each matrix composition (no std. deviations are given).

<table>
<thead>
<tr>
<th>Matrix wt% PZT</th>
<th>Matrix wt% Carbon</th>
<th>Predicted Dielectric</th>
<th>Predicted $d_{33}$ (e-12 m/V)</th>
<th>Measured $d_{33}$ (e-12 m/V)</th>
</tr>
</thead>
<tbody>
<tr>
<td>0</td>
<td>0</td>
<td>100%</td>
<td>100%</td>
<td>95</td>
</tr>
<tr>
<td>0</td>
<td>5,7,9</td>
<td>187%-308%</td>
<td>Breakdown</td>
<td>181-297</td>
</tr>
<tr>
<td>60</td>
<td>0</td>
<td>112%</td>
<td>112%</td>
<td>108</td>
</tr>
<tr>
<td>60</td>
<td>3</td>
<td>140%</td>
<td>121%</td>
<td>136</td>
</tr>
<tr>
<td>60</td>
<td>4</td>
<td>144%</td>
<td>Breakdown</td>
<td>142</td>
</tr>
<tr>
<td>75</td>
<td>0</td>
<td>138%</td>
<td>138%</td>
<td>136</td>
</tr>
<tr>
<td>75</td>
<td>2</td>
<td>179%</td>
<td>124%</td>
<td>176</td>
</tr>
</tbody>
</table>

the electrode area (as discussed previously). The last two columns display the predicted and measured free strain constants for the different matrix compositions.

All of the composites with matrix compositions that did not contain carbon black, performed as predicted. The 60% PZT composition, (60,0), had performance comparable to the pure epoxy case (0,0), showing an improvement of only 12%. The higher matrix dielectric was mostly offset by the thicker composite caused by the filler particles. The 75% PZT composition, (75,0), showed a greater increase in dielectric and $d_{33}$ constant, and had a thickness similar to the (60,0) composite. This seemed to indicate that the composite thickness may be as much due to the finite particle size as to the processing viscosity. If the performance of each of these composites was normalized by mass, the undoped matrix (0,0) would perform slightly better. In each case, the Uniform Fields model accurately predicted the response.

The composites with carbon black filler as a part of the matrix composition did not experience the improvements expected. The Uniform Fields model predicted an increase in composite performance directly proportional to the measured increase in matrix dielectric, as presented in Chapter 2. Instead, the composites had performance similar to those composites with the same level of PZT, without carbon black. Both (60,3) and (75,2) had $d_{33}$ constants close to (60,0) and (75,0) respectively. The (60,4) samples all experienced dielectric breakdown prior to poling, which established a portion of the boundary for safe designs with carbon black filler. The composites with pure carbon black and epoxy matrix, (0,5), (0,7), and (0,9), also did not survive the prepole testing. In
all of these breakdown cases, the prepole capacitance of the samples varied widely, indicating the formation of random chains of carbon black. Unfortunately, the dielectric breakdown is a weakest link phenomenon, so that the composites break down before it is possible to take advantage of the overall lowered dielectric mismatch which would lower the required field.

The lack of improvement in the combined carbon black/PZT filler composites could be caused by several reasons, linked to the random percolation of the carbon black. As chapter 2 showed, the dielectric of the matrix samples varied from no additional benefit with carbon black, to a doubling of the dielectric over the PZT filler alone. This was attributed to the varying degrees of dispersion in the different samples, even though the processing was carefully controlled. This variation was also seen in the composite samples that experienced dielectric breakdown. For example, the unpoled dielectric of the (4,60) samples ranged from 560 to 933, similar to the increase experienced in the matrix. The (7,0) composites demonstrated unpoled dielectric constants from 800 to 35000.

Another contributing factor could be due to mechanical strain in the matrix. Work with conductively loaded epoxy has shown changes in electrical properties with expansion of the material (Strumpler et al., 1995). Expansion during curing or temperature changes caused contacts between particles to open, producing an increase of seven to nine orders of magnitude in the resistivity of the matrix. This size change occurred because the system properties were percolation dominated, and relied upon the continuous pathways between the electrodes. For expansions on the order of 6000 ppm, these pathways opened, causing the resistivity to approach the values for an unpercolated system. A similar effect on matrix dielectric might be expected. Another work, with combined carbon black and PZT fillers, showed that the dielectric and d constant of these 0-0-3 connectivity composites varied as a function of the processing pressure (Sa-Gong et al., 1986). The values increased 20% for a 50 MPa pressure.

The matrix samples tested in this work were manufactured using procedures that would likely leave the samples macroscopically stress-free: slow cooling and heating during cure, and no restriction on the volume expansion/contraction during cure. In the composite system, the high thermal coefficient mismatch between the fiber and matrix causes a compressive pre-stress in the fibers, and a tensile stress in the matrix upon cooling. Since the carbon black is near percolation at 2-3%, it plays the role of creating low resistivity paths between the dielectric particles with small conductive chains, increasing the apparent dielectric. With the application of a tensile stress, these chains are broken, and the matrix dielectric relies on the intrinsic increase from the dielectric particles. The increase due to the PZT filler is not affected in the same way, as the dielectric filler is

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not at percolation, and the effective dielectric from combining dielectric materials depends on a completely different phenomenon. As a result, the composites with carbon black would exhibit the same behavior as their counterparts without. For matrix with high values of carbon black (5-9%), the density of carbon black may be sufficient that breakdown still occurs, or the lack of PZT may contribute to the potential of breakdown. A further complication is the fact that the PZT powder may become partially polarized, causing changes in the local stress fields.

6.4 High Field Properties - Performance

The previous section summarized the low field testing and properties of the active composites. The low field properties are a measure of the effectiveness of the fiber and matrix combination - the efficiency of the design. The piezoelectric free strain and induced stress constants provide a metric for the composite capability. This section showed that the tradeoffs in bulk ceramic properties allowed fibers with otherwise low electro-mechanical coupling (such as PZT 4), to perform extremely well in composite form. The section also showed that the response of the composites at low field could be captured accurately by the linear Uniform Fields model.

The high field regime is an extension of the low field effectiveness, but additional material behaviors come into play. This section looks at the response of the composites at high electric fields, using the Representative Work Cycle as a measure of the capability, for both free and laminated operating conditions.

6.4.1 Representative Work Cycle Performance

Representative Work Cycles were first introduced in section 2.2.1 in the characterization of different ceramics, as a means of comparison between different PZT types at high fields. This metric was created to deal with the use of ceramics in structural actuation applications, where the low field constants \((d, e)\) are not adequate to describe the high field capability. The Rep Cycle is also ideal for comparing the relative performance of active composites, either with different constituent materials, or with the same materials to determine variability in properties due to manufacturing/materials. The cycle is defined as a voltage cycle that maximizes the work output while avoiding electrical damage or repolarization, which can cause numerous problems including fatigue and loss of performance. The size of the cycle is defined by the electric field applied to the composite, with respect to its coercive field \((E_c)\). Since most applications will require harmonic inputs, the Rep cycle is assumed to be a sinusoidal voltage with a DC bias, and for a given bandwidth of frequencies.
Figure 6.7 Typical Representative Work Cycle performance for a baseline actuator with nominal materials (actuator #60 shown); (a) Comparison of the nominal and extended Rep Cycles; (b) Reduction of strain with frequency for the nominal Rep Cycle.

Figure 6.7a shows two potential Rep Cycles for a baseline composite with the nominal material set (PZT 5H fibers, and 60 wt% PZT filler). The lower (nominal) Rep Cycle was defined as a voltage cycle from -75% $E_c$ against the direction of poling, to +200% $E_c$ with the direction of poling. This resulted in a strain for this particular sample of ~840 ppm. The larger (extended) Rep Cycle was defined as a voltage cycle from -75% $E_c$, to +450% $E_c$. The lower limit does not change due to repolarization, while the upper value can increase indefinitely until electrical damage occurs or the strain saturates. This resulted in a much higher strain, approximately 1750 ppm for the given cycle. The difficulty of using such a high Rep Cycle, however, was the increased likelihood of damage. Up to 50% of the actuators tested to this level failed through dielectric breakdown (attributed to voids and fiber porosity). Since the objective of the Rep Cycle was to avoid damage, the lower Rep Cycle was chosen for comparison of all composites. This cycle was equivalent to voltage between -900 volts to +2100 volts (3000 $V_{pp}$, with a 600 $V_{dc}$ offset).

Figure 6.7b shows the Rep Cycle performance for the same actuator as a function of frequency, for the nominal cycle. There was a slight roll-off with frequency, at approximately 10%/decade over the range shown. This was commensurate with the roll-off seen in low field composite transfer functions (not shown). Bulk PZT 5H ceramic also exhibits a roll-off behavior, although smaller in size: ~5%/decade. The additional decrease may be due to a viscoelastic effect in the matrix material, or a decrease in field reaching the fiber from the change in electrical properties of the matrix and fiber as a function of frequency. This trend was not seen in the Uniform Fields model predictions of Chapter 4, but could be a smaller second order effect not captured by the material models.
The Representative Work Cycle results are summarized in Figure 6.8, where all composites are tested for the same cycle. The strains are shown as mean values, with the divisions representing the standard deviations above and below the mean. All of the composites have comparable Rep Cycles strains, in the range of ~900-1000 ppm, except for PZT 4 which is significantly lower. This is in contrast to the low field constants, where the PZT 4 composites had piezoelectric response similar to the softer ceramics. However, this was expected. As the scaled Representative Work Cycles in Chapter 4 showed, PZT 4 had a lower high field response than the softer ceramics. PZT 4 is a much harder piezoceramic, with lower nonlinearity in the strain response, and a much lower saturation strain than PZT 5H or 5A. Both of these contribute to the lowered Rep Cycle strain.

The nonlinear features of the high field response can be attributed to the ferroelectric nature of the material. The coercive field of the crystal dipoles are actually a distribution of values, with some switching far below the reported value. As the applied field is increased, more of the dipoles are reoriented by 180° and back with each cycle of applied field. This nonconservative action contributes to the material hysteresis, and also results in larger apparent strain from the volumetric changes associated with repolarization. As it will be shown, all of this increased strain is not available for useful work.

This nonlinearity is strain based, rather than electric field based. This was evidenced in Figure 6.3b. This figure showed the $d_{33}$ constant as a function of applied field, for a laminated and un laminated composite sample, normalized by the zero field value. The $d$ constant of the free composite rapidly increases with increasing field, while
the clamped composite $d_{33}$ is almost constant with applied field in comparison. The application of a high stress decreases the ease with which the dipoles can switch. In this case, the stress is compressive along the polarization direction, so that weaker dipoles would prefer to be oriented in the 90° directions rather than switch between the 180° positions. Note that the laminated result shown is for a highly clamped active composite. Since the nonlinearity is strain-based, less clamped actuators would result in a curve falling somewhere between the two shown. The large digression of these two curves hints that the laminated Rep Cycle strains will be far less than expected.

This was indeed the case. The results for the laminated Representative Work Cycle strains are summarized in Figure 6.9, shown as a percentage of the free Rep Cycle strains. Also shown is the low field residual strain ratio for the same composites, originally presented in section 6.3.3. This was taken as the ratio of laminated to free $d_{33}$ constants. The lamination to the two plies of E-glass fabric resulted in a reduction of Rep Cycle strain to between 34% and 43% of the un laminated values. Again, the PZT 4 exhibited a smaller decrease in actuation capability under load, attributed to the smaller degree of nonlinearity in the unclamped actuation. There was little difference in residual Rep Cycle strains in the softer PZT materials, whether circular or square. However, as anticipated, in all cases the residual strains were significantly less than the values expected based on the low field results. The discrepancy between the two ratios varied from 30% to 50%.
Figure 6.10 Comparison of the free and laminated performance for a typical baseline actuator (#75 shown), illustrating discrepancy between actual strain reduction and that predicted using the low field behavior (equivalent to laminated plate theory predictions).

The effect of the high strain nonlinearity on residual actuation is illustrated in Figure 6.10. This shows a comparison of the measured un laminated and laminated Rep Cycle strain for a single actuator (PZT 5H fibers), and the laminated Rep Cycle strain predicted using the low field cutdown factor. Clearly, this discrepancy is significant, and must be accounted for in the design of active structures. Since the discrepancy depends on the loading condition, it is important to characterize the actuators in a stress environment as close to the application as possible. There is no obvious method for extending the linear models for incorporating such effects. Instead, better material models that allow for strain-based actuation and repolarization effects are needed, and are being advanced at this time (Ghandi and Hagood, 1997).

6.4.2 Application - Discrete Flap Actuator

Flap actuation was introduced in section 5.3.2 as a method for controlling blade vibrations using a discrete trailing edge flap actuator. The application used IDEPFCs incorporated as the actuator mechanism in a laminated 10 ply bender configuration. The purpose of this application was to demonstrate the use of active composites in a laminated structure. Monolithic ceramics had been used in the past as the actuation, but IDEPFCs potentially offered some advantages in terms of weight distribution and performance.

The actuators used in the manufactured flap (Figure 5.7) were fully characterized prior to lamination in the bender. The average of all Representative Work Cycle results were used as an input to the model. Two types of tests were conducted on the flap actuator. Low field transfer functions were taken to determine the natural frequencies of the system. This was used to compare to models in order to assess modeling accuracy.
Table 6.7 Comparison of Discrete Flap Actuator Performance to Model (from Lutz, 1995; Ilvedson, 1995).

<table>
<thead>
<tr>
<th></th>
<th>Tip Deflection (1 Hz)</th>
<th>Natural Frequency</th>
<th>Efficiency</th>
</tr>
</thead>
<tbody>
<tr>
<td>Model</td>
<td>±7°</td>
<td>$f_r=81$ Hz</td>
<td>10-13%</td>
</tr>
<tr>
<td>Experiment</td>
<td>±15°</td>
<td>$f_r=64-77$ Hz</td>
<td>58-76%</td>
</tr>
</tbody>
</table>

High field testing was also conducted over a range of frequencies, to determine the maximum motion of the flap. This test was an evaluation of the high field capability of the laminated actuators and the total system performance.

Experimental results were compared to a simple two beam model, where each beam represented a five ply IDEPFC grouping (Ilvedson, 1995; Lutz, 1995). The model utilized two Bernoulli-Euler beams with the tip motion of each constrained by a compatibility condition that ensured the tips moved together. The stiffness of the foam core was ignored. The beams were assumed clamped at the base, and in-plane forces in the actuators caused by the compatibility condition were accounted for. Damping was ignored.

The transfer function tests were taken from voltage input to flap deflection output, measured with a diode laser. The results of the test showed a first natural frequency on the order of 81 Hz, which was close to the range of predicted frequency (Table 6.7). This indicated that the system was accurately modeled in terms of stiffness and mass distribution. However, the non-resonant flap deflection was on the order of 50% of the model predicted deflection. There were several possible explanations for this cut-down. At least 20% of this reduction was expected from the strain associated with the high field nonlinearity, as described in the last section. Another 6% may be accounted for by the fact that the actuator was tested at 2800 Vpp, rather than the standard 3000 Vpp Rep cycle (which was used as the model input). Other sources of error may have been introduced by the model, the bonding among actuators, and the linkage to the flap.

Perhaps the largest contribution to the discrepancy was due to the compressive field depolarization which occurred during testing near resonance. After testing, the peak-to-peak strains were reduced to half the original value. Although the actuator groupings were re-poled, this was done at room temperature. Chapter 2 showed that only about 80% of the full poling is achieved for these conditions.

In summary, this application showed the importance of accurate modeling of the system in order to perform effective design. The system efficiency was reduced to 10-13%, due to the squared relationship with output deflection. However, the results still showed comparable actuation energy density with monolithic piezoceramics, due to the
lower mass density. Overall, this application demonstrated the use of IDEPFC actuators in a stacked configuration for high field actuation, and illustrated many of the key issues to operation and lamination.

6.4.3 Application - Integral Blade Actuator:

The integral blade model was an application that incorporated IDEPFC actuators into the spar of a model helicopter blade to demonstrate the main spar twist concept for individual blade control (IBC). This application was introduced in section 5.3.2 for the actuator manufacturing relevance, and the modeling and testing is discussed in detail in (duPlessis, 1996). The purpose of the experiment was to verify box beam models developed to predict the response of anisotropic composite structures to induced twist, and validate the use of IDEPFC actuators in such applications.

Although the passive materials used in the experiment were well characterized, the ±45° actuators were not directly characterized. The actuators were close to the nominal actuator materials that have been well characterized, with the exception that the matrix contained 75 wt% PZT filler\(^5\). It was assumed that the stiffness remained unchanged, and only the free strain would be increased, due to the higher matrix dielectric. The Uniform Fields model was used to predict the composite low field \(d\) constants for this matrix material. The representative work cycles for the nominal actuators (60 wt% PZT) were multiplied by the ratio of the \(d\) constants to arrive at a prediction of the 75% PZT Rep Cycle, estimated to be 1000 ppm strain. This was then divided by the voltage for this cycle (3 kV) to determine a linearized high field \(d\) constant. This was input into the structural model.

Passive box beams were manufactured and tested to calibrate the structural models, and to validate the test setup. The test setup involved clamping the blade at the base, and measuring the tip twist. Quasi-static methods were used to measure the tip twist of the blade from a voltage applied to the ±45° actuators embedded in the blade. A 1 Hz sinusoidal voltage was applied to the actuators, with a DC offset to maintain positive fields only (primarily for an extra measure of safety against depoling). The tip twist was measured using two methods. The first measured twist directly using an indicator fixed to the tip along the chord direction, which indicated the twist on a protractor. The second used two lasers to measure the differential displacement of two points on the tip, from which the twist could be easily calculated.

\(^5\) These actuators had not yet been manufactured in the baseline series.
**Figure 6.11** Results for 1/16th scale model blade, showing tip twist as a function of applied voltage to ±45° IDEPFC actuators. Taken from (du Plessis, 1996).

The results are shown in Figure 6.11 for two separate tests. The first test showed the response prior to electrical damage, while the second was taken after multiple damage sites were repaired. There was a close match to the model for the first experiment. Almost 1.4° of twist at the tip was achieved for the 2000 volts applied. A lower level of actuation was expected, due to the nonlinear free strain effects discussed in section 6.4.1. It is possible that the actuator capabilities were underestimated, or inaccuracies might exist in modeling the geometry of the spar. The second test showed lower response due to the incurred damage. Five damage sites were repaired after pushing the actuator past the 2000 volt level in the first test, using the ‘drill and fill’ method described earlier. Although this significantly reduced the capability (~25%), it demonstrates the robustness of the material.

Overall, the application showed the possibility of integral blade actuation for controlling helicopter vibrations, based on the measured deflections. More specifically, it showed the possibilities of using IDEPFC actuators for highly integrated actuation that is robust to damage. Higher twist levels are possible with increased coverage and better placement of the active plies, in order to match the initial specifications set out for Individual Blade Control. Extension of the models to full sized helicopter blades (Derham and Hagood, 1996) has shown that the use of IDEPFC actuators will permit increased cruising speeds (+12%) and achieve significant vibration reduction (65% in forward flight), with only a modest increase in power (2.5%) and cost. These attributes alone demonstrate the merit of pursuing this technology for future applications in this area.
6.5 Summary

This chapter presented the experimental results for the active fiber composite actuators, representing closure in the design loop process. Details of the composite microstructure provided insight into the effects of fiber geometry and filler loading on composite performance. Prepole tests for the remnant polarization and coercive field demonstrated the inability of the linear Uniform Fields model to predict high field phenomenon, which is largely dominated by the localized behavior near the electrodes. However, low field composite properties were well captured by the Uniform Fields method. Both the trends and magnitudes of effective properties were predicted by the model for samples with varying ceramic fraction, and for different combinations of fiber materials and matrix composition. PZT fillers added to the matrix did show an improvement in composite response as predicted, whereas the carbon black fillers did not improve the composite response. This was attributed to the random effectiveness of the conductive filler, which required percolation in order to be effective at low loading fractions. Comparisons of properties and performance with different PZT fiber materials indicated that while the hard ceramics may maximize the low field response, the soft ceramics provide higher actuation capability at high fields. Finally, two applications demonstrated the effective use of active fiber composites in the control of laminated structures. These applications showed that the active composites could be successfully operate in a number of modes, laminated in bender configuration (like monolithic wafers), or used as shear mode actuators, a characteristic of anisotropic actuators.
Chapter 7

Conclusions and Recommendations

7.1 Summary

This thesis was directed at the development of active fiber composites for structural actuation. Active control of structures is a rapidly growing field that incorporates actuators and sensors into an otherwise ‘passive’ structure for the purpose of improving performance. Performance can take a number of forms, depending on the particular application, but usually focuses on the attenuation of structural vibrations. A reduction in vibration levels can result in longer structural lifetimes for aircraft wings and helicopter blades, extended operational envelopes in high performance aircraft, and signature reduction in acoustic control of stealthy aircraft and submersibles.

Aerospace structures place a number of restrictions on the active materials used in these environments. These include high actuation energy density, conformability to curved surfaces, large area coverage, high strength, and directional anisotropy to name a few. The current selection of active materials is not capable of meeting all of these demands, which are often considered mutually exclusive. The active material most commonly used is piezoceramics, but these materials suffer from a number of drawbacks related to reliability and large scale integration. Piezoelectric Fiber Composites (PFCs) with uniform electrodes were introduced a number of years ago in an attempt to overcome the weaknesses in monolithic ceramic technology. These new materials offered higher strength and conformability, potential for large area coverage, and directional actuation. However, the maximum performance of these materials suffered from the mismatch in the dielectric of the bulk constituents. A new material was needed that combined the best attributes of available actuation materials: a material that had the structural nature of composites, yet had actuation performance that equaled or surpassed current piezoceramics. This thesis was motivated by this challenge.

This challenge was successfully met in the development of the current active fiber composites, known here as interdigitated electrode piezoelectric fiber composites
The concept was a fusion of piezoelectric fiber composites and a performance enhancement technology developed for monolithic ceramics: the interdigital electrode. The interdigitated electrode pattern greatly enhanced the performance of active fiber composites. This arrangement increased the effective ceramic volume fraction along the electric field pathlength, partially reducing the difficulty with dielectric mismatch. In addition, the primary axis of piezoelectricity was now oriented along the fiber direction, greatly increasing the maximum strain. This also improved the anisotropy, causing the planar directions to actuate out of phase for improved modal sensing and control. Finally, since the interdigitated electrode pattern was etched onto a separate interlaminar layer, this enabled the composite to remain functional in the presence of damage to the ceramic.

The major goal of this work was to develop this concept into a viable technology for real applications. The realization of this goal depended on a number of tasks, which formed the objectives for this thesis. These goals, and the structure of this thesis, were organized in a hierarchy that followed the natural evolution of a structure from the most basic building blocks to the built-up active structure:

1. Develop methods for characterizing constituent materials for use in structural actuation and active composites systems
2. Advance the field of composite modeling for coupled field materials
3. Identify the key issues and tradeoffs in the design of composite actuators
4. Develop the ability to manufacture and characterize active composites
5. Demonstrate the capability of active composites in structural control of laminated structures

The ultimate objective was to eventually enable highly distributed actuation and sensing for structural control applications.

The first objective was investigated in chapter 2. This chapter dealt with fundamental issues of the bulk materials, and understanding how the choice of these materials affected the overall composite behavior and ease of manufacturing. The first part of the chapter characterized the ceramic material. A family of bulk ceramics, ranging from piezoelectrically soft to very hard, were characterized and compared using new metrics particular to the needs of structural actuation. One such metric was the introduction of the Representative Work Cycle. The soft ceramics, such as PZT 5H and 5A, exhibited better actuation capability at low and high field regimes of operation, while the hard ceramics were better suited for the high stress environments that might be encountered in structural applications. PZT 5H was chosen for further characterization at this point for its high electro-mechanical coupling, and ease of poling. Poling studies of PZT 5H determined the
necessary conditions for full poling, as a function of poling temperature, electric field, and time. The required electric field could be related to the composite through a common metric known as the coercive field. An aging study identified the aging trend for PZT 5H, showing it to be linear with decades in time, and substantiated the use of a 24 hour aging time found in most references. Finally, a method was developed for characterization of individual PZT fibers, allowing measurement of the maximum strain and coercive field. This demonstrated that the extruded fibers had substantially degraded performance, compared to bulk materials. This information was used as inputs to the models of Chapter 3.

The second half of Chapter 2 characterized the matrix properties as a function of conductive (carbon black) and dielectric (PZT) fillers in the system. This focused on the matrix properties as they relate to the ability to manufacture the composites, and the effects on the electrical properties which influence the composite performance. A rheology study was conducted that demonstrated the substantial change of viscosity with added fillers. With elevated temperatures, the viscosity of some matrix compositions was shown to decrease to levels that were acceptable for composite processing. To fully characterize the effect of the matrix on performance of the composite, a three electrical parameter material model was developed. The three parameters (dielectric, resistivity, and dissipation) were determined for the matrix as a function of the fillers. This was used later with the models developed in Chapter 3 to determine the composite response as a function of frequency for poling and operational issues. The random nature of the percolative chain formation with carbon black fillers became evident from the wide range of values measured for the electrical properties. This would be demonstrated again later in the composites. Finally, dielectric breakdown tests were conducted on the matrix samples. Samples with purely conductive fillers showed a rapid drop in dielectric strength at very low concentrations (2-3% by weight), indicating a percolation phenomenon. The dielectric fillers also reduced the dielectric strength, but for much higher concentrations (60-75% by weight), and at a more moderate rate. Introduction of the dielectric filler into the conductively loaded systems showed a stabilizing effect on the breakdown of the samples.

The second objective was examined in Chapter 3, with the modeling of active fiber composites. Review of previous work showed little advancement in the area of micro-mechanical modeling for coupled field materials in composites. Two models were applied to this problem. The first used a closed form approach that modeled the approximate geometry based on combining rules. This technique, called the Uniform Fields method, was a generalization of the mechanics of materials approach, but extended to three dimensions and to incorporate electromechanical coupling. This method had also been
used in the original PFC problem, but the novel geometry created by the interdigitated electrode required more careful consideration of field compatibility between the materials. This model predicted the composite effective properties by assuming uniform electrical and mechanical fields within each material. Due to its closed form nature, this model could provide physical insight into the governing material and geometrical terms that dominated the composite response. It was also extended to accommodate the complex electrical behavior of the three parameter material model.

The second model employed the finite element approach to predict the effective properties using a commercial program. This technique applied a series of hypothetical load cases and boundary conditions, and determined the response of the discretized model. The two model approaches were compared as a function of ceramic volume fraction, and for different families of materials that covered the ranges of interest in this problem. Excellent agreement was seen throughout the range of materials and ceramic fraction. Although a portion of agreement can be attributed to the similar assumptions and boundary conditions imposed on the representative element used for modeling, it demonstrated the effectiveness of the simpler, closed form technique. The agreement showed that field uniformity is a good assumption, and effective composite properties were adequately modeled when considered in an average sense. This validated the use of the Uniform Fields model for design, where it has far greater utility than a finite element model due to its adaptability and ease of use.

The third objective involved identifying the key issues in design of active fiber composites, and was considered in Chapter 4. This objective was accomplished by examining the change in composite properties with different material combinations in order to map a design space for composite selection. This portion of the work combined the Uniform Fields model developed in Chapter 3 with the material characterization of Chapter 2. The use of the model permitted predictions for the effectiveness of the different bulk ceramics in composite form. Surprisingly, hard ceramics such as PZT 4 were shown to be most effective in low field regimes, due to the lower ceramic dielectric which more than offset the lower electro-mechanical coupling. In an attempt to extend the linear model to the prediction of high field performance, the concept of a scaled Representative Work Cycle was introduced. This demonstrated that soft ceramics provided the largest high field response, coupled to their increased dipole mobility which results in higher nonlinearity and saturation strains.

The design choices in matrix materials involved both performance and reliability issues. Conductive fillers were predicted to provide the largest benefit to the performance of active fiber composites. The addition of the high dielectric particles (PZT) was also
predicted to increase performance, but at the cost of increased composite mass. It was expected that the introduction of dielectric fillers would also increase the composite thickness, which had a strong impact on performance. In such a case, the dielectric fillers would not be beneficial to the composite. Reliability studies were conducted using a combination of the matrix dielectric strength data from Chapter 2, and predictions of the maximum field concentrations in the matrix between the electrode and fiber. These studies predicted the relative reliability of different designs based on filler fractions using a Risk Factor. This Risk Factor illustrated that composites with pure conductively filled matrix was at high risk for breakdown, and the inclusion of dielectric fillers substantially reduced this risk.

A final design study examined the role of fillers in modifying the frequency response of the composite actuators. For the range of composite geometry in this work, it was found that the high material mismatches in all cases limited any useful behavior to outside the frequency range of interest, approximately 1 Hz to 10 kHz for typical vibration suppression. The Uniform Fields model augmented with the three parameter material model illustrated a transition from DC to AC behavior that occurred well below 1 Hz. Accompanying this transition was a sharp reduction in apparent dielectric and field in the fiber, an increase in composite stiffness, and a peak in the material loss factor. Although this indicated that damping applications would require external electrical components, it also ensured that full poling would be achieved under the conditions prescribed. The culmination of this third objective was a set of selected designs for experimental verification. These designs included varying fiber geometry and ceramic type, and designs for different matrix compositions based on the performance and reliability factors.

The fourth objective of this thesis, to develop the capability to manufacture and characterize active fiber composites, was brought together in Chapters 5 and 6. Chapter 5 focused on the techniques developed for repeatable batch manufacturing of composites, which allowed evaluation of the different materials and geometry. The key requirements for the production of successful composite actuators were outlined, including factors that affect both the performance (such as compaction and electrode alignment) and reliability (voids). This lead to a step-by-step procedure for manufacturing that borrowed concepts from the manufacture of graphite/epoxy composites, and included modifications for incorporation of the electrodes. These techniques were developed to allow a minimum of changes for adapting to different materials or ceramic concentrations.

Chapter 6 focused on the techniques for characterization of the composites, and a comparison of the measured properties to the models of Chapter 3. A crucial part of the characterization of the composites was an examination of the resulting composite geometry.
and microstructure. Microphotographs demonstrated the differences in a variety of fiber types (circular and square) and matrix compositions (fillers and no fillers). Square fibers showed a reduction in composite compaction due to the decreased area for matrix flow and aspect ratio of the so-called square fibers. Added fillers also increased the composite thickness. Both of these would later show a reduction in possible performance. Other preparations prior to poling the materials included determining the coercive field, which was used to set the poling conditions based on the poling study of Chapter 2. Measurement of the coercive field showed levels far below those expected, based on Uniform Fields predictions of the field in the fiber. This indicated that some portions of the fiber were experiencing dipole realignment before others. This was most apparent in the circular fibers, where proximity of the ceramic to the electrode depended on the location along the perimeter of the fiber. However, this phenomenon was also seen in the square fibers, evidence of an advancing repolarization along the fiber and local field distortions near the electrode. These effects were seen in the very rounded repolarization shape of the butterfly and hysteresis curves.

Low field characterization of the composite piezoelectric free strain properties were accomplished using developed quasi-static methods, which measured the strain for applied voltage. This was extended to the induced stress constants by bonding the composites to materials of known stiffness, and using laminated plate theory to determine the values. Results for varying ceramic fraction showed very good correlation with the Uniform Fields model. Composite dielectric, piezoelectric free strain, and piezoelectric induced stress all showed good agreement. Although the stiffness was not directly determined, full fiber stiffness was validated implicitly by the agreement with induced stress constants. A comparison of different fiber types (PZT 5H, 5A, and 4) also demonstrated good agreement with the model for piezoelectric free strain and induced stress. A direct comparison between fiber capabilities based on free strain was difficult due to the different geometry, but the clamped actuation tests highlighted differences in ceramic composition. As predicted, the harder ceramic (PZT 4) exhibited a higher level of residual actuation after clamping. High field characterization of the free composites showed higher free strain performance of the soft ceramics (PZT 5A, PZT 5H), as suggested by the extension of the linear model predictions based on the scaled Representative Work Cycles. However, the inability to effectively model high field performance was made strongly apparent in the high field laminated testing results. In all cases, the strain-based nonlinearity inherent in piezoceramics reduced the residual actuation otherwise predicted by linear theory, particularly for the softer ceramics.
Composites with varying matrix compositions were also characterized to compare the effects of fillers on performance and reliability. Composites with PZT fillers were thicker than those without, reducing the benefits of increased dielectric. Composites with pure conductive fillers did not survive the pre-pole testing, but experienced dielectric breakdown as predicted for these high risk designs. Composites with a mixture of conductive and dielectric fillers were more stable, and generally survived the high fields. However, these composites had performance similar to composites without conductive fillers, exhibiting no benefit from the apparent increase in matrix dielectric. This was not surprising, for a number of reasons. First, similar results were found for the matrix samples of Chapter 2, which varied from no benefit, to a doubling of dielectric over the PZT filled matrix. This lack of increased performance could also be attributed to the high tensile pre-stress developed in the composite matrix upon cooling from cure, which removed the dominant percolative effect of the conductive fillers. Works by other researchers have shown that large reductions in conductivity can accompany tensile strains in carbon black polymer composites. This percolative chain formation is relied upon for increases of dielectric at such low filler levels, but this same chain formation causes rapid decreases in dielectric strength.

The fifth and final objective of the thesis was the demonstration of active composites in laminated structures. Active composites were successfully laminated to a variety of host materials, including structural fiberglass, glass, and metal substrates. A number of integration issues were identified when using these laminates. Poling was always done prior to lamination due to the high temperature and oil environment needed, and the more difficult dipole orientation when under compressive stress. An operational issue is caused by the interdigitated electrode, which does not provide one side of the actuator with a ground potential, as is encountered with monolithic ceramics. Instead, the high potential and ground electrodes are distributed across both surfaces, requiring additional care with electrical isolation. Operation at high electric field caused shorting to any adjacent conductive materials, and capacitive coupling to mounted strain gages was evident for even low frequencies (>10 Hz). Thus, a thicker insulating layer is required in these cases than what was provided by the Kapton interlaminar electrode.

Active fiber composites were applied to two applications for rotorblade control. These applications provided additional lessons associated with the preparation and operation of active composites for real structures. The first of these was a demonstration of active composites to a trailing edge flap actuator. Although the performance was not as large as expected, this discrepancy was explained by consideration of several factors, including the strain-based nonlinearity and the occurrence of compressive depolarization of
the actuators in service. This demonstrated the difficulty of using soft ceramics in structural applications. Other issues that were examined included lamination and operation of active composites in a stacked bender configuration, including efficient operation for in-phase drive with biased voltage cycles. The second laminated structure was the application of active composites to integral twist of a 1/16th composite rotorblade. This validated the manufacture and use of fully integrated 45° active plies in a structure, used to induce shear stress in the spar structure to twist the blade. This application also demonstrated the successful use of damage repair techniques to active composites. In the duration of testing, electrical damage sites were repaired by drilling out the damage and filling the holes with epoxy, showing the robustness of the composite material. In both cases, active fiber composites represented several advantages over monolithic piezoceramics. In addition to robustness and conformability issues, the lower density material allowed better weight distribution and showed the potential for highly integrated, large area coverage active materials.

7.2 Conclusions and Contributions

Conclusions

Material Characterization

Characterization was a critical element in the development of active fiber composites. Accurate modeling and design was not possible based on constitutive relations reported in the literature alone. The novel combination of active and passive materials in this work necessitated a full understanding of the electrical, mechanical, and piezoelectric behavior of all materials involved. For example, characterization of the fiber materials determined a two third reduction of the reported bulk properties in materials that were originally assumed to be equivalent to bulk. Decisions on manufacturing, design, performance, and reliability all depended on an in-depth knowledge of the properties. Both bulk and composite materials needed to be characterized at field levels comparable to the operational environment. Linear low field characterization is usually the only characterization performed on active materials, but does not capture factors associated with dipole mobility that affects the degree of nonlinearity and level of saturation strain. In this case, this would result in incorrect conclusions about the capability of different designs.

Fiber Material

Selection of the PZT ceramic type and fiber geometry was shown to depend on the application. For relatively low field applications, a hard ceramic in the PZT 4 family (such
as 4S) could provide high composite electromechanical coupling and high stress capability, including the ability to handle compressive stress. For high field, but low stress applications, PZT 5A could provide the maximum actuation capability. If high stress is expected, the lower actuation of PZT 4S must be accepted for the added reliability. Square fibers offered no obvious benefits over round fibers, except for a slightly higher mass efficiency and more reliable properties in the current configuration. The effect of fiber cross section on composite strength was not known.

**Fillers**

The use of filler materials was not beneficial to the performance of active fiber composites, in the ranges of concentrations studied. The increase in matrix dielectric realized by using dielectric fillers was largely offset by the lower composite compaction. Mass normalization of the performance further reduced the apparent improvements due to the high percentages of lead-based filler needed. There is a possibility, however, that increased pressure may have further increased the compaction. Conductive fillers were only viable when used in conjunction with the stabilizing effects of dielectric fillers, but no added improvement was gained over the pure dielectric fillers. The introduction of fillers may only be beneficial if it affects the strength considerations through toughening mechanisms, or decreasing the composite sensitivity to manufacturing inconsistencies such as compaction.

**Linear Models**

Linear models were well suited to modeling the low field averaged behavior of active composites, but were inadequate at capturing high field response, or behavior caused by distortions in the uniformity of the electric field. Both the Uniform Fields and Finite Element based models were accurate when determining the constitutive properties of IDEPFCs, which considered only the average macroscopic response of the material. Good agreement was seen between models, and with experimental data. High field behavior, characterized by strain based nonlinear increases in free strain, could not be accounted for. Accurate representation in this regime was critical for design. Local field behavior near the electrodes was not captured by the Uniform Fields model, but could be captured by the Finite Element model. However, these field distortions had little effect on the behavior at low field, but would be important for accurate prediction of high field characteristics such as composite coercive field and saturation strain, which could not be captured by a linear finite element in any case.
Actuator Performance

Active Fiber Composites represented an increase in performance over currently available actuator materials. These materials provided two to three times the actuation energy density of its closest competitor, monolithic piezoceramics. Although the composite was lower in stiffness, this was more than offset by the lower weight and higher maximum strains. The bandwidth of the material indicated material applicability to a range of actuation and sensing applications. The highly directional nature of the composite was evidenced in both predicted and experimental response, which will permit anisotropic actuation at levels not previously possible.

Structural Applications

Active Fiber Composites could effectively perform the actuator role in real structural actuation applications. Two rotorblade applications demonstrated the capability of active composites, both in terms of performance, and in integration. IDEPFCs performed as predicted when used as integral twist actuators, and were easily integrated within passive structural materials. Electrical damage could be effectively repaired and materials returned to service. Composites were also used in the more traditional monolithic ceramic modes of operation, as laminated and operated in bender configuration.

Contributions

The general contribution of this thesis was the development of a new actuator/sensor material for use in the actuation structures. These composites represent an advancement in the current state of the art of commercially available materials. The lower density and higher actuation capability doubles the energy density over its closest competitor, monolithic piezoceramics. With the introduction of the active material into composite form, a number of other benefits were now possible, previously unrealized in monolithic technology. Highly directional actuation permits induced shear stresses for integral twist of structures without relying on host material anisotropy. The separate interlaminar layer allows a retention of actuation properties even in the presence of ceramic damage. The polymer matrix provides a highly conformable and robust material through load transfer mechanisms. Perhaps the most important contribution of the composite material, however, is the new possibility of a continuous highly integrated active material, which could conceivably cover entire structures. This level of coverage must be realized to achieve the performance of the majority of upcoming applications.

Other contributions, specific to the individual objectives of this thesis, are discussed below:
Characterization Methodologies

This thesis developed a number of methods and metrics for characterization of active materials, particular to the field of structural actuation. A method for testing individual 100 micron diameter fibers was devised and used to determine the properties in relation to expected bulk values. Quasi-static methods were developed for the determination of the electromechanical properties of bulk ceramics, and for composites in free and laminated operation. Metrics for high field characterization, such as the Representative Work Cycle and Representative energy density, were introduced in order to permit comparison of materials in the regime where operation actually occurs. Methods for determining poling conditions and aging requirements of bulk ceramics were advanced to ensure maximal properties and accurate results in composites. Key matrix properties were also identified and methods developed to determine a range of characteristics, including rheology, electrical properties, and dielectric breakdown. Many of these methods are of general importance for the field of structural actuation, and all can be applied to characterization for development of other combinations of active and passive materials.

Modeling of Coupled Field Composites

This thesis advanced an approach to modeling multi-phase coupled materials that accurately predicts the linear effective constitutive relations. The Uniform Fields method represents a simple approach for combining active and passive materials that provides insightful prediction of composite response. This type of model demonstrated its utility during the design stage of this thesis, where more cumbersome methods such as FEM represent a much higher cost. This method was able to model the complex geometry of the current composite with interdigitial electrodes, and can easily be adapted for other composite types such as particulate and rod composites. The model was validated using comparisons with a finite element model, and with experimental data.

Design Methods

This thesis developed a systematic approach to the design of active fiber composites. The approach looked at how the choice of fiber materials, matrix composition, and geometry affected the important output metrics, such as viscosity, induced stress, or operational risk. Inter-relationships between design variables lead to general conclusions on ‘best’ designs for active fiber composites. Although new materials will become available and the best composite design may change, the approach and important issues identified will remain. This approach can also be applied to other combinations of active and passive components, wherever tradeoffs in material selection affect the processing, performance, and reliability.
Method for Manufacturing

This work developed a method for manufacturing composites that incorporated electroceramic fibers, thermosetting resins with various fillers, and interlaminar electrodes. The process developed was repeatable and uniform, allowing comparisons for different constituent materials and composite geometry. This permitted evaluations of different designs. This technique was also successful in that it produced composite actuators with properties that were superior to the best commercial actuators available. These actuators were used in a number of proof-of-concept designs for structural actuation, allowing work to advance in areas of aeroelastic design of rotorblade structures.

7.3 Recommendations for Future Work

Fiber Materials

Although only a few piezoceramic compositions were available for use as fibers in this work, they represented the important spectrum of possible types. However, the present work in fine diameter piezoceramic fibers may soon introduce alternatives to the current composite geometry. Fibers on the order of 5-10 micron will permit composites of multiple fibers across the thickness. This will introduce a host of new issues. It is unclear whether the performance will be degraded due to the increased number of matrix gaps among fibers, or will be offset by the higher maximum volume fractions. Small fibers are worth pursuing for issues of potentially increased fiber strength, which will increase composite strength both directly, and indirectly from the increased number of alternate load paths. The increased strength may also result in some form of automation in manufacturing, using automated placement of individual fibers or tows, or methods for weaving. To investigate these possibilities, the Uniform Fields model may be adapted, or new models may need to be generated if the composite performance is more dependent on the local behavior near the electrodes. This may be the case, as the field may be much less uniform due to the distribution of fiber and matrix as a function of field path length.

Matrix Materials

This thesis explored the use of fillers on the performance and manufacturability of active fiber composites, but did not deal with different polymers for the base matrix material. The base resin chosen for this work was carefully selected for processing and structural reasons, and probably represented the best choice of thermoset resins. However, thermoplastic materials may also offer some significant benefits. One particular material is PVDF - polyvinyl difluoride, a piezoelectric polymer. It offers an intrinsically higher dielectric (~8 for random processing) than the undoped thermoset resins, and may not be as
susceptible to voids during processing. Although the manufacturing would need to change to accommodate the new matrix, these changes could be small. The added benefits might include higher performance and potential for large scale forming and manufacturing.

**Nonlinear Models**

Development of a nonlinear model for the response of active fiber composites would be a great benefit to this work. This could be based on nonlinear finite elements or an energy based approach coupled with accurate material models. This need arose from the inability to predict high field behavior, which is key to design of structural actuation systems. A prime example existed in the flap actuator application, where goals based on impedance matched design were not met, and combinations of high field and high stress resulted actuator depolarization during use. A priori prediction of such behavior would be beneficial. The desired model would be capable of predicting repolarization and actuation on the basis of electric field and stress conditions, that vary as a function of material location according to the governing continuum equations.

**Strength and Stiffness**

Issues related to the strength and stiffness of active fiber composites needs to be addressed. This thesis dealt primarily with optimization of the actuation capability, without regard to the resulting strength. Even its present form, however, IDEPFCs are significantly more robust than monolithic ceramics. Although the ultimate tensile strength will be on the same order as the monolithic ceramic (because the fibers dominate the ultimate strength), the material is conformable and resistant to damage. Cracks in the ceramic can be tolerated by virtue of the load transfer capability provided by the matrix. The ultimate goal is replacement of structural plies with active structural plies, so that the active material serves a dual role. Before this is possible, more work needs to be done on evaluating the effects of reinforcing materials on the actuation, strength, and fatigue properties of IDEPFCs.
References


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Appendices

A. Piezoelectric Constitutive Relations

Standard forms for the linear piezoelectric constitutive relations are set by the IEEE, given in (IEEE Std-1986). These forms relate the dependent mechanical and electrical fields to their conjugate independent field variables. Since there are four field variables, four forms of the constitutive relations exist. The two most commonly used for actuation are ones that set the applied electric field $E$ as an input, or independent, variable:

$$\begin{bmatrix} D \\ S \end{bmatrix} = \begin{bmatrix} \varepsilon^T & d \\ d^T & s^E \end{bmatrix} \begin{bmatrix} E \\ T \end{bmatrix}$$

(\text{A.1})

or,

$$\begin{bmatrix} D \\ T \end{bmatrix} = \begin{bmatrix} \varepsilon^S & e \\ -e^T & \varepsilon^E \end{bmatrix} \begin{bmatrix} E \\ S \end{bmatrix}$$

(\text{A.2})

where $D$ is the electrical displacement (Coulombs/m$^2$), $S$ the strain, $E$ the electric field (kV/cm), and $T$ the stress (Mpa). Subscript ‘t’ denotes a matrix transpose.

The first of these relations gives a description of the strain as a function of applied electric field and stress. These complementary electrical and mechanical fields are related through the material properties: the dielectric ($\varepsilon^T$), the induced strain constant ($d$), and the compliance ($s^E$). The superscripts are a stipulation on the boundary conditions, which are important to specify for coupled field materials. Superscript $T$ represents a constant stress condition, often taken as unclamped or free mechanical boundary conditions. Superscript $E$ represents a constant electric field condition, satisfied with short circuited electrodes.

The second commonly used form (A.2) describes the induced piezoelectric stress as a function of the applied electric field and mechanical strains. For this relational form, the material constants are: $\varepsilon^S$, the clamped dielectric, $\varepsilon^E$, the short circuit stiffness, and $e$, the induced stress. Superscript $S$ refers to fully clamped (zero strain, or constant strain) conditions. Relationships between the material constants are found by simple manipulation of the constitutive relations. Taking, for example, the first row of the matrix expansion of Equation (A.1), and rewriting to express stress as a function of strain and electric field,
\[ T = (s^E)^{-1} S - (s^E)^{-1} d_i E \]  \hspace{1cm} (A.3)

If (A.3) is substituted into the second row of the matrix expansion of A.1, the result is

\[ D = d(s^E)^{-1} S - d(s^E)^{-1} d_i E + \varepsilon^T E \]  \hspace{1cm} (A.4)

Comparing Equations (A.3) and (A.4) to the second form of the relations in (A.2), the following is apparent:

\[ c^E = (s^E)^{-1} \quad e = dc^E \quad \varepsilon^S = \varepsilon^T - dc^E d_i \]  \hspace{1cm} (A.5)

The first relationship demonstrates that the short circuit stiffness and short circuit compliance are simply matrix inverses. The second demonstrates that the piezoelectric induced stress (e), often used to describe *actuator authority*, is a product of the free strain capability of the material (d) and the stiffness (c^E). The last relationship demonstrates that the clamped dielectric is smaller than the free stress dielectric. Other relationships can be found by comparisons with the other forms of the constitutive relations, and are summarized in the IEEE Standards (IEEE, 1978).

The above notation is a convenient representation of the full forms of the matrix equations. Using Voight notation, it is possible to write the full constitutive relations in a 9 by 9 array of material properties. Several simplifications can be made. Since the electric field variables are first order tensors, the dielectric block matrix is diagonal. The mechanical fields are second order tensors, but the act of poling causes the material to be transversely isotropic about the ‘3’ axis. This results in only five independent stiffness terms (s_{66}^E can be written in terms of s_{11}^E and s_{12}^E), and an absence of coupling between shear and normal directions. Piezoelectrically, stresses are only developed in the normal directions by a field applied in the direction of poling. In the unusual case that different electrodes for poling and actuation are used, shear can be induced. For the first form of the constitutive realtions, the fully expanded equations for linear bulk piezoceramics are,

\[
\begin{bmatrix}
D_1 \\
D_2 \\
D_3 \\
S_1 \\
S_2 \\
S_3 \\
S_4 \\
S_5 \\
S_6 \\
\end{bmatrix} \begin{bmatrix}
\varepsilon_{11}^T \\
\varepsilon_{11}^T \\
\varepsilon_{33}^T \\
\cdot \\
\cdot \\
\cdot \\
\cdot \\
\cdot \\
\cdot \\
\end{bmatrix} = \begin{bmatrix}
d_{15} \\
d_{15} \\
d_{31} \\
\cdot \\
\cdot \\
\cdot \\
\cdot \\
\cdot \\
\cdot \\
\end{bmatrix} \begin{bmatrix}
E_1 \\
E_2 \\
E_3 \\
\cdot \\
\cdot \\
\cdot \\
\cdot \\
\cdot \\
\cdot \\
\end{bmatrix}
\]

\hspace{1cm} (A.6)
For the *Uniform Fields* modeling approach, described in section 3.3, the uncoupled shear problem is ignored, as are electric fields not in the direction of poling. This results in only needing to consider a 4x4 block of material properties (see Equation 3.1). However, a higher degree of orthotropy is assumed in this case, to account for the different combination cases.

For use of the material (both bulk and composite) as planar actuators, the assumptions of plane stress are applied. Using the Kirchoff assumption in simple plate and shell theory (Jones, 1975), the out-of-plane normal and shear stresses are zero,

$$T_3 = T_4 = T_5 = 0$$  \hspace{1cm} (A.7)

Applying the assumption of plane stress to the above constitutive relations (A.6) simply requires removing the rows and columns associated with these stresses. The corresponding strains may be nonzero, but are functions only of the in-plane variables, and can be solved for separately. If other forms of the constitutive relations are needed (such as that given by A.2), the plane stress conditions must first be enforced in forms where stress is an *independent* variable, before manipulating to determine the other forms. Thus, although the 3-D induced strain ($d_{ij}$) and compliance ($s_{ij}^E$) constants are equal to their plane stress counterparts, the stiffness ($e_{ij}^E$) and induced stress ($e_{ij}$) are not.

### B. Error Analysis for Experimental Data

Simple error analysis has been performed for the experimental data presented throughout this thesis, following methods outlined in the following reference (Beckwith and Marangoni, 1990). For the most part, the data contained herein is not a large sampling of the existing population, and thus is not subject to analysis by statistical methods (i.e. standard deviations and confidence levels). A few exceptions are the sampling of fiber properties from a particular batch (including diameter, coercive field, etc), and composites where a number were manufactured with the same exact materials and geometry. Generally, however, the data is single sample data, and it is important to quantify the maximum level of *bias or systematic* errors inherent in the measurement techniques.

Bias errors are ‘the difference between the average of the total population and the true value’, and can be caused by a number of problems (Beckwith and Marangoni, 1990):

- calibration errors
- consistently recurring human error
- errors of technique
- limitations of system resolution
• ‘loading’ errors

Values for the various uncertainties come from several sources. For equipment, such as capacitance meters and viscometers, basic accuracies for measurements are taken from manuals or calibration certificates. Additional uncertainty is estimated from ranges of values seen in multiple measures of the same quantity, or changes in measurements with environment (such as capacitance with temperature). Other errors cannot be accounted for, such as variations in bulk material properties, material processing, or nonlinear errors such as the effect of d constant errors on the slope which defines the low field d value.

The majority of reported data depends on more than one measurement, so that the overall error estimate depends on a propagation of uncertainty. In these cases, the maximum error can be determined from

\[ u_f \leq \left| u_1 \frac{\partial f}{\partial x_1} \right| + \left| u_2 \frac{\partial f}{\partial x_2} \right| + K \left| u_n \frac{\partial f}{\partial x_n} \right| \quad (B.1) \]

where \( f \) is the function describing the overall measured quantity, \( u_f \) is the total uncertainty in \( f \) from the individual uncertainties \( u_i \), and the partial derivatives of the function with respect to the the different terms of \( f \). An example would be the total uncertainty in the dielectric constant,

\[ \varepsilon = \frac{C t}{\varepsilon_0 A} \quad (B.2) \]

which depends on the electrode area, \( A \), thickness, \( t \), and measured capacitance \( C \). The permittivity of free space is not considered uncertain. The total uncertainty in this case would be given by

\[ u_\varepsilon \leq \left| u_c \frac{\partial \varepsilon}{\partial C} \right| + \left| u_t \frac{\partial \varepsilon}{\partial t} \right| + \left| u_A \frac{\partial \varepsilon}{\partial A} \right| \quad (B.3) \]

The error as a fraction of the measured dielectric is given by dividing this equation by B.2. Also substituting for the partial derivatives for this example gives

\[ \frac{u_\varepsilon}{\varepsilon} \leq \left| \frac{u_c}{C} \right| + \left| \frac{u_t}{t} \right| + \left| \frac{u_A}{A} \right| \quad (B.4) \]

What follows is a brief description of the measurement errors and their estimated/reported values.

• Viscosity: \( \eta \) Viscometer was calibrated against standardized fluid, resulting in the reported maximum accuracy of 1% of the working range, where the working
range was defined by percentage of full motor torque. Since comparisons between samples of different viscosities were made at equal shear rates, the torque was often as low as 10% of the full range, resulting in a maximum sample error of 10%.

- **Displacement**: (x) Displacement was measured using a laser sensor, which had been calibrated with a micrometer, and shown to be within 1% of the measured output.

- **Capacitance and dissipation**: (C, D) HP meter had a minimum accuracy of 0.17% for the conditions used in measurement of the matrix and composite samples.

- **Resistance**: (R) Error is specified to be 1.15% of working range. Since ranges are spaced by one order of magnitude, the maximum error possible is 11.5% (i.e. a sample at the very low end of the range).

- **Thickness** (composites and dielectric breakdown samples): (h) Variations in repeated measurements showed an uncertainty of 0.0001 inches, over a total thickness of approximately 0.006 inches (error = 2%).

- **Electrode Width** (composites): (w) Thickness of electrode finger assumed accurate to 0.00025 inches (0.25 mil) by measuring under microscope, compared to the nominal value of 0.0075 inches, giving possible error of 3.3% for total electrode area.

- **Electrode Spacing** (composites): (p) Although specified to be 0.045 inches, could not accurately measure to better than 0.002", giving possible error of 4.4%.

- **Electrode Length** (composites): (l) Assumed to be measured accurately to within 1%.

- **Gage Length** (composites): (L) Test length between sliding and stationary clamps, over which actuation displacement was measured. Assumed measured to within 1%.

- **Thickness** (matrix disks): (t) Thickness of disks was not constant, and micrometer reads only highest point over measurement surface. Maximum estimate of thickness uncertainty is 0.0005 inches, over nominal 0.015 inches, resulting in error of 3.3%.

- **Electrode Area** (matrix disks): (A) Variation in disk diameters on order of 0.025 inches (over a 1 inch nominal diameter), resulting in error of 5% for area.

- **Voltage** (general): (V) Function generator and high voltage amplifier assumed to be accurate to within 1% of setting.

- **Voltage** (dielectric strength tests): \((V_{ds})\) Voltage steps in ‘step by step’ test method could not be done instantaneously, but were dialed up quickly, resulting in
a possible error of one voltage step for breakdown (due to time requirements). Maximum error was approximately 5% of breakdown field.

Propagation Errors:

- **Dielectric**: (Matrix Disks) 8.3%

  \[ \varepsilon = \frac{C_t}{\varepsilon_o A} \]  
  \hspace{1cm} (B.5)

- **Dielectric**: (Composites) 8.7%

  \[ \varepsilon = \frac{C_p}{\varepsilon_o (w \cdot l)} \]  
  \hspace{1cm} (B.6)

- **Resistivity**: (Matrix Disks) 20%

  \[ \rho = \frac{R_t}{A} \]  
  \hspace{1cm} (B.7)

- **Induced Stress/Strain**: (Composites) 7.4%

  \[ d_{33} = \frac{(x/L)}{(V/p)} \]  
  \hspace{1cm} (B.8)

- **Dielectric Strength**: (Composites) 7%

  \[ D.S. = \frac{V_{DS}}{h} \]  
  \hspace{1cm} (B.9)

C. Index of Suppliers


PZT 5A/4 Square Fibers, *Stavely Sensors Inc.*, 91 Prestige Park Circle, East Hartford, CT, 06108.


Baseline Epoxy, Epon 9405/9470, *Shell Chemical Company*, Houston, TX.

Dielectric Filler, 1 \textmu m dia 5H Powder; *Morgan-Matroc Inc.*, 232 Forbes Road, Bedford, Ohio 44146.


Electrode Material, 0.5 mil Copper/Kapton, *Southwall Technologies*, 1029 Corporation Way, Palo Alto, CA 94030.
Ultrasonicator, Model VC-50, Sonics and Materials, Danbury, CT 06810
High Voltage Amplifier, Model 664, *Trek Inc.*, 3932 Salt Works Road, Medina, NY 14103
Oil Bath, Exacal EX-250HT, *Neslab Instruments Inc.*, 25 Nimble Hill Road, Midland, MI 48686.
Electrometer, Model 6517, Keithley, 28775 Aurora Rd., Cleveland, OH, 44139.
Impedance Analyzer, Model 4194a, Hewlett Packard, 4 Choke Cherry Road, Rockville, MD, 20850.