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Department of Mechanical and Nuclear Engineering

CHARACTERIZATION AND MODELING OF ACTIVE FIBER COMPOSITES

A Dissertation in
Mechanical Engineering

by
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ABSTRACT

Active Fiber Composites (AFCs) are long fiber composites, where the fibers are made from lead zirconate titanate (PZT), which is one of the most widely used piezoelectric materials owing to its high electro-mechanical coupling and high piezoelectric coefficients. The PZT fibers are embedded in an epoxy matrix to give the composite more flexibility and more ruggedness. AFC utilizes interdigitated electrodes (IDE) along the direction of the fibers resulting in a $d_{33}$-mode actuation (actuation parallel to the poling direction), which is advantageous since the $d_{33}$ coefficient is almost twice the value of the $d_{31}$ coefficient (actuation perpendicular to the poling direction). The development of AFCs offers a great potential for advancing structural health monitoring techniques and active vibration control and suppression thanks to their flexibility and lightweight, however AFCs piezoelectric performance is still not comparable to that of PZT transducers, where their effective actuation coefficient is 120 compared to $400 \times 10^{-12}$ m/V for the bulk PZT. In addition, while in use, AFCs could be subjected to simultaneous mechanical loading, extreme environments, and moderate to high electric fields, leading to nonlinear and inelastic behaviors, strong coupling between various physical properties, and complex failure mechanisms. Moreover, AFCs have more than 50% volume content of polymer; the viscoelastic effect of the polymer matrix on the overall response of AFC can be significant especially at elevated temperatures. The effects of time and temperature on the mechanical and electrical characterization of AFCs have not been studied. Modeling these phenomena for the AFCs is an important step in redesigning them and obtaining reliable properties. However, there is not an available model that
takes into consideration combined physical phenomena like time-dependence and piezoelectric non-linearity. In addition many models assume uniform electric field or perfect contact between the fibers and the electrodes, which neglects the effects of any IDE geometry parameters and electrical property of the matrix.

The overall objective of this proposed dissertation is to use a combination of experimental and numerical approaches to examine the overall behavior of the AFC with the goals of 1) quantifying the impact of the constituent properties (polymer matrix and PZT fiber) on coupled response of AFCs, 2) conducting an exhaustive parametric study focused on design of AFCs, and 3) building a model which takes into consideration non-uniform electric field behavior, non-linear behavior and time-dependent properties.

Mechanical, electrical and electro-mechanical experimental characterization of AFC and its constituents at various temperatures and loading rates were carried out to bring to light the behavior of AFC, including the time and temperature dependence of some of its properties. Numerical parametric study on the design of the composites that takes into effect the IDE geometry and matrix dielectric constant and modulus is conducted. In addition, the experimental work quantified the non-linear and time-dependent responses of the AFCs, which are then included in the developed model. One outcome of this study is to propose a re-design of an improved AFC device with better electro-mechanical coupling response.
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Chapter 1

Introduction

1.1 Background

Smart materials are capable of achieving more than structural utility; they can also contribute electrical, magnetic, and optical power generative roles to the host structure. The development of these materials and their incorporation into the host structure have the potential to reduce its weight, cost, and power consumption and improve its efficiency and safety [1]. Among the most commonly used smart materials are shape memory alloys (SMA), magnetostrictive materials, electrostrictive materials, and piezoelectric materials.

1.1.1 Piezoelectric materials

Due to their electro-mechanical coupling, piezoelectric materials are one of the widely used multifunctional materials for sensing, actuation and energy harvesting. Piezoelectric materials can convert electrical energy to mechanical energy and vice versa. This electro-mechanical coupling consists of the conversion of mechanical loading to electric charges, called the direct effect, whereas the converse effect is the conversion of the applied electric field to mechanical deformation. The Curie brothers first discovered piezoelectricity in 1880. Through their collaboration, they demonstrated piezoelectricity in tourmaline, quartz and Rochelle salt [2]. The use of piezoelectric materials in sonar
applications during World War I increased research interest in them, leading to discovery and synthesis of new classes of ceramics that exhibit piezoelectricity [2-4].

Materials can be divided into 32 point crystal groups, 11 of them are centrosymmetric (also known as centric) and the other 20 are noncentrosymmetric (also known as acentric). Piezoelectric materials are materials belonging to the acentric point groups that can be further divided to two equal groups: Non-pyroelectric (non-polar) and pyroelectric (polar) materials that have spontaneous electric polarization in the absence of an applied electric field, with polar point groups given as 1, 2, m, 2mm, 4, 4mm, 3, 3m, 6 and 6mm [5]. A subset of polar materials can switch the spontaneous polarization to a different and stable direction when an electric field of a sufficient magnitude, it is called ferroelectric materials [6]. They are the most commonly used piezoelectric materials thanks to their high coupling coefficients.

An important crystal structure for ferroelectric materials is the perovskite structure with the formula ABO$_3$ [4]. Perovskites can exhibit rhombohedral, orthorhombic as well as tetragonal structures in the ferroelectric phase or cubic structure in the paraelectric phase. There is a critical temperature for ferroelectric materials, called Curie temperature ($T_c$), above which the phase is paraelectric and below which the phase is ferroelectric. Figure 1-1 shows crystal cell for cubic structure and tetragonal structure. Close inspection of the tetragonal structure shows that the center of mass of positive and negative ions does not coincide, forming a permanent dipole. Dipole size can be changed either by applying an electric field or applying pressure to change the distance separating the centers of mass of positive and negative ions. The most common ferroelectric ceramics with this structure
are barium titanate (BaTiO$_3$) and lead zirconate titanate (PbZr$_x$Ti$_{1-x}$O$_3$), also known as PZT, thanks to their high coupling coefficients and high dielectric permittivity.

Adjacent dipoles oriented in the same direction form what is called ferroelectric domains. In a polycrystalline material, the dipoles of domains are oriented randomly and will cancel each other on a global scale. To align the domains in polycrystalline materials, a poling step is introduced: i.e., applying a strong DC electric field at a temperature slightly below the Curie temperature, for a given amount of time. The higher temperature will increase the mobility of the domain walls. As the electric field is applied, the domains align in the direction of the field; and upon removal of the field, most of the domains remain pointing in that direction as shown in Figure 1-2 and result in a remnant

Figure 1-1: Elementary crystal cell; (a) cubic configuration and (b) tetragonal configuration [7].
polarization. The applied field, the temperature, and the time are all parameters that affect the degree of poling.

![Diagram of polarization](image)

Figure 1-2: Poling a piezoelectric material: a) random orientation of polar domain prior to polarization, b) Polarization in DC electric field and c) Remnant polarization after electric field is removed.

When the sample is poled and a high magnitude electric field is applied in the direction opposite to the poling, the resulting dipole rotates. Figure 1-3 shows the polarization as a function of the electric field after applying a high amplitude sinusoidal electric field. The obtained plot is called a hysteresis loop and three ferroelectric properties can be extracted from it as shown in the figure. The saturation polarization ($P_s$) is the maximum value of the polarization that represents the sum of all the dipoles aligned in the direction of the electric field. The remnant polarization ($P_r$) is the value of the polarization when the applied field is zero and it represents the sum of the dipoles aligned in the poling direction in the absence of the electric field (known also as spontaneous polarization). The coercive field ($E_c$) is the electric field when the measured polarization is equal to zero that corresponds to the minimum electric field necessary for polarization switching (switching the poling direction).
Experimental studies have also shown that mechanical stress can cause orientation of dipoles, poling or phase transformation [8-10] in some ferroelectric materials. This transition is called ferroelastic switching. For example, when a stress above a critical magnitude is applied in the poling direction, the domains rotate by 90° and become perpendicular to the initial poling direction. The 90° domains rotation can cause irreversible loss of sensing or actuation as the polarization in the poling direction becomes close to zero. The critical magnitude is called the coercive stress $\sigma_c$ in an analogy to the coercive electric field and it can be as low as 10 MPa for some materials.

The non-linear mechanical response observed in some ferroelectric materials is mainly due to ferroelasticity or mechanically initiated domains reorientation when the stress is above the coercive magnitude [11, 12]. Even though most of the ferroelectric materials are ceramics, they exhibit creep behavior at room temperature. The creep response is also
mainly due to ferroelasticity as it was shown that the ferroelastic switching is a time dependent process [13]. These observations show that studying the mechanical properties and behavior of piezoelectric materials is as important as their electro-mechanical behavior even though their primary use is not structural.

1.1.2 Constitutive equations of piezoelectric materials

Piezoelectric materials exhibit mechanical, dielectric, and piezoelectric behavior. In this section basic constitutive laws for these different behaviors will be explained.

The mechanical behavior of a linear elastic material can be described by a relationship between stress and strain as shown in (1.1), where $T$ is the second-rank stress tensor expresses in N m$^{-2}$, $S$ is the second-rank strain tensor, and $s$ is the fourth-rank elastic compliance tensor expressed in m$^2$ N$^{-1}$:

$$S_{ij} = s_{ijkl} T_{kl}$$ (1.1)

The dielectric behavior of an insulating polarizable material can be expressed by the relationship between polarization and applied electric field as shown in (1.2). $P$ is the induced polarization vector expressed in C m$^{-2}$, $E$ is the applied electric field vector expressed in V m$^{-1}$, and $\chi$ is the second rank dielectric susceptibility tensor expressed in F m$^{-1}$.

$$P_i = \chi_{ij} E_j$$ (1.2)
The measured variable when applying the electric field is usually the dielectric displacement $D$ expressed in C m$^{-2}$ that is given by (1.3), where $\varepsilon_0 = 8.854 \times 10^{-12}$ F m$^{-1}$ is the dielectric permittivity of vacuum.

$$D_i = P_i + \varepsilon_0 E_i$$  \hspace{1cm} (1.3)

From equations (1.2) and (1.3) the dielectric displacement can be expressed as:

$$D_i = \varepsilon_0 E_i + \chi_{ij} E_j = \varepsilon_0 \delta_{ij} E_j + \chi_{ij} E_j = \left( \varepsilon_0 \delta_{ij} + \chi_{ij} \right) E_j = \varepsilon_{ij} E_j$$  \hspace{1cm} (1.4)

where $\varepsilon_{ij}$ is the dielectric permittivity of the material and $\delta_{ij}$ is Kronecker’s symbol. The relationship between the permittivity and susceptibility can be expressed as:

$$\varepsilon_{ij} = \left( \varepsilon_0 \delta_{ij} + \chi_{ij} \right)$$  \hspace{1cm} (1.5)

For most of the widely used piezoelectric ceramics like PZT $\chi_{ij} \gg \varepsilon_0 \delta_{ij}$, and as a result, $\varepsilon_{ij} \approx \chi_{ij}$.

The piezoelectric behavior can be explained in two different ways: the direct effect and converse effect. Applying mechanical stress will produce dielectric displacement and polarize the material—the direct effect. The constitutive equation for this effect is written in (1.6), where $d_{ijk}$ expressed in C N$^{-1}$ is the third-order piezoelectric coefficients tensor.

In the converse effect the material can contract or expand when an electric field is applied. The constitutive law in this effect is the relationship between the strain and the electric field shown in (1.7), where the superscript $tr$ denotes the transposed matrix. The unit of the piezoelectric coefficients in this case is m V$^{-1}$.

$$D_i = d_{ijk} T_{jk}$$  \hspace{1cm} (1.6)

$$S_{ij} = d_{kij} E_k = d_{ijk} E_k$$  \hspace{1cm} (1.7)
Piezoelectric materials exhibit coupling of the previously stated behaviors. The equations for this coupling are shown in (1.8).

\[
D_i = \varepsilon_{ij}E_j + d_{ijk}T_{jk} \\
S_{ij} = d_{kij}E_k + s_{ijkl}T_{kl}
\]

Due to the symmetry of the stress and strain tensors, by definition the compliance tensor and piezoelectric coefficients tensors can be written in matrix form following the Voigt convention given in Table 1-1 [14]. The constitutive law can be written as shown in (1.9) or (1.10).

\[
D_i = \varepsilon_{ij}E_j + d_{ij}T_k \\
S_i = d_{ik}E_k + s_{ij}T_j
\]

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

(1.10)

Table 1-1: Rules of matrix notation.

<table>
<thead>
<tr>
<th>Tensor notation</th>
<th>Corresponding matrix notation</th>
</tr>
</thead>
<tbody>
<tr>
<td>( ii ) = 11, 22, 33</td>
<td>( m = 1, 2, 3 )</td>
</tr>
<tr>
<td>( ij ) = 23 or 32, 13 or 31, 12 or 21</td>
<td>( m = 4, 5, 6 )</td>
</tr>
<tr>
<td>( s_{ijkl} )</td>
<td>( s_{mn}, ) both ( m ) and ( n = 1, 2, 3 )</td>
</tr>
<tr>
<td>( 2 \ s_{ijkl} )</td>
<td>( s_{mn}, ) ( m ) or ( n = 4, 5, 6 )</td>
</tr>
<tr>
<td>( 4 \ s_{ijkl} )</td>
<td>( s_{mn}, ) both ( m ) and ( n = 4, 5, 6 )</td>
</tr>
<tr>
<td>( d_{ijkl} )</td>
<td>( d_{lm}, ) ( m = 1, 2, 3 )</td>
</tr>
<tr>
<td>( 2 \ d_{ijk} )</td>
<td>( d_{lm}, ) ( m = 4, 5, 6 )</td>
</tr>
</tbody>
</table>

Piezoelectric materials are anisotropic and their properties depend on the poling direction, which corresponds to the 3-direction. The properties in the 1-direction and 2-direction are equal to each other and are different from the ones in poling direction. Typical properties of soft PZT (the one used throughout this dissertation) and hard PZT are shown in the table below and the full tensors of soft PZT are shown in Appendix.
Table 1-2: Typical materials properties for soft and hard PZT [15].

<table>
<thead>
<tr>
<th></th>
<th>Symbol</th>
<th>Unit</th>
<th>Soft PZT</th>
<th>Hard PZT</th>
</tr>
</thead>
<tbody>
<tr>
<td>Density</td>
<td>( \rho )</td>
<td>g/cm(^3)</td>
<td>7.80</td>
<td>7.80</td>
</tr>
<tr>
<td>Temperature</td>
<td>( T_c )</td>
<td>°C</td>
<td>350</td>
<td>330</td>
</tr>
<tr>
<td>Relative permittivity in poling direction</td>
<td>( \varepsilon_{33}^T / \varepsilon_0 )</td>
<td></td>
<td>1750</td>
<td>1200</td>
</tr>
<tr>
<td>Relative permittivity ( \perp ) to polarity</td>
<td>( \varepsilon_{11}^T / \varepsilon_0 )</td>
<td></td>
<td>1650</td>
<td>1500</td>
</tr>
<tr>
<td>Dielectric loss factor</td>
<td>( \tan \delta )</td>
<td>( 10^{-3} )</td>
<td>20</td>
<td>3</td>
</tr>
<tr>
<td>Piezoelectric charge coefficient</td>
<td>( d_{31} )</td>
<td>( 10^{-12} ) C/N</td>
<td>-180</td>
<td>-120</td>
</tr>
<tr>
<td></td>
<td>( d_{33} )</td>
<td></td>
<td>400</td>
<td>265</td>
</tr>
<tr>
<td></td>
<td>( d_{15} )</td>
<td></td>
<td>550</td>
<td>475</td>
</tr>
<tr>
<td>Piezoelectric voltage coefficient</td>
<td>( g_{31} )</td>
<td>( 10^{-3} ) Vm/N</td>
<td>-11.5</td>
<td>-11.2</td>
</tr>
<tr>
<td></td>
<td>( g_{33} )</td>
<td></td>
<td>22</td>
<td>25</td>
</tr>
<tr>
<td>Elastic compliance coefficient</td>
<td>( s_{11} )</td>
<td>( 10^{-12} ) m(^2)/N</td>
<td>16.1</td>
<td>11.8</td>
</tr>
<tr>
<td></td>
<td>( s_{33} )</td>
<td></td>
<td>20.7</td>
<td>14.2</td>
</tr>
</tbody>
</table>

1.1.3 Piezoelectric composites

Piezoelectric materials are materials of choice for both sensing and actuation. Piezoelectric materials can be used in structural health monitoring as embedded sensors [16]; they can also be used as actuators in active damping and vibration control [17]. The most common piezoelectric materials used in devices are PZT, a piezoelectric ceramic or polyvinylidene fluoride (PVDF), which is a piezoelectric polymer. Piezoelectric ceramics have some size and shape limitations because of their brittle behavior and relatively high density [18], while the piezoelectric polymer has relatively low electro-mechanical and mechanical properties compared to the ceramic. In order to take advantage of the best properties of ceramics and polymers, researchers have focused on a two-phase active
composite. Active composites were designed using different components and different connectivity patterns. The connectivity patterns define the arrangement of the phases and the way that the single phases are connected [19, 20]. A nomenclature of two digits was used to define the connectivity patterns for diphasic composites; each number can go from zero to three. Each number defines the number of directions in/to which each phase may self-connect. The first digit usually corresponds to the piezoelectric phase; different connectivity patterns in composites with the same phases can produce physical properties that are different by orders of magnitude. Figure 1-4 shows 0–3, 1–3 and other composites with different connectivity.

The first piezoelectric composites developed had 0–3 connectivity, where the polymer is connected in three directions and embedded with piezoelectric powder [21, 22]. The main advantage of 0–3 composites is their ease of fabrication in various shapes and in mass production. More 0–3 composites were later developed using different polymers and different piezoelectric powders. Chloroprene rubber and PZT powders of different sizes, ranging from 3 to 30 micrometers in diameter, have been mixed, producing a composite with a $d_{33}$ up to $50 \times 10^{-12}$ C/N [23-25]. Despite the simplicity of the processing of 0-3 composites they face some challenges when the concentration of dispersed particles is high due to the non-homogeneous distribution of piezoelectric particles, their tendency to agglomerate and the presence of air voids that reduce the dielectric breakdown and make the poling of the composites more difficult. To overcome these challenges, colloid processing methods discussed in [26] were used on piezoelectric particles: (1) dispersion of the ceramic powders in a dilute polymer solution where polymer is adsorbed onto
ceramic powder surfaces; (2) addition of a polymer nonsolvent to effect phase separation of the polymer-powder coacervate, a colloid-rich viscous liquid phase, from the dispersion; (3) colloidal filtration of polymer-powder coacervate; and (4) cold pressing [26]. Using the colloid process method improves the homogeneity of the phase distributions and increases the $d_{33}$ coefficient from 27 to 65 pC/N in a (Pb$_{0.5}$, Bi$_{0.5}$)(Ti$_{0.5}$, (Fe$_{1-x}$, Mn$_x$)$_{0.5}$O$_3$ ($x = 0.00$ to 0.02) powder epoxy polymer. Moreover, composites processed via the colloidal route were found to withstand poling fields up to 150 kV/cm as compared to less than 120 kV/cm for conventional composites (composition and solids loading). However, when poling a 0–3 composites, the electric field is mostly controlled by the dielectric constant of the polymer phase which is known to have a lower dielectric constant compared to piezoelectric materials. Therefore the electric field lines will concentrate in the polymeric phase due to its lower dielectric constant. This issue was addressed by introducing a third conductive phase in the polymeric phase like carbon, germanium, or silicon [27] which results in a decrease of the dielectric contrast between polymer and ceramic phases.

In parallel to 0–3 composites development, researchers were exploring 1–3 composites using piezoelectric rods instead of powders. In 1–3 composites, the rods are usually aligned perpendicular to the electrodes and embedded in a polymer matrix [21].
In order to overcome the challenges of manipulating and aligning the brittle rods, a different technique was used to produce 1–3 composites [29]. In this method, a PZT disc is sliced in one direction and filled with epoxy; then, the same disc is sliced in a direction perpendicular to the first and again filled with epoxy leading to a diced composite. In order to improve the piezoelectric coefficients of the composite, a foamed polyurethane matrix was used [30]. The compliant matrix allowed better stress transfer to the rod, increasing the sensing ability of the composite. For other applications like ultrasonic transducers used for medical imaging, 2–2 piezoelectric composites made of stacks of PZT and epoxy are used [31]. The 2–2 composites use the resonance in the thickness mode in order to propagate waves.

In order to couple stresses and strains associated with planar bending and twisting, other
forms of composites have been developed. In 1993, Bent and Hagood introduced Piezoelectric Fiber Composites (PFC) consisting of thin and long continuous PZT fibers aligned in an epoxy matrix in the same plane; the electrodes are parallel to the fiber in order to apply an electric field through the thickness (Figure 1-5)[32].

![Figure 1-5: Schematic of PFC][32]

These piezoelectric fiber composites (PFC) work on the transverse mode, taking advantage of the piezoelectric coefficient $d_{31}$ of the fibers. PFCs are thin; they can be inserted in a variety of structures, and can be manufactured along large surfaces without the risk of fracture present in large PZT plates. Moreover, they are flexible and so can fit curved surfaces. Flexibility in these composites was made possible by the development of thin PZT fibers manufactured by different processes: the Viscous Suspension Spinning Process (VSSP) developed by Advanced Cerametrics Inc [33] and the ALCERU Process [34]. Fibers are manufactured with various diameters from a few micrometers to hundreds of micrometers. The low diameters of the fibers and their sintering process
(slightly different from bulk PZT) offer more flexibility to the fibers, and as a result, to the composite, due to the presence of epoxy. Because of the application potential of PFCs, researchers have tried to maximize PFC’s efficiency and effective piezoelectric behavior. One way to do that was to use interdigitated electrodes (IDEs) that produce electric field lines in the direction of the fibers instead of through the thickness of the composite itself, hence taking advantage of the $d_{33}$ coefficient rather than the $d_{31}$. As shown in Section 1.1.2, the $d_{33}$ coefficient is twice as high as $d_{31}$, therefore this is a promising strategy. These developed composites are called Active Fiber Composites (AFCs) and are discussed in detail next.

1.2 Active Fiber Composites

1.2.1 Overview

Bent and Hagood [35] have developed a composite of a ceramic and a polymer, taking advantage of the best properties of each component, and have named it an Active Fiber Composite. AFCs are long fiber composites in which the fibers are made from PZT. A schematic of an AFC is shown in Figure 1-6 below. The PZT fibers are nominally 250 micrometers in diameter and are embedded in an epoxy matrix to give the composite more flexibility and ductility. As shown, the AFC utilizes interdigitated electrodes along the direction of the fibers [36]. This electrode configuration was previously used by Hagood [37] to improve the actuation of piezoceramic plates: The use of the conventional parallel electrodes actuates the plate transversally based on the $d_{31}$ coefficient, whereas
interdigitated electrodes result in a $d_{33}$ actuation in which the $d_{33}$ coefficient is almost twice the value of the $d_{31}$ coefficient. The small electrode gap in the IDEs makes it possible to apply a high electric field through the fibers without applying high voltage. To encapsulate the device, and therefore improve mechanical integrity and lower the chances of breakdown and shorting, a Kapton tape is used over and below it [38]. The IDEs are usually printed on the Kapton layers and then are glued to the epoxy and fiber composite on the top and bottom. The whole set up is then cured, leaving a small layer of epoxy between the fibers and the electrodes to ensure the bonding of the Kapton layers as shown in Figure 1-6.

![Figure 1-6: Representation of Active Fiber Composites: a) Constituents, b) Typical dimensions.](image)

1.2.2 Applications and properties of AFCs

The development of AFCs offers great potential to many applications, including the advancement of structural health monitoring techniques, stealth and morphing aircrafts, high-speed vehicles, and energy harvesting. Some studies [39, 40] have discussed the use
of AFCs as sensors for self-health monitoring (SHM) and damage detection, along with their limitations. AFCs were first installed on glass-reinforced polymer composites and their SHM were compared to using commercial resonant Acoustic emission (AE) sensors. The limited use for low-frequency applications with a resonance frequency around 50 kHz and the non-availability of long-term behavior are the main disadvantages of AFCs compared to commercial sensors. AFCs were also tested on pipes of diameters as low as 50 mm, and showed an ability to detect leaks in those pipes. However, when subjected to strains of more than 0.1 %, their sensitivity decreases due to damage in the piezoelectric fibers. AFCs also can be used as actuators for vibration suppression [17]. They were compared to monolithic piezoelectric ceramic actuators experimentally and numerically. It was shown that their performance in vibration suppression per active unit volume is equivalent to monolithic actuators in the 3-1 mode and better than those in the 3-3 mode. AFCs can furthermore be used for pre-twisted rotating composite thin-walled beams [41, 42] or as actuators where they have been installed on a bionic flapping wing and showed an improvement in aerodynamics of the wing [43].

An in-depth look at the literature confirms that although the flexibility and lightweight of AFCs opened up a wide range of applications, their piezoelectric performance is still not comparable to that of PZT transducers (120 to 400 $10^{-12}$ m/V). In addition, each of the above applications could subject the AFC to simultaneous mechanical loading, extreme environments, and moderate to high electric fields. These could well lead to non-linear behaviors and strong coupling between various physical properties in the AFC composites, causing the constituents of the AFC to experience non-linear and inelastic behaviors and so result in complex failure mechanisms. For example, heat generation and
shape changes at different rates during high-speed flights, wherein dynamic effects such as vibration are pronounced, cause complex multi-field responses at multiple time-scales. Similarly, a high electric field applied opposite to the poling axis can cause polarization switching in the PZT fibers, affecting the actuation performance of the AFC. In addition, since the fiber volume content of AFCs is generally below 45%, the volume content of polymer is above 50%—and so the viscoelastic effect of the polymer matrix on the overall response of AFC can be significant, especially at elevated temperatures. The softening of the matrix at high temperatures transfers more stress to the fiber and may cause damage in the fibers, affecting the sensitivity and performance of AFCs [39]. In addition, the polarization-switching response of AFCs under different frequencies and temperatures has not been fully explored. In an attempt to further identify the issues and challenges still associated with AFCs, we next analyze each of the constituents (namely, the PZT fibers and the epoxy matrix) separately.

The piezoelectric fibers embedded in the AFC’s are what make this composites active. In fact, these fibers are made of Lead Zirconate Titanate (PZT) which is a ferroelectric polycrystalline ceramic widely used in industrial, medical, military and domestic applications. These PZT fibers are produced through three different techniques: extrusion[44], spinning [45] and sol-gel processing[46]. The main difference between these three methods is the nature of the precursor involved and the way the green fibers are formed before firing and sintering. Due to these processing conditions, a number of authors pointed out the importance of characterizing single PZT fibers versus bulk PZT. Dent et al.[47] studied the microstructure of PZT fibers in the context of the different
manufacturing routes used to produce said-fibers. Kornmann et al. correlated the microstructure of the fibers to their mechanical properties [48, 49]. Dittmer studied the microstructural and mechanical properties of PZT fibers by using different powders, fiber shaping techniques as well as different fiber diameters [50]. However, in all of these studies there is a lack of knowledge concerning the dependence of the tensile properties with respect to temperature and strain rates. In addition, due to the specific applications where the PZT fibers are being used, such as in AFC for instance, it is of a primary interest to characterize their mechanical behavior under creep, relaxation and tensile test under different strain rates. For example, in load-bearing applications where AFCs are used as actuators, they are subjected to a constant stress during an extended period of time. Thus, it is crucial to understand the PZT fibers’ response and therefore the whole AFC response over this period of time. Moreover, although PZT exhibit outstanding electro-mechanical properties, their non-linear mechanical behavior has impeded their wide use [51]. In fact, it has been found that PZT ceramics exhibit a strong non-linearity in stress-strain curves [52, 53] and present an apparent “plasticity”. This was attributed to ferroelastic domain switching [54, 55] which results in an “apparent” plasticity and crack growth resistance behavior [56]. Although a great number of authors studied the non-linear mechanical behavior of bulk PZT there are only few studies regarding the mechanical performance of single PZT fibers. Consequently there is scant data about tensile strength and ferroelastic behavior of PZT fibers. Several experimental studies suggest time-dependent electro-mechanical behavior of the PZT as well. Schäufele studied the impact of loading and unloading compressive stress on the polarization and on strain; it was shown than when the loading is held for one hour the remnant strain and
polarization after unloading are more than one order of magnitude larger than when the loading is one second [55]. It was also observed that despite their brittle nature, PZT ceramics show creep response under constant stress at room temperature [57]. Other studies [58, 59] have examined the creep response in soft PZT under static electric fields and compressive stresses. The creep responses were more pronounced at higher stresses and at electric fields near the coercive electric field. When subjected to cyclic electric fields, the ferroelectric ceramics show hysteretic responses that strongly depend on the amplitude of electric fields, frequency, the existence of stresses, and ambient temperatures. Other experimental studies have investigated the electro-mechanical response and the effect of cyclic frequencies on changes in the elastic modulus of active fiber composites [60, 61]. In summary, the time-temperature effects on the overall electro-mechanical behavior of PZT fibers and AFC are of a prime interest in order to exhaustively characterize the overall behavior of AFC and thus need to be investigated as literature review showed scant data.

In general, there is not much focus in the literature on the experimental determination of AFCs’ mechanical properties, most likely because their function is not to carry a load in the structure. It is still important, however, to know these properties in order to prevent failure or loss of sensing capability. A small number of papers have done experimental studies to obtain the mechanical properties of AFC for different volume contents and have investigated the failure modes for different fiber volume fractions [62, 63]. A polymer matrix is attractive because polymers are lightweight and compliant, resulting in flexible active composites. The existence of polymer in AFCs also prevents
catastrophic failure due to fiber breaking. However, the presence of the polymer also has disadvantages related to the inherent viscoelastic and viscoplastic responses of the polymer. Also, the presence of the polymer is responsible for the large contrast in mechanical and electrical properties between polymer and ceramic, which affects electric and mechanical load transfer at the interphase. There have been extensive experimental and theoretical studies conducted on the physical and chemical mechanisms underlying the time-dependent and hysteretic behaviors of polymers, attributed to the viscoelastic and/or viscoplastic response of polymers [64-67]. The polymer used in AFC is usually epoxy, which is an amorphous thermosetting polymer. Below its $T_g$, it is glassy; above its $T_g$, its viscoelastic behavior is more dominant. Determining the extent of the polymer’s role in the AFC’s electro-mechanical coupled response has not been investigated previously and it is crucial for determining what attributes need to be optimized to further improve active composites.

There are currently limited micromechanical models addressing time-dependent (viscoelastic) behaviors of active composites. Most of the work studying viscoelastic behavior is numerical; examples of such studies can be found in Li and Dunn [68], Jiang and Batra [69], and Muliana and Li [70]. In these studies, fibers were assumed to be linear piezo-elastic and the matrix was modeled as linear viscoelastic. Muliana [71] developed a simplified micromechanical model assuming non-linear piezoelectric fibers and with a viscoelastic polymer matrix that predicts the saturated polarization and strain during polarization switching in active fiber composites, as well as the overall electro-mechanical properties of the composite. Most other micromechanical models were
developed to predict linear piezo-elastic properties of active composites. Additional work has been done to evaluate the homogeneous properties of the composites [71-76]. So far, however, such work has not considered the IDE electrodes’ geometry that will create non-uniform electric fields and any electrical interaction between the matrix and the fiber; nor does this work, with the exception of a few, consider the non-linear and time dependent behavior of PZT fibers. The non-linear and time-dependent electro-mechanical constitutive models for ferroelectric ceramics, i.e., PZT, will account for polarization switching and dissipation of energy. The macroscopic responses of materials to external stimuli depends upon their microstructural changes at various length scales, such as atomistic, molecular, and polycrystalline. Models that incorporate changes in the polycrystalline structure of ceramics have been developed to predict the hysteretic responses of ferroelectric ceramics [77-81]. These studies, though, were all conducted for bulk PZT that is expected to have different behavior than fibers; this behavior has not been implemented in models of AFCs. Apart from material behavior, modeling the geometry and choosing the representative volume element (RVE) for the simulations is a crucial task for AFCs because of their complicated electrode geometry. Therefore, there is a need for Finite Element (FE) simulation to address these details and to examine their impact on the homogeneous properties of the composites. Many FE models of AFCs have been developed with different RVEs and under different assumptions. Tan [82] considered one fiber and one electrode gap as an RVE and proposed the electrodes to be rings surrounding the fibers, a circumstance termed circular-linked interdigitated electrodes. A micromechanical model of this was developed and properties were calculated for different volume fractions and different fibers’ cross sections (circular and
One other way to model the AFCs is to consider a quarter fiber, assume a uniform electric field, and study the effect of volume fraction [83]. There have also been studies on the IDE geometry parameters’ impact on the performance, but usually in the absence of the epoxy matrix around the fibers; i.e., these researchers assume direct contact between the electrodes and PZT [84, 85]. Paradies and Schläpfer examined the electro-mechanical response of a piezoelectric material with the use of IDE electrodes. This study investigated the effects of polarization and material orientation due to a non-uniform electric field. For AFC, 2D simulation was conducted and stress distribution was measured and compared to a uniform field model [86]. In 2010, the same authors [87] focused on AFC with a 3D model including the effect of material orientation variation. The authors assumed perfect contact between the electrodes and fibers at the zone of intersection; in addition, this study focused on the state of stress on the AFC, but did not investigate the electro-mechanical response. Another study by Belloli et al. considered the impact of the contact angle between electrodes and fibers, and the electrode gap on the $d_{33}$ coefficient of AFC in ANSYS [88] but other geometry parameters and epoxy matrix electrical properties were not taken into consideration.

Modeling AFC’s performance is of great importance in the quest to understand the impact of each design parameter and to obtain homogenized properties of the composite. However, there is no model available that takes into consideration combined physical phenomena such as viscoelasticity and piezoelectric non-linearity. In addition, many models assume a uniform electric field, and so neglect the effect of any IDE geometry parameters and they also do not consider the electrical properties of the matrix.
1.3 Problem statement

The objective of this proposed dissertation is to use a combination of experimental and numerical approaches to examine the overall behavior of AFCs. The specific goals are 1) quantifying the impact of AFCs’ constituent properties (polymer matrix and PZT fiber) on the coupled response of AFCs; 2) conducting an exhaustive parametric study focused on the design of AFCs; and 3) building a FE model that takes into consideration non-uniform electric field behavior and time-dependent properties. One outcome of this study is to provide guidance to redesign an AFC device to maximize its effective electro-mechanical coupling response. The experimental approach will consist of a full characterization of AFCs mechanically, electrically, and electro-mechanically at various temperatures and loading rates, which will better illuminate the behavior of AFCs and the time and temperature dependence of some of their properties, especially considering that epoxy represents around 50% volume fraction of the composite. Comparing the behavior of the composite with the behavior of its components (PZT fibers and epoxy matrix) will highlight the advantages and shortcomings of AFCs as compared to monolithic piezoelectric devices, and will quantify the impact of the constituents on overall properties of AFCs. The results of the experimental study will indicate the properties that are more affected by the design parameters. Understanding the behavior of AFCs and knowing their drawbacks will enable their redesign by taking advantage of technology advances in manufacturing and electronics. For an optimal design of AFCs, there should be a parametric study on the design of the composites that takes into account the fiber volume fraction, IDE geometry, and the
matrix dielectric constant and modulus. Previous studies in the literature have not specifically considered these design parameters. For example, it is important to consider the polymer layer separating the fibers from the matrix while investigating the electrode geometry because the thickness of that layer will affect the electric field distribution. Also, by assuming a uniform electric field inside the fiber, IDE geometry parameters such as electrode width will be neglected, so a parametric study with a uniform field will give incomplete results. In this study, an IDE electrode setup is used and also modeled to produce a non-uniform electric field on the fibers; in addition, a matrix interface between the electrodes and the fibers is introduced in the model to demonstrate the impact of the matrix properties on the behavior of AFCs. In addition, since previous experimental work has shown that AFCs and their constituents’ behavior is non-linear, temperature-dependent, and time-dependent, this suite of performances should be combined in one model.

1.4 Scope of the thesis

The first chapter presented a background on piezoelectric materials and the constitutive equations governing them. Furthermore, an overview on piezoelectric composites is given and a summary of the literature focusing on active fiber composites is discussed. Finally, in light of this background, the problem statement guiding this research is presented.
In chapter two, the experimental methods used for mechanically testing of AFCs, PZT fibers and epoxy are described. The results of the different tests (Tensile, Creep, Stress relaxation and DMA) are presented and discussed. The mechanical properties of the different samples are evaluated at several loading rates and temperatures.

In chapter three, the experimental procedures used for electrical and electro-mechanical characterization of AFCs, PZT fibers and PZT plates are described. Ferroelectric and electro-mechanical properties of the different samples are calculated for different temperatures and frequencies and compared to each other.

In chapter four, an AFC model is developed and implemented using finite element method (FEM). The model examines the effect of the IDEs parameters, the fiber diameter and matrix properties. This parametric study will lead to an improved geometry and matrix for an AFC with better actuation performance.

In chapter five, non-linear piezoelectricity, time-dependent polarization and polarization switching behaviors are included in the FE model of AFC’s and PZT fibers, to validate experimental results and better describe their true performances.

In chapter six, a summary of the obtained results is presented and future tasks are suggested to augment this current study.
Chapter 2

Mechanical Characterization of AFC and its Constituents

2.1 Materials and Equipment

2.1.1 Materials

Three different materials have been used for mechanical testing: AFCs, PZT fibers, and epoxy. The used AFCs were purchased from Advanced Cerametrics Incorporated. The AFC is 120 mm long, 20 mm wide, and 0.4 mm thick. In order to test them on tensile machines, 20 mm are used on each side to be attached to the grips. A photograph of the specimen is shown in Figure 2-1.

![Fracture](image)

Figure 2-1: Picture of an AFC specimen used in tensile experiment.

Though we wanted to test the fibers embedded in the AFCs, Advanced Cerametrics Incorporated did not have the required quantities commercially available. Instead, we looked for fibers with the same diameter and made of the same material (PZT 5A). PZT
5A fibers 150 mm long and 0.25 mm in diameter were purchased from Smart-Material Corp.

The matrix used in the AFC is epoxy Flex-A-Bond/740 supplied from United Resin Corporation. A kit was provided by them and was used to prepare tensile specimens. The kit is made up of two parts: a resin and a hardener. The two parts are mixed with a magnetic stirrer with equal mass ratios for approximately 20 minutes until the mixture is visually homogeneous. The mixture is then degassed in a vacuum oven at room temperature to remove air bubbles (approximately 30 minutes). The epoxy is then poured in the silicone mold and cured for four weeks at room temperature. The properties of this epoxy provided by the manufacturer are reported in Appendix. A summary of materials sources, dimensions and measured modulus are shown in Table 2-1.

Table 2-1: Summary of materials supplier dimensions and modulus

<table>
<thead>
<tr>
<th>Source</th>
<th>Dimensions</th>
<th>Modulus</th>
</tr>
</thead>
<tbody>
<tr>
<td>AFC</td>
<td>Advanced Cerametrics Inc</td>
<td>120 mm x 20 mm x 0.4 mm</td>
</tr>
<tr>
<td>PZT fibers</td>
<td>Smart Materials Corp.</td>
<td>150 mm long 0.25 mm diameter</td>
</tr>
<tr>
<td>Epoxy</td>
<td>United Resin Corp.</td>
<td>Figure 2-27</td>
</tr>
</tbody>
</table>

2.1.2 Equipment

Three different mechanical testing instruments were used to fit all material types used and the needed mechanical properties; a picture of these three set-ups is shown in Figure 2-2. A hydraulic MTS uniaxial test frame with a 30 kN load cell was used for tensile
experiments on AFCs. The MTS is equipped with an environmental chamber allowing temperature range from room temperature up to 250°C. For this system, strain was measured via crosshead displacement. A TA Instruments RSA G2 Dynamic Mechanical Analysis (DMA) was used for tensile testing of PZT fiber in addition to creep, relaxation, and dynamic tests. The DMA is equipped with grips designed specifically for fibers and thin film tensile configuration; the tightening of these grips is performed manually. The force resolution of the DMA is less than 1 mN and its displacement resolution is less than 1 µm. It is also equipped with a forced convection oven to control temperatures from -150°C to 400°C. An Instron Tensile 5800 series was used for tensile experiments conducted on epoxy samples. A 1kN load cell and clip-on extensometer were used with this equipment to record force and strain. Pneumatic pressure was used to attach the specimen to the grips.

Figure 2-2: Different equipment used for mechanical testing; from left to right: MTS 810, DMA RSA-G2, and Intron Tensile 5800.
2.2 A Mechanical Characterization of AFC

2.2.1 Tensile Experiments

AFCs are long thin fiber composites, which make their longitudinal properties the most important to study. The mechanical properties of these AFCs were evaluated using tensile testing in the longitudinal fiber direction. The tests were conducted under three different displacement rates (0.5, 5, and 25 mm/min, respectively equivalent to 0.625, 6.25, and 31.25%/min) in order to understand the time dependence of the mechanical behavior, and at different isothermal temperatures (25, 50, and 75º C) in order to ascertain temperature dependence. A displacement rate of 0.5mm/min is a relatively slow rate that allows for the rate-dependent plastic contribution of ferroelastic switching [89] and stress relaxation (creep deformation) in the composites to be ascertained, while at 5 and 25 mm/min the time-dependent effect should be negligible. The manufacturer specified the maximum operational temperature of the AFCs to be 80ºC, and so our temperatures were chosen to be between room temperature and this maximum value. Based on the specimens’ dimensions, the gage length for tensile tests was chosen to be 80 mm. The effective longitudinal properties found for the AFC were elastic (instantaneous) modulus, tensile strength, and maximum strain (also known as % elongation). The elastic modulus was determined by taking the slope of the stress-strain curve close to the initial (zero) value of stress. At least three tests were conducted for each temperature condition and loading rate.
Several stress-strain curves were obtained from the tensile tests, and the loading rate and impact of temperature on these longitudinal properties was investigated. Figure 2-3 shows a comparison of the stress–strain curves at 25°C for the different displacement rates. The response from the 0.5 mm/min rate (more than 30 minutes of loading) shows higher ductility and toughness for the specimens than those from the 5 and 25 mm/min rates (less than 2 minutes of loading), which resulted in relatively brittle responses. The strength of the AFCs obtained from different strain rates is almost the same. It can be concluded that the ductile behavior in the AFC with a slow loading rate might be due to a significant viscoelastic effect in the polymer matrix.

When we compare the temperature effect at the strain rate 25 mm/min in Figure 2-4, we observe differences in ductility, toughness, and strength. The temperature increase raises the ductility and the toughness and decreases the strength.

![Figure 2-3: Loading rates comparison of stress–strain curves of AFCs at 25°C.](image)
The effective elastic modulus, tensile strength, and maximum strain at various temperatures and loading rates are summarized in Figure 2-5 through Figure 2-7. These figures demonstrate that the mechanical properties of AFCs depend strongly on time and temperature.
Figure 2-5: Elastic modulus for different temperatures and loading rates.

Figure 2-6: Maximum strain for different temperatures and loading rates.
At room temperature, the elastic modulus of AFCs (Figure 2-5) is varying between 9 and 22 GPa depending at the strain rate. These values are in agreement with the maximum possible overall modulus. The fibers’ modulus varies between 25 and 40 GPa and the epoxy’s modulus varies between 0.5 and 1 GPa and volume fraction of each component is about 0.5. Using the arithmetic average, the modulus of the AFC can be expressed as:

\[
E_{AFC} = V_f E_{fiber} + (1-V_f) E_{matrix}
\]

\[
V_f E_{fiber}^{min} + (1-V_f) E_{matrix}^{min} \leq E_{AFC} \leq V_f E_{fiber}^{max} + (1-V_f) E_{matrix}^{max}
\]  \hspace{1cm} (2.1)

\[
12.75 \leq E_{AFC} \leq 20.5
\]

The mechanical properties strongly depend on the loading rate at room temperature. The elastic modulus (Figure 2-5), for example, increased by almost 100% from 0.5 mm/min to 25 mm/min at 25°C. At 50°C, the difference between strain rates was less pronounced. At 75°C the effect of the loading rate disappeared and the values were very close because the matrix became very soft and most of the load is carried by the fibers. Higher strain rates increased the overall stiffness and ultimate strength of AFC. Lower
strain rates, associated with a longer loading duration, indicated more pronounced effects of the viscoelastic matrix. The increase in temperature from 25°C to 50°C decreased the elastic modulus by 40% and the tensile strength by (25%) of the AFCs, and increased the maximum strain by 80%. This observed change in properties is most likely caused by the viscoelastic behavior of the epoxy matrix and the Kapton tape used to protect the electrodes. Another reason for this change is the existence of small air voids, which expand at high temperatures and so weaken the PZT fiber-epoxy, and Kapton-epoxy interfaces. The effect of temperature discussed above is verified by looking at the Scanning Electron Microscope (SEM) of the AFC fracture surfaces shown in Figure 2-8. At room temperature, the failure is caused by the fracture of the fibers inside the matrix, as evidenced by the cracks traversing the fibers. At 50°C and 75°C, the SEM images show the fibers intact and illustrate PZT fiber pull-out from the matrix, which confirms that the interface is weaker at higher temperatures. It was also observed that at elevated temperatures, matrix yielding and creeping increases failure strains without any fiber breaking.

Figure 2-8: SEM pictures for fractured surfaces at different temperatures (50µm scale): a) at 25°C; b) at 50°C; and c) at 75°C.
2.2.2 Creep and Relaxation

The results of the previous experiments show that the overall stress-strain relations of the AFCs subjected to uniaxial tension along the fiber direction are rate- and temperature-dependent. To further investigate the rate-dependent behavior, creep and stress relaxation were conducted using the DMA machine at different temperatures. AFC specimens were of the following dimensions: thickness=0.38mm; width=5 mm; and length=60 mm. The samples were clamped in the DMA grips and the gap length used in these experiments was around 40 mm. The specimens were cut from full size AFCs; their gap length and width were chosen to meet the DMA’s recommended specimen dimensions.

For stress relaxation, an instantaneous uniaxial tensile strain of 0.3% in the direction of the fibers was applied and maintained for 30 minutes before the stress was measured. 0.3% was chosen to avoid forces exceeding the load cell’s capability. For creep tests, an instantaneous uniaxial tensile stress of 15MPa was applied and held for 30 minutes before the strain was measured. These tests were performed at three different temperatures used for tensile experiment: 25°C, 50°C, and 75°C. The relaxation results are shown in Figure 2-9, where it can be seen that an increase in temperature reduces the initial level of stress. The initial stress is the elastic component of the relaxation curve and this confirms the temperature impact seen in the tensile tests. The room temperature stress relaxation showed a 28% decrease in stress after 30 minutes. A similar decrease of 29% was seen at 50°C, and a larger decrease of 44% was seen at 75°C. The temperature increase caused a drop of the remnant stress compared to room temperature: at 50°C the drop was 11% and at 75°C the drop was 38%.
The creep results (shown in Figure 2-10) are similar to the relaxation results. The creep strain at room temperature and 50°C are close; however, at 75°C, the strain level rises. The initial strain at 50°C and 75°C is higher than at 25°C which is in agreement with the tensile experiments. The room temperature creep showed a 69% increase in strain after 30 minutes. A similar increase of 70% was seen at 50°C, and a larger increase of 250% at 75°C. At elevated temperatures the matrix softens and becomes more viscoelastic and the interface between fibers and matrix is weakened and can undergo frictional sliding. These results confirm that the mechanical behavior is time- and temperature-dependent.

Figure 2-9: 0.3% stress relaxation results at different temperatures.
To better study the impact of temperature on the mechanical behavior of AFCs, a temperature sweep under dynamic loading experiment was conducted. The same DMA machine and sample sizes used for creep and stress relaxation have been used in this test with temperature increases at a rate of 2°C/minute. The strain oscillates with a maximum value of 0.02% and 1Hz frequency and the stress is measured and recorded as shown in Figure 2-11. 0.02% was chosen because it is within the elastic regime of the composite and the results it produces have a low noise level. 1Hz was chosen to avoid viscoelastic behavior and focus the results on the effect of temperature. The temperature ranged from room temperature to 100°C; at higher temperatures the specimen is damaged. The results obtained from this test are $E'$, $E''$, and $\tan(\delta)$ where:
• $E'$ is the elastic or storage modulus, the contribution of the elastic solid-like behavior to the complex modulus.

• $E''$ is the viscous or loss modulus, the contribution of viscous fluid-like behavior to the complex modulus.

• $\delta$ is the phase angle, the phase difference between the measured stress and the applied strain. $\delta$ can be obtained from a plot similar to that shown in Figure 2-11, using (2.2).

\[
\delta = 2\pi \frac{\text{Time delay}}{\text{Time period}} \tag{2.2}
\]

Its tangent is $\tan(\delta) = \frac{E''}{E'}$. For polymers, $\tan(\delta)$ is at its maximum at the transition from the glassy state to the rubbery state.

Figure 2-11: Generic DMA stress and strain.

In Figure 2-12, we see the $E'$ dropping to half of its magnitude, from room temperature to 100°C and an increasing $\tan(\delta)$. $E'''$ is much smaller than $E'$, which is why there is no visible change. Upon closer inspection, however, the value of $E'''$ increases from 87MPa to 140MPa at 90°C and then decreases again to 130 MPa at 100°C.
2.3 A Mechanical Characterization of PZT Fibers

2.3.1 Tensile Experiment

The ASTM Standard Test Method for Tensile Strength and Young’s Modulus of Fibers C1557 [90] recommends gluing the fiber on a mounting tab. When this method was used, however, the fibers broke at the bonding point, either during the specimen’s installation or at the beginning of the experiment. The DMA’s grips are designed for fibers; as shown in Figure 2-14, the grips have threads parallel to the loading direction where the fibers can be placed to ensure good alignment. Mounting the fibers directly on those grips solves the bonding issues. In addition, the grips use two screws to tighten the sample and

Figure 2-12: Temperature ramp DMA results for AFC.
apply uniform pressure on it. The grip displacement is the source of strain in the tensile experiment. In order to make sure that this experimental setup does induce extra strain due to the slippage of the sample, tensile tests were conducted on specimens of different gage lengths (20, 40 and 50 mm). The stress–strain curves of these tests on fibers are shown in Figure 2-14. The curves are on top of each other at lower strain which is the portion used to evaluate the elastic modulus. At higher strains, small differences are observed on the curves are random and not related to the gage length of the specimen however they are due the difference of the specimens used. This experiment confirms that there is slippage at the grips and that there is not an induced strain. The gage length chosen for the rest of the experiments for the fibers is 40 mm to maximize displacement and to fit the environmental chamber for different temperature experiments.

The unpoled fibers were subjected to uniaxial tensile with different displacement rates (0.2, 0.04, and 0.004 mm/s) at different temperatures (25°C, 50°C, and 75°C) in order to study temperature- and rate-dependence. The displacement rates were chosen to match the strain rates used while testing AFCs (31.25, 6.25 and 0.625 %/min).
Figure 2-13: DMA grips.

![DMA grips](image)

Figure 2-14: Stress–strain curves of different gage lengths PZT fibers.

Figure 2-15 presents the typical stress–strain behavior of PZT fibers at room temperature (each curve corresponds to a strain rate). Independently of the strain rate used, we noticed a non-linear behavior typical of PZT ceramics. This non-linearity finds its origin in ferroelasticity, which consists of an additional elongation due to domain switching. In
order to estimate this non-linearity, a threshold $\sigma_{\text{prop}}$ (proportional limit) is used to mark the beginning of the non-linear portion.

This limit for domain switching has also been used by Guillon et al. [51]. Dittmer et al. [91] defined $\sigma_{\text{prop}}$ as the stress point at which the discrepancy from linear behavior exceeds 5%. As shown in Figure 2-16, the proportional limit was found to range between 15MPa and 30MPa. More interestingly, $\sigma_{\text{prop}}$ was found to decrease with temperature. In other words, non-linear behavior was found to be more prominent at high temperatures.

It is reasonable to assume that the ALCERU® process contributes to this “plastic-like” behavior. In fact, the ALCERU® process involves some organic binders that might be contributing to the non-linear behavior in addition to ferroelasticity. To the best of our knowledge, no previous work has studied the temperature dependence of the proportional limit for PZT fibers under tensile testing. Rossetti et al. [92] estimated the end of the linear portion in the stress–strain curve of PZT fibers at 18 MPa but the authors did not
mention the temperature of the experiment. Cao and Evans [12] found that non-linearity begins beyond 20 MPa, but this was for bulk PZTs.

![Figure 2-16: Proportional limit $\sigma_{prop}$ for different temperatures.](image)

The other properties obtained for the stress-strain curves are elastic modulus, tensile strength, and strain at failure. Figure 2-17 shows the elastic modulus as a function of temperature for different strain rates. The elastic modulus of the piezoelectric fibers does not show a rate-dependence except for high temperatures (75°C). On the other hand, a significant temperature-dependence appears.
Figure 2-17: Elastic modulus for different temperatures and different strain rates

Figure 2-18 and Figure 2-19 show the tensile strength and strain at failure of the tested fibers for different temperatures and different strain rates. Interestingly, these mechanical properties are rate-dependent. This time-dependence is peculiar to ferroelastic materials [89]. At 75°C and with a strain rate of 0.2 mm/s, PZT fibers exhibit a tensile strength that reaches 80 MPa and a strain at failure of 0.6 %, which is in agreement with Dittmer et al. [91].
The elastic modulus values of PZT fibers are lower than the typical modulus of bulk PZT, which is around 60 GPa. Most literature values refer to bulk samples that have an elastic modulus that ranges from 40 to 77.5 GPa [91]. Part of this difference may be due to the microscopic porosity observed in the piezoelectric fibers organic binders residues. The process used to manufacture and thermally treat the piezoelectric fibers apparently
generates more microscopic porosity. A Scanning Electron Microscopy (SEM) was performed on the surface of a non-tested fiber Figure 2-20. The fibers grain size shows a wide distribution. In fact, grain size range from 0.8 to 3 µm. We also notice the presence of micro defects consisting of pores having sizes up to 6 µm and representing approximately 5% of the cross-section. These defects are probably linked to the manufacturing process since air may be trapped into the spin mix. It is worthwhile to mention that some notches are present on the outer surface of the fiber. These notches may reduce the effective cross-section area of the fiber and give rise to stress concentration and thus earlier failure.

![SEM of PZT fiber’s cross-section.](image)

**2.3.2 Creep and Relaxation**

In order to further investigate the time- and temperature-dependence, we studied the mechanical response of the fibers under creep and stress relaxation. The RSA G2 instrument was used for these experiments. The samples were unpoled. For creep,
different stress thresholds were applied (10 MPa, 20 MPa, and 30 MPa) and held for approximately 1h at different temperatures (25°C, 50°C, and 75°C). The stress dependence observed during the creep experiments indicate that in addition to a local saturation mechanism, there is a global saturation mechanism that may originate in the contribution of extrinsic (binder) elements that are involved with the ALCERU® process.

Figure 2-21: PZT fibers creep at room temperature.

Figure 2-22: PZT 5A creep at different temperatures.
Stress relaxation was performed on PZT fibers under three different maximum strains: 0.1%, 0.2%, and 0.3%. It is seen that increasing the strain level results in more pronounced relaxation responses. The initial stress values corresponding to the maximum strains are within the stress levels obtained from the tensile testing as detailed above.

![Stress relaxation of PZT fibers at room temperature.](image)

Figure 2-23: Stress relaxation of PZT fibers at room temperature.

### 2.3.3 Cyclic Loading

In order to quantify the impact of cyclic loading on the behavior of the material, repeated loading and unloading cycles were performed under different stress levels. Figure 2-24 shows an example of the stress-strain curve of cyclic loading made of three cycles. The fiber was under tension with a constant displacement rate until the stress reaches the desired value and then the fiber is unloaded with the same displacement rate until the stress reaches zero. The elastic modulus was evaluated at the beginning of each loading cycle; these values are reported in Figure 2-25. At 15 and 40 MPa stress, the elastic
modulus remained almost unchanged during the different loading cycles. However, at 30 MPa a noticeable increase of 30% was recorded. Under a low stress level (15 MPa), the domain switching is not considerable enough to see a consequent jump in elastic modulus values from cycle to cycle. However when the load is higher, the domain switching i.e ferroelasticity start affecting the stiffness of the material.

![Stress-strain curve of cyclic loading on PZT fibers.](image)

Figure 2-24: Stress-strain curve of cyclic loading on PZT fibers.

Nevertheless, the stiffening mechanism due to ferroelasticity is less pronounced at very high stress (e.g., 40 MPa) due to damage initiation, which competes with the stiffening effect of ferroelasticity. In order to confirm this behavior, we added three additional stress levels (20, 25, and 35 MPa) and plotted the % increase of the elastic modulus between the first and second cycles. From Figure 2-26 we notice that this percentage
becomes considerable starting at 20 MPa and reaching a maximum around 30 MPa. This leads us to conclude that optimal mechanical poling occurred between these two values. Above 30 MPa, the observed mechanical poling is less pronounced due to damage. The values of stress for which ferroelasticity affect the mechanical response is in agreement with the estimated values of $\sigma_{\text{prop}}$ extracted from tensile testing as explained above.

Figure 2-25: Elastic modulus evolution with loading cycles.

Figure 2-26: Elastic modulus % increase for different stress levels.
2.4 A Mechanical Characterization of Epoxy

Since Flex-A-Bond/740 epoxy is not commercially available, United Resin Corp provided us with a 400 g free sample. Due to this limited quantity, only selected mechanical tests were conducted on the epoxy. The epoxy was prepared as recommended by the manufacturer. The resin and the hardener were mixed in equal weight ratios, and then the mix was degassed under vacuum for 30 minutes. The degassed solution was then poured into silicone molds and cured at room temperature. For tensile experiments, the mold was chosen to conform with the ASTM Standard Test Method for Tensile Properties of Plastics D638-10 [93]. The ASTM standard-dictated dimensions are shown in Figure 2-27.

Figure 2-27: Dimensions of ASTM D638 Type IV in mm.
2.4.1 Tensile Experiment

The focus for the tensile experiments of epoxy is limited to temperature impact because of the scarcity of epoxy material. Tensile experiments were conducted at the three different temperatures used for previous tensile experiments: 25°C, 50°C, and 75°C. The strain in these experiments was measured using a clip-on Instron extensometer with a gage length of 25mm. This strain measurement worked well for room temperature, but at higher temperatures the extensometer blades used to attach the specimen caused early failure. Figure 2-28 shows the stress–strain curves for the tensile experiments at three different temperatures with a displacement rate of 1.6mm/min (strain rate =6.25%/min). The tensile strength and the modulus decrease with an increase of temperature. No conclusions can be made concerning the strain at failure because at high temperatures the sample softens and the extensometer blades penetrate the sample, resulting in early failure.
Three specimens were tested at each temperature, and the Young’s modulus and tensile strength were extracted from the stress–strain curves. In some of points plotted in Figure 2-29 and Figure 2-30 the error bars are not visible because of the small standard deviation compared to the scale. The Young’s modulus decreased by one order of magnitude at 50°C compared to 25°C; this decrease is due to the softening of the epoxy and the transition from a glassy state to a rubbery one. The temperature of transition between both states is called glass transition temperature $T_g$. The modulus dropped by 78% from 50°C to 75°C, which means that the $T_g$ of this epoxy is around 50°C (Figure 2-29). The tensile strength decreased by 70% between 25°C and 50°C and by 75% between 50°C and 75°C as shown in Figure 2-30. These measurements confirm that the strength and toughness of the epoxy greatly decrease with temperature, and this behavior explains the results of the AFC tensile experiments discussed in the previous sections.

Figure 2-28: Stress–strain curves for epoxy tensile at different temperatures.
Figure 2-29: Young’s modulus for different temperatures.

Figure 2-30: Tensile strength for different temperatures.
2.4.2 Dynamic Tests

To accurately evaluate the value of the epoxy’s $T_g$, a dynamic mechanical test was conducted under a temperature ramp across a wide range of temperatures (from -50°C to 130°C). $E'$, $E''$, and $\tan(\delta)$ are plotted in Figure 2-31 where we notice a two order of magnitude drop in $E'$ from room temperature to 50°C. We also notice that $\tan(\delta)$ reaches its maximum around 49°C. That means that the epoxy is moving from the glassy state to the rubbery, which explains the decrease of the epoxy’s mechanical properties (Section 2.4.1) and the AFCs’ at temperatures equal and exceeding 50°C (Section 2.2.1).

Figure 2-31: Temperature ramp DMA results for epoxy
2.5 Summary

In this work, mechanical properties of AFCs were determined experimentally at different loading conditions and temperatures. Tensile tests were conducted at three different temperatures and three strain rates. Our findings show that the composite properties are time and temperature dependent. At higher temperatures, the modulus and tensile strength decreased for all strain rates while strain at failure increased. The change in the mechanical properties with changes in temperature and strain rates is attributed to the viscoelastic nature of the epoxy surrounding the fibers and the weakening of the fiber/epoxy and epoxy/Kapton interfaces. These results were further confirmed by the experiments conducted on the epoxy; the drop in modulus and strength of AFCs at high temperatures is due to the drop of the epoxy’s mechanical properties. Above 50°C, the epoxy switched to the rubbery state which increased the loss modulus of epoxy and AFCs. As a result higher creep and relaxation responses were seen at high temperatures. An overview of the mechanical behavior of PZT fibers processed by the ALCERU® was given. For the first time for PZT fibers, non-linear mechanical behavior under tensile testing was investigated with respect to temperature, and creep and relaxation experiments were performed. The organic binders used during the ALCREU® may have a substantial effect on the non-linear mechanical behavior. Furthermore, we were able to quantify the amount of mechanical poling using cyclic loading and unloading testing. The study relating to creep shows a stress and temperature dependence and suggests the existence of an additional global saturation mechanism. We can conclude then that creep and stress relaxation results obtained for AFCs were affected both by the behavior of
epoxy and the behavior of PZT fibers. It was also noticed that some of the mechanical properties of the PZT fibers were different from what was reported in literature for bulk PZT 5A while others were similar. For example the elastic modulus of fibers was varying between 25 and 40 GPa, however it varies between 40 and 77 GPa for bulk [91]. Proportional limit for PZT fibers however was similar to what was reported for bulk PZT [12]. Some of the properties obtained for epoxy and PZT fibers will be used as inputs when modeling the AFCs. Other results can be used as reference for future modeling of mechanical non-linearity and time dependence of piezoelectric fibers.
Chapter 3

An Electrical and Electro-Mechanical Characterization of AFC and its Constituents

Apart from mechanical behavior, AFCs exhibit ferroelectric behavior, which makes the characterization of their electrical and electro-mechanical properties very important. In the previous chapter, it was shown that the mechanical behavior is time- and temperature-dependent. It remains, however, to characterize the ferroelectric and electro-mechanical properties of AFCs, specifically, the coercive electric field ($E_c$), remnant polarization ($P_r$), saturation polarization ($P_s$), and the piezoelectric strain coefficient $d_{33}$. AFCs are subjected to sinusoidal electric fields at different frequencies, namely 0.2, 1, and 5Hz. These frequencies are all low in order to stay within a pseudostatic regime to avoid resonance and Joule heating. These experiments are also done under different isothermal temperatures, namely 25, 50, and 75º C for the same reasons stated in Chapter 2, which is to be within the operational temperature range of AFCs.

3.1 Materials and Equipment

3.1.1 Materials

Three different materials have been used for electrical and electro-mechanical testing: AFCs, PZT 5A fibers and PZT 5A plates. The AFCs and epoxy used here are the same as
described in Chapter 2. PZT fibers were purchased from Advanced Cerametrics, Inc. These fibers are the same fibers used to manufacture AFCs and have a diameter of 0.25 mm. PZT 5A plates were purchased from Omega Piezo with the dimensions 38.12 mm x 3.825 mm x 0.55 mm.

3.1.2 Equipment

Several measurement setups are used for electrical and electro-mechanical measurements. The experimental setups used for measuring AFCs and bulk PZT wafers cannot be used for PZT fibers because of their geometry, brittleness, and low capacitance.

The electrical measurements of AFCs and bulk PZT were completed in a silicone oil bath as a dielectric medium to avoid shorting and arcing due to the high voltage. The sample was connected to a voltage generator through a high voltage power amplifier and the circuit was connected to an oscilloscope to measure the current and the voltage as shown in Figure 3-1. The data was collected on a laptop using GPIB cable and Labview program. The current was then integrated with respect to time using a numerical function on Matlab before being divided by the surface area crossed by charges to obtain the electric displacement, $D$. 
The electric displacement is given by \( D = \varepsilon_0 \varepsilon_r E \). The polarization \( P \) is given by

\[
P = \varepsilon_0 (\varepsilon_r - 1) E.
\]

Since for PZT 5A, \( \varepsilon_r \) is around 1800, the values of \( D \) and \( P \) are very similar. The definition of the surface area that the charges are crossing in parallel electrodes set up is simply the area of the electrode. However, for the interdigitated electrodes, the electric field is not uniformly distributed in the fibers: depending on the electrodes distance and the fiber diameter, the charges are collected on the top and bottom of AFCs by the finger electrodes (Figure 3-2). In this study, this surface area is the sum of the cross sections of AFC multiplied by the number of electrode gaps.

Figure 3-1: Experimental setup for electrical characterization of AFCs and bulk PZT.

Figure 3-2: A schematic of electric field distribution inside an AFC.
Because of the geometry and small capacitance of a single fiber, it was not possible to use the same setup that we used for the AFCs. The current generated from one fiber is within the noise level generated by the amplifier’s current output. In addition, the small diameter of the fibers made it very difficult to attach any electrodes. In order to overcome these challenges, two methods were used to characterize the PZT fibers. The first consisted of printing IDE electrodes on Kapton sheets and using them to apply the field and collect charges as shown in Figure 3-3. PZT fibers were placed between two sheets of Kapton and a vacuum was applied to ensure contact between the fibers and the electrodes. The current was measured and integrated with respect to time, and then it was divided by the sum of the cross section of the fibers multiplied by the number of electrode gaps to obtain electric displacement or D. The second method consisted of testing a single fiber of a length of 2.75mm, as in the setup shown in Figure 3-4. The short fiber is placed between two semi-spherical electrodes where a uniform electric field is applied on the fiber. The entire setup is submersed in Galden oil and then placed in an oven to control the temperature. The setup is connected to a Sawyer-Tower circuit to measure polarization and an LVDT to measure the displacement generated by the fibers.
For the electro-mechanical tests on the AFCs, the sample was placed in an environmental chamber for at least 10 minutes to ensure a uniform temperature. The sample is then connected to a voltage generator through a high-voltage power amplifier. The electric field generates a displacement in the sample, which was measured using a high-resolution fiber optic sensor (MTI 2100), as shown in Figure 3-5. The displacement and the voltage...
signals were both collected from the oscilloscope and the strain was plotted as a function of electric field.

3.2 Electrical Characterization of AFCs and PZT

3.2.1 Ferroelectric Properties

In order to obtain the most important electrical properties of ferroelectric materials we plot the polarization as a function of the electric field after applying a high amplitude sinusoidal electric field. In order to study the effect of the presence of epoxy and the IDE electrodes in AFCs, PZT plate samples will also be tested. In Chapter 2, it was shown that the mechanical properties of PZT fibers are different from those of bulk PZTs. For the electrical characterization, both PZT plates and PZT fibers will be compared to the properties of AFCs. Figure 3-6 shows the hysteresis loops for AFCs at different temperatures and frequencies. Increasing the frequency decreases the height of the loop.
(Figure 3-6 a). At low frequency, there is more time for the charges to be collected by the electrodes. Therefore, $P_r$ and $P_s$ increase with the decrease of frequency whereas $E_c$ is almost constant. We also notice an increase of $P_r$ and $P_s$ with a temperature increase as shown in Figure 3-6 b. The increase in those values is abrupt between 25 and 50ºC. To better analyze impact of temperature on the polarization, Figure 3-7 and Figure 3-8 illustrate change in $P_r$ and $P_s$ for AFC a), bulk PZT 5A b) and PZT 5A fibers c).

![Ferroelectric hysteresis loop of AFCs at different frequencies and temperatures](image)

$P_s$ results for AFCs are plotted in Figure 3-7 a; at room temperature the $P_s$ increases by 20% between 5 Hz and 1 Hz; however, between 1 Hz and 200 mHz, the increase was around 12%. Similar $P_s$ increase with frequency decrease was also seen for the temperatures 50º C and 75º C. The temperature also had an impact on the behavior of $P_s$; in fact, $P_s$ increased by 30% between 25º C and 50º C and partially decreased at 75º C. For bulk PZT plates, the results shown in Figure 3-7 b); higher $P_s$ was measured for lower frequencies, which is similar to the behavior seen for AFCs. $P_s$ also increases
slightly with an increase in temperature. The $P_s$ of bulk PZT 5A is almost twice the values of $P_s$ of AFC.

For the PZT fibers results shown in Figure 3-7 c), the data was more scattered and the error bars were wider. The sensitivity of the measurement setup used shown in Figure 3-4 and the possible bad alignment of the fiber with the electrodes are the main reasons for data scattering. Slightly higher values of $P_s$ were seen at low frequencies. The impact of temperature on PZT fibers is not as important as it was for AFCs; $P_s$ increased slightly with temperature. The $P_s$ of PZT fibers are higher than those of AFCs and bulk PZT values. This difference between PZT fibers and PZT plates, despite the same composition in both cases, may be due to different manufacturing processes and sintering temperatures, resulting in different microstructures and grain sizes. This could also explain the lower magnitude of $P_s$ in the PZT plates compared to PZT fibers. In AFCs, the presence of epoxy amplifies the frequency and temperature effects on the ferroelectric behavior. The remnant polarization results are plotted in Figure 3-8 and the impact of temperature and frequency is very similar to what was observed for the saturation polarization.
Figure 3-7: Temperature and frequency effects on $P_s$: a) AFC, b) PZT 5A plates, and c) PZT 5A fibers.
The coercive field results are plotted in Figure 3-9. For AFCs, $E_c$ does not show any dependence on the frequency, but shows a slight decrease with temperature. For PZT fibers and plates the coercive field is higher at higher frequencies. The orientation of dipoles and domain wall motion is a time-dependent process and this can explain the dependence of the coercive field on the frequency. It can be seen also that $E_c$ decreases with temperature for PZT fibers and plates; this can be explained by the increased mobility of dipoles, which facilitates poling and unpoling at higher temperatures. The temperature impact on ferroelectric properties of bulk PZT 5A is in agreement with experimental results obtained in the study conducted on different ferroelectric materials by Hooker [94].
The values of the coercive field in the AFCs are higher than the values of PZT fibers, though they are the same fibers that are used in AFCs. In AFCs, the electric field inside the PZT fiber is not uniformly distributed (Figure 3-2) because of the use of IDE electrodes. As a result the local electric field values along the longitudinal direction will be lower than the applied one. There is a thin layer of epoxy also separating the fibers from the electrodes fingers. The high contrast in dielectric properties between the epoxy
and the PZT results in charge dissipation and reduced electric potential at the fiber section under the electrodes. The non-uniformity of the electric field results in a lower local electric field inside the fibers compared to the applied field in the PZT fibers, which requires higher applied field to reach the coercive field inside the fibers. The low values of $P_r$ and $P_s$ in the AFCs compared to pure PZT are mainly caused by the low local electric field inside the fibers and by charge dissipation at the interface between PZT and epoxy.

The effects of temperature and frequency are more dominant in the AFCs because the polymer component represents more than 50% of their volume fraction. Electric field lines can penetrate the interface between matrix and fibers easier at higher temperature because the dielectric constant of the matrix increases by more than 25% when the temperature rises from 25°C to 50°C, thus reducing the dielectric mismatch between the matrix and fiber. AFC’s properties also show a frequency-dependent ferroelectric behavior that is analogous to the viscoelastic behavior discussed in the previous chapter. This viscoelastic like behavior for ferroelectric properties have been reported for bulk PZT in some studies [59, 95]. It was shown that the PZT materials exhibit creep-like behavior when a constant electric field is applied.

### 3.2.2 The Impact of IDE Electrodes

To better understand the impact of IDE electrodes, an electric field was applied to PZT fibers using the setup shown in Figure 3-3. The screen-printing process while preparing
the electrodes resulted in some imperfections in the electrodes, which lead to some arcing and limited the maximum possible field to be applied. The measured hysteresis loops were plotted for different frequencies and electric field magnitudes, as shown in Figure 3-10. The loops are very lossy. The impact of frequency is similar to what has been seen for the parallel electrode setups. In Table 3-1, the properties obtained from these loops are compared with properties obtained in 3.2.1. We notice that the coercive field values are much higher than in the PZT fiber results with a parallel electrodes setup, and are closer to the results obtained from AFCs. The $P_s$ obtained from the IDE setup is almost one-third of what was found for a parallel setup. Therefore, it can be concluded that the high coercive field and the low polarization in the IDE setup are due to the non-uniform field and low local electric field inside the fibers. In AFCs, in addition to the previous two reasons, the epoxy interface separating the electrodes from the PZT fibers also further lowers the polarization and increases the Ec.

Figure 3-10: Ferroelectric hysteresis loop of PZT fibers at: a) different electric field
magnitude and b) different frequencies.

Table 3-1: Comparison of ferroelectric properties for different experimental setups.

<table>
<thead>
<tr>
<th>Property</th>
<th>$P_s$(C/m$^2$)</th>
<th>$P_r$(C/m$^2$)</th>
<th>$E_c$(MV/m)</th>
<th>Applied electric field (MV/m)</th>
</tr>
</thead>
<tbody>
<tr>
<td>AFC</td>
<td>0.14</td>
<td>0.11</td>
<td>1.4</td>
<td>2.5</td>
</tr>
<tr>
<td>PZT fiber (parallel electrodes)</td>
<td>0.34</td>
<td>0.21</td>
<td>1.02</td>
<td>2.5</td>
</tr>
<tr>
<td>PZT fiber (IDE)</td>
<td>0.14</td>
<td>0.11</td>
<td>1.7</td>
<td>2.1</td>
</tr>
</tbody>
</table>

It is not technically possible to measure the local electric field inside the fibers; therefore Abaqus is used to model the fiber/electrode/epoxy and investigate the electric field distribution. The model consists of a cylinder (representing the fiber) surrounded by air, as can be seen in Figure 3-11. The purpose of this model is to estimate the local electric field inside the fibers and to compare the homogeneous dielectric constant between parallel and IDE electrodes. With IDE electrodes, the electric field is not uniform in magnitude or direction. Figure 3-12 a shows the results of the electric field distribution, where all the arrows are represented using the same length independently of magnitude for better visualization. Under the electrodes, the electric field arrows are aligned in the $y$ direction toward the electrodes. In order to investigate the impact of IDEs on the magnitude of the electric field in the longitudinal direction, the longitudinal component of the electric field at the center of the fiber was plotted as a function of $z$ in Figure 3-12 b. At the center of the fiber, electric field can reach only 0.7MV/m, though the applied electric field is 1MV/m. Under the electrodes the electric field component can go as low
as 0.1\text{MV/m}. The homogeneous dielectric constant was calculated from the model by adding the positive reactive charges at the electrodes and divided by the cross section of the fiber and the applied electric field. The homogeneous dielectric constant of the model with IDE electrodes is around 940; however, it is 1700 for the parallel electrodes. This could explain the lower polarization and high coercive field observed in the experiments with IDE electrodes.

Figure 3-11: Abaqus model for PZT fibers.
3.3 Electro-Mechanical Characterization of AFCs and PZT Fibers

For the electro-mechanical characterization, only the linear region of piezoelectricity is investigated by applying electric fields less than half the value of the coercive field in order to avoid polarization switching. Steady state electro-mechanical loops of AFCs are
plotted in Figure 3-13 to observe the impact of frequency and temperature change on the hysteresis. Though the loops are very close to one another, a decrease in the slope of the loop can be observed when increasing the frequency. Figure 3-14 shows the loops for different electric fields where the slope is smaller for minor loops. The piezoelectric coefficient can be divided to intrinsic part related to ionic response and extrinsic part related to ferroelectric domains. The intrinsic part is independent of the electric field, however the extrinsic part does depend on the magnitude of the electric field and is caused by vibration, translation and switching of ferroelectric domains. For electric field lower than the coercive as in our case, the extrinsic contribution will be caused only by domains vibration and translations [95, 96]. For AFCs, when the electric field is higher, more electric field lines can go through the center of the fiber, thus increasing the actuation performance of AFCs.

![Graphs showing hysteresis loops](image)

Figure 3-13: Electro-mechanical hysteresis of AFCs at: a) different frequencies at 25°C and b) different temperatures at 1Hz.
In order to better understand the electro-mechanical behavior of AFCs, the $d_{33}$ coefficients were plotted for different electric field magnitudes, frequencies, and temperatures in Figure 3-15 for AFC and in Figure 3-16 for PZT fibers. The $d_{33}$ coefficients were obtained by evaluating the linear slope of the loop. For AFCs, these coefficients increase linearly with the magnitude of the electric field. The slower the frequency, the larger the coefficient; this can be explained by the frequency dependence of the PZT fiber response (Figure 3-16) and the viscoelastic behavior of the epoxy, which increases under longer loading durations. This behavior is more pronounced at higher electric fields where the stress presence in the fibers is higher. The $d_{33}$ coefficients for AFCs increase when the temperature increased from 25°C to 50°C, however they remained almost constant when the temperature increased from 50°C to 75°C. For PZT fibers, $d_{33}$ coefficients increase linearly with temperature, which follows the trend of polarization. For AFC, apart from the temperature impact on the fibers, the temperature increase softens the matrix, which reduces the constraints on the fibers’ elongation. From
25°C to 50°C, the matrix transitions from the glassy to the rubbery state (Temperature > $T_g$) and its elastic modulus drops by more than one order of magnitude. Above 50°C, the elastic modulus starts to reach a constant value as seen in DMA results of epoxy shown in Chapter two. This can be the reason for not observing a change in the $d_{33}$ coefficients for AFCs when the temperature increased from 50°C to 75°C. When the piezoelectric coefficients of AFCs and PZT fibers are compared, we notice that the AFCs’ coefficients are less than half of PZT fibers’ coefficients. The reduced strain in AFCs is due to the low local electric field inside the fiber (non-uniform field and dielectric mismatch at the interface epoxy/fiber) and to the constraint subjected on the fibers by the epoxy.

Figure 3-15: $d_{33}$ coefficients of AFC for: a) different frequencies and electric fields at 25°C and b) different temperatures and electric fields at 1 Hz.
In this chapter, an experimental study was conducted on the ferroelectric and electro-mechanical response of AFCs, bulk PZT and PZT fibers with a focus on temperature and time dependence. It was found that the remnant and saturation polarizations increase with the decrease of frequency and increase of temperature for AFCs and pure PZT (bulk and fiber). The coercive field decreased with temperature for the three types of materials tested. The coercive field for bulk PZT fiber and bulk PZT increased with the increase of frequency. The values of polarization in AFCs were smaller than the ones measured for bulk PZT and PZT fibers because of the lower local electric field inside the PZT fibers. The non-uniform electric field distribution and the epoxy layer separating the electrodes from the fibers are the reasons for the low electric field inside the fibers. As to the electro-mechanical measurements, $d_{33}$ increased with temperature increase and frequency.

Figure 3-16: $d_{33}$ coefficients PZT fibers for: a) different frequencies and electric fields at $25^\circ C$ and b) different temperatures and electric fields at 1 Hz.

3.4 Summary

In this chapter, an experimental study was conducted on the ferroelectric and electro-mechanical response of AFCs, bulk PZT and PZT fibers with a focus on temperature and time dependence. It was found that the remnant and saturation polarizations increase with the decrease of frequency and increase of temperature for AFCs and pure PZT (bulk and fiber). The coercive field decreased with temperature for the three types of materials tested. The coercive field for bulk PZT fiber and bulk PZT increased with the increase of frequency. The values of polarization in AFCs were smaller than the ones measured for bulk PZT and PZT fibers because of the lower local electric field inside the PZT fibers. The non-uniform electric field distribution and the epoxy layer separating the electrodes from the fibers are the reasons for the low electric field inside the fibers. As to the electro-mechanical measurements, $d_{33}$ increased with temperature increase and frequency.
decrease for AFCs and PZT fibers. For AFCs, the frequency and temperature dependence are due to the behavior of the PZT fiber and also to the viscoelastic behavior of the epoxy that is constraining the PZT fibers. The electro-mechanical behavior of the AFCs and PZT fibers was non-linear and depended on the magnitude of the applied electric field. The obtained results in this chapter will guide us in the modeling work conducted in the next chapters. In addition, the experimental procedures discussed in this chapter will be used to calibrate some of the materials parameters and properties that will be used for the modeling of AFCs and PZT fibers.
Chapter 4
A parametric study on flexible electro-active composites: importance of geometry and matrix properties*

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In the previous chapter, we established that most of the properties of AFCs depend on the local electric field inside the fiber. One way to evaluate the AFCs’ properties based on the local field is by modeling the composite. Several studies focused on modeling AFCs, some of them to obtain homogenized properties in order to predict and validate system level performance while the rest focused on AFC design parameters that can improve actuation or stress distribution, however, none of the studies, to the best of our knowledge, considered combining all design parameters related to the fibers, IDE geometry and epoxy matrix, the latter being generally neglected in all of them. In addition, the opposite poling of the PZT fiber along its length due to adjacent electrodes was not investigated either. For an optimal design of AFCs, there needs to be a parametric study on the design of the composites while simultaneously studying the effect of IDE geometry, and presence of epoxy layer between electrode and PZT fiber. It is especially important to consider the IDE electrode geometry and the epoxy surrounding the fibers together because they are inter-dependent where the change in one affects the behavior of the other components. In our study, IDE electrode setup will be used to realistically model the non-uniform electric field present in the PZT fiber in a real AFC; the electric field distribution inside the fiber is very important because it is a major factor in actuation. In addition, we use three electrode gaps for better observation of field distribution and calculation of homogenized dielectric constant. We will also focus on the
epoxy matrix and its properties, since the presence of this polymer interface between the electrodes and the fibers will affect the electric field reaching the fibers due to its low dielectric constant. Furthermore, for a better understanding of the mechanism of actuation for different parameters, we will use an RVE with four fibers and three electrode gaps with the presence of polymer matrix around the fibers. Four fibers are used in the RVE to visualize electric field and stress distribution without the effect of the boundary faces. The parametric study will allow not only varying the geometry but also studying the matrix materials properties effect on the behavior of the composite.

4.1 Modeling Procedure

4.1.1 Geometry and material modeling

The focus of this chapter is on actuation performance of the AFC. Most of the previous work done on AFC considered only quarter of fiber or a fiber with only one electrode gap and used uniform electric field lines crossing the PZT fibers. The RVE that we considered in this work is a representative part of the whole AFC. It is composed of four adjacent fibers embedded in epoxy and the composite is placed between four equidistant electrode fingers in the top and the bottom sides. The volume fraction of the fibers is 50%. The volume fraction and the dimensions of the RVE were based on analysis of micrographs taken by SEM of the cross section of commercial AFCs shown in Figure 4-1.
The modeling was done using Abaqus software by creating one part in the shape of a Cuboid and then using the partition tool to divide it to different components. This method provides common nodes at the interfaces between the components, so there will be perfect bonding between fibers-matrix and matrix-electrodes (the perfect bonding is in all degrees of freedom, i.e., electric potential and displacement continuity).

There are three different components, which are the electrodes, the fibers and the matrix. As shown in Figure 4-2, the RVE includes three different electrodes gaps with four electrodes on top and four on the bottom. The electric field is simulated by applying different electric potentials, where some of them are positive and the others are negative or zero. The electrodes at the edges have half width to take into consideration the symmetry and continuity of the AFC. The real thickness of the electrodes is less than 100 nanometers out of around 300 micrometers (total dimension of electrodes plus epoxy plus PZT fibers), and they are usually made of copper or silver. Due to this negligible relative
thickness, the electrodes will not affect the mechanical behavior of the AFC, which is why their presence is ignored in terms of impact on mechanical properties by assigning them matrix mechanical properties. They appear solely in the way the electric potential boundary conditions are defined. Note that in this case, the electrodes are modeled with a thickness of 10 micrometers for easier meshing due to the complexity of the geometry.

The fibers are modeled as cylinders embedded in the epoxy; to take into consideration the opposite poling, the fibers themselves were partitioned using the cutting plane tool at half the electrode width. To model opposite poling conditions, fiber sections in the poling direction will have the properties of PZT while others will have properties of PZT with opposite piezoelectric coefficients. The electrodes are placed 2 micrometers from the fibers to take into consideration the epoxy interface and its impact on the behavior of AFCs. The matrix component will then fill the rest of the space around the fibers and the

Figure 4-2: Different components of AFC after partition.
electrodes. The aim of the current work is to evaluate the impact of parameter variations on the actuation performance; to do that we will start with a nominal case with specified geometry and materials properties and then we will vary one parameter at a time, using a 50% fiber volume fraction. The nominal case geometry was chosen based on SEM of commercially available AFCs (Figure 4-1). The stiffness matrix and piezoelectric tensor for PZT 5A are shown in equations (4.1) and (4.2), and the rest of material properties and dimensions of the nominal case are shown in Table 4-1 [98]. In the current chapter, we use a linear piezoelectric material model for the PZT and linear dielectric and linear elastic models for the epoxy matrix.

$$
K = \begin{bmatrix}
1.2035 & 0.7518 & 0.7509 & 0 & 0 & 0 \\
0.7518 & 1.2035 & 0.7509 & 0 & 0 & 0 \\
0.7509 & 0.7509 & 1.1087 & 0 & 0 & 0 \\
0 & 0 & 0 & 0.2105 & 0 & 0 \\
0 & 0 & 0 & 0 & 0.2105 & 0 \\
0 & 0 & 0 & 0 & 0 & 0.2105 \\
\end{bmatrix} \cdot 10^9 \left( \frac{N}{m^2} \right) \quad (4.1)
$$

$$
d = \begin{bmatrix}
0 & 0 & 0 & 0 & 584 & 0 \\
0 & 0 & 0 & 584 & 0 & 0 \\
-171 & -171 & 374 & 0 & 0 & 0 \\
\end{bmatrix} \cdot 10^{-12} \left( \frac{C}{N} \right) \quad (4.2)
$$

Table 4-1: List of geometric parameters and material properties for the nominal case.

<table>
<thead>
<tr>
<th>Geometric parameter</th>
<th>Value</th>
</tr>
</thead>
<tbody>
<tr>
<td>Electrodes gap (µm)</td>
<td>500</td>
</tr>
<tr>
<td>Electrodes width (µm)</td>
<td>125</td>
</tr>
<tr>
<td>Fibers diameter (µm)</td>
<td>250</td>
</tr>
<tr>
<td>Inter-fiber distance (µm)</td>
<td>30</td>
</tr>
<tr>
<td>Property</td>
<td>Value</td>
</tr>
<tr>
<td>----------------------------------------------</td>
<td>---------</td>
</tr>
<tr>
<td>Overall thickness (µm)</td>
<td>350</td>
</tr>
<tr>
<td>Matrix dielectric constant $\varepsilon_m$</td>
<td>3.75</td>
</tr>
<tr>
<td>Fibers dielectric constant $\varepsilon_{11}, \varepsilon_{22}$</td>
<td>1730</td>
</tr>
<tr>
<td>Fibers dielectric constant $\varepsilon_{33}$</td>
<td>1700</td>
</tr>
<tr>
<td>Matrix Poisson’s ratio</td>
<td>0.3</td>
</tr>
<tr>
<td>Matrix Young’s modulus $Y_m$ (GPa)</td>
<td>1.5</td>
</tr>
<tr>
<td>Matrix piezoelectric coefficients $d_{ij}$ (m/V)</td>
<td>0</td>
</tr>
<tr>
<td>Fibers mechanical properties</td>
<td>See Eq. (4.1)</td>
</tr>
<tr>
<td>Fibers piezoelectric properties</td>
<td>See Eq. (4.2)</td>
</tr>
</tbody>
</table>

### 4.1.2 Boundary conditions

To assess the actuation capability, effective $d_{33}$ of the AFC is quantified, where the electric field is applied and the resulting displacement at one end is ascertained while constraining the other end. The overall strain is then divided by the electric field to obtain the piezoelectric coefficient $d_{33}$. The electrical boundary conditions applied to measure the piezoelectric coefficient are 0V potential at alternating electrodes above and below the fibers and a positive potential $V$ at the other electrodes. In the RVE, there are four zero potential electrodes (blue in Figure 4-3 a) and four positive potential electrodes (red in Figure 4-3 a). The mechanical boundary conditions are set in order to block the displacement in one side and leave it free in the opposite side. The displacement will be blocked in three sides as shown in Figure 4-3 b: surface $(0,y,z)$, surface $(x,0,z)$ and
surface (x,y,0). The prescribed displacement for the selected sides can be described as follows:

- At surface (0,y,z): $U_x = UR_y = UR_z = 0$;
- At surface (x,0,z): $U_y = UR_x = UR_z = 0$;
- At surface (x,y,0): $U_z = UR_x = UR_y = 0$.

4.2 Parametric Study

The performance of the AFCs depends on several parameters, some of them are related to the geometry, and others are related to materials properties of epoxy matrix and PZT fibers. In this study, we are focusing on those parameters that can be modified when manufacturing AFCs (see Table 4-2). The matrix is assumed as linear elastic and isotropic.
Table 4-2. List of parameters to be varied.

<table>
<thead>
<tr>
<th>Parameter</th>
<th>Range of values</th>
</tr>
</thead>
<tbody>
<tr>
<td>Electrodes gap</td>
<td>0.1-1 mm</td>
</tr>
<tr>
<td>Electrodes width</td>
<td>0.125-0.4 mm</td>
</tr>
<tr>
<td>Matrix Young’s Modulus</td>
<td>0.1-3 GPa</td>
</tr>
<tr>
<td>Matrix Dielectric Constant</td>
<td>4-1400</td>
</tr>
</tbody>
</table>

4.2.1 Impact on homogeneous elastic modulus and dielectric constant

The first AFC property studied is its Young’s modulus along the fiber longitudinal axis, $Y_{AFC}$; it was plotted in Figure 4-4 as a function the matrix Young’s modulus, $Y_m$. The modulus of the composite $Y_{AFC}$ increased only by 7% when $Y_m$ increases by almost 30 times. We can conclude that the matrix variation will not have a big impact on the modulus of the composite. However, results in Figure 4-5 show that the AFC’s dielectric constant, $\varepsilon_{AFC}$ is very dependent on the matrix dielectric constant, $\varepsilon_m$, in a non-linear manner. It is noted that $\varepsilon_f$ refers to the PZT fibers’ dielectric constant.
Figure 4-4: AFC axial modulus for different values of the matrix modulus.

Figure 4-5: AFC’s dielectric constant for different values of the matrix dielectric constant.

4.2.2 IDE geometry impact on actuation

Figure 4-6 shows the plots of $d_{33}$ coefficient as a function of electrode gap and width. The piezoelectric coefficient increases linearly with the electrode gap. However, for the electrode width, the variation is not linear where an inflection point occurs at around 250 micrometers, which represents the fiber diameter. In the above analysis, the modulus and dielectric properties of the matrix are 1.5 GPa and 3.75, respectively. To better
understand the dependence of the piezoelectric coefficient on the electrodes gap, the electric flux was plotted in Figure 4-7. It is seen that for the short electrode gap, only a small portion of the electric field lines are in the direction of the fiber whereas for the longer electrode gap, a bigger portion of the electric field lines is parallel to the fibers, which increase the local longitudinal electric field inside the fibers. Figure 4-8 displays the electric potential for different electrode width as an attempt to better understand the inflection point seen in Figure 4-6.

Figure 4-6: \( d_{33} \) as a function of electrode gap and width.

Figure 4-7: Electric flux orientation for narrow and wide electrode gap: a) 0.1 mm and b)
1 mm.

Figure 4-8: Electric potential distribution for narrow and wide electrode fingers: a) 0.125 mm and b) 0.5 mm.

Figure 4-8 a) shows that the electric potential does not reach the middle of the fiber for the low electrodes width, i.e., the fiber mostly acts as a passive material since a major portion of it does not experience the electric potential. However, for the larger width (Figure 4-8 b)) there is a better distribution of the electric potential under the electrodes. The penetration of the electric potential does not depend only on the electrode width, but it also depends on the fiber diameter, which explains the jump in the piezoelectric coefficient when the value of the electrode width is close to the fiber diameter (Figure 4-6).

4.2.3 Matrix properties impact on actuation

The design of the IDEs has a big impact on the actuation performance of AFCs; this impact is measured by taking into consideration the presence of a polymer matrix around the fibers. In the next part of this parametric study we will assess the impact of the polymer matrix properties on the performance of AFCs. The coefficients that we vary are
the elastic modulus and dielectric constant, as shown in Figure 4-9. The matrix elastic modulus, which has a very important impact of the mechanical properties of the AFCs, does not have a measurable impact on the piezoelectric coefficient. As we vary the modulus from 100 MPa to 3 GPa, the $d_{33}$ coefficient is almost constant. The reason is due to the high modulus of the PZT fibers that is at least 20 times higher than that of the matrix. On the other hand, the variation of the dielectric constant has a significant impact on the actuation capability of AFCs. The dielectric mismatch between the matrix and the fibers prevents the electric potential from crossing the interface between them, and instead concentrates the electric field in the polymer. The local electric field inside the fibers is almost a tenth of that applied in the epoxy matrix. When the dielectric constant of the matrix is increased, the effective $d_{33}$ coefficient dramatically increases in response (Figure 4-9 b)). As the dielectric ratio $\varepsilon_m/\varepsilon_f$ reaches then exceeds 0.1, the increase in the $d_{33}$ coefficient becomes more gradual and finally plateaus. To explain the reason for the $d_{33}$ behavior, Figure 4-10 shows the effect of a high dielectric mismatch a) and a low dielectric mismatch b) on the electric potential distribution. The potential distribution for the low dielectric constant is found only around the electrode area; whereas for high dielectric constant we see a more uniform potential distribution reaching the center of the fiber.
The typical range of polymer’s dielectric constant $\varepsilon$ is between 2 and 11, which creates a large dielectric contrast with the ceramic fibers ($\varepsilon_f \sim 1700$). Bent [99] proposed the inclusion of carbon black and PZT particles in the epoxy to reduce this contrast. Although the inclusion of these particles decreased the mismatch, it created other challenges in composite processing due to the higher concentrations of particles leading.
to agglomeration and increase in viscosity. Thanks to the advances made in dielectric polymer nano-composites, lower concentration of inclusion could be sufficient to improve the effective dielectric properties. In a previous study on epoxy/carbon nano fiber (CNF) [100] it was shown that addition of 2 weight % of CNF in epoxy led to a 20-time increase in the dielectric constant. Furthermore this increase in dielectric constant was not accompanied by an increase in viscosity or presence of voids or agglomerates, which is encouraging in terms of using the CNF-epoxy composite as an alternative matrix in AFCs. In this current study, materials properties, measured experimentally from [100], are listed in Table 4-3 and are used as inputs to simulate the piezoelectric behavior of AFCs with the different matrices. Figure 4-11 shows the results of the corresponding $d_{33}$ simulations. The presence of the CNFs increases the effective dielectric constant of the matrix, $\varepsilon_m$, without detrimentally affecting the other properties; in addition, the higher the CNF concentration, the higher the dielectric constant. As a result, the piezoelectric coefficient $d_{33}$ is predicted to increase and reach as high as 110 pm/V for 2 wt % CNF/epoxy (Figure 4-11).

Table 4-3: List of matrix materials properties.

<table>
<thead>
<tr>
<th>Matrix</th>
<th>$Y_m$ (GPa)</th>
<th>Poisson’s ratio</th>
<th>$\varepsilon_m$</th>
</tr>
</thead>
<tbody>
<tr>
<td>Epoxy</td>
<td>1.5</td>
<td>0.3</td>
<td>3.75</td>
</tr>
<tr>
<td>Epon 862</td>
<td>1.04</td>
<td>0.353</td>
<td>4</td>
</tr>
<tr>
<td>PVDF</td>
<td>2.2</td>
<td>0.3</td>
<td>11</td>
</tr>
<tr>
<td>0.1 wt% CNF-Epoxy</td>
<td>1.09</td>
<td>0.35</td>
<td>35</td>
</tr>
<tr>
<td>0.5 wt% CNF-Epoxy</td>
<td>1.16</td>
<td>0.35</td>
<td>55</td>
</tr>
<tr>
<td>2 wt% CNF-Epoxy</td>
<td>1.24</td>
<td>0.35</td>
<td>70</td>
</tr>
</tbody>
</table>
In this study a detailed model of AFCs that includes several fibers and electrode gaps, and taking into consideration the opposite poling in the fibers under the centerline of the electrodes, is developed and analyzed. Another unique feature of the model is the presence of the polymer matrix between fibers and electrodes, whereas in other studies in the literature the electrodes are assumed to be in direct contact with the fibers. In addition, by modeling the IDE electrodes as a separate component, we obtain a non-uniform electric field inside the fibers which is more realistic than assuming a constant...
electric field. The parametric study showed that in all the parameters varied, the improvement is always related to the amount of electric potential transferred to the fiber, which will create a local electric field in the piezoelectric material with a different magnitude than the applied one. The model provided information related to important geometrical and material parameters with the goal to improve the actuation performance of the AFC; namely, larger electrode gap, wider electrodes, thinner fibers and especially a higher matrix dielectric constant improve the actuation performance of the AFC dramatically. The biggest parameter impact was increasing the matrix dielectric constant, which is possible by reinforcing the matrix with high dielectric nano-particles. CNFs for example can increase the relative dielectric constant of the matrix from 4 to up to 70 at a relatively low concentration (2 wt %) without detrimentally affecting its mechanical behavior or creating voids and other manufacturing defects. In addition to identifying key parameters and related mechanisms that impact and inform actuation performance, the current model can also be used as a design tool. For example, engineers can explore use of a low modulus matrix and resulting AFC mechanical strength for a given application, or investigate limitations of achievable voltages in dictating electrode gap dimensions and actuator size. This parametric study is a useful tool to identify an optimized geometry and matrix that can be used for the manufacturing of next-generation AFCs with higher performance than what is currently available. However, it is noted that the modeling results do not match the experimental results, mainly because of the assumption of linear piezoelectric material used in the model. In order to estimate more realistic behavior of AFCs and use it as an input for models where AFCs are acting on host structures, non-linear piezoelectric behavior observed in section 3.3. In the next chapter, some of the
assumptions will be relaxed, and non-linear piezoelectricity and time-dependent behaviors will be included in the model, with the modeling results compared to experimental results.
Chapter 5  
Non-linear piezoelectricity and time-dependent modeling of AFCs  

5.1 Motivation  

When the experimental results obtained in Chapter 2 and 3 are compared to the modeling results obtained in Chapter 4, a large discrepancy was observed. Experimental $d_{33}$ coefficients of AFCs were found to vary between 100 and 200 pm/V, which is higher than the modeling results obtained in the previous chapter, where $d_{33}$ values of 12pm/V were found for similar geometry and material properties. The model assumed a linear piezoelectric material, however the experimental results on PZT fibers show that the $d_{33}$ and the polarization P are not linearly related to the electric field, see Figure 5-1, and results discussed in section 3.3. Furthermore, polarization and strain responses to an electric field are time-dependent as shown in results of section 3.3 and in Figure 5-1 c. One of the main applications of AFCs is actuation, and since in order to obtain higher strains, high electric fields need to be applied at different frequencies, a useful model has to account for the observed non-linear behavior and time-dependence. In this section, we will present a model that will take into consideration non-linear piezoelectricity, polarization switching and time-dependence.
5.2 Non-linear Piezoelectricity

5.2.1 Constitutive Equations

The constitutive equations for a piezoelectric material stated in Chapter 1 can be rewritten in different forms using the stiffness tensor $C$ instead of the compliance tensor $s$, stress coefficient piezoelectric tensor $e$ instead of the strain piezoelectric coefficient.

Figure 5-1: a) Non linear piezoelectricity and b) Polarization switching in PZT fibers.
tensor $d$, and the dielectric tensor at a constant strain instead of constant stress as shown in (5.1). For active materials undergoing large electric driving fields and small strains, higher order terms as a function of $E$ need to be included. The constitutive equation is rewritten as shown in (5.2) with electric field dependent piezoelectric and dielectric tensors respectively $h$ and $l$. The equations of $h$ and $l$ are shown in (5.3), where, $b$ is fourth-order electro-mechanical tensor, and $\chi$ is third-order electric permeability\cite{4, 101}. $b$ and $\chi$ can also be called Rayleigh coefficients. The coefficients $e$ and $\varepsilon$ are responsible for the reversible terms in (5.3), whereas the coefficients $b$ and $\chi$ are responsible of the irreversible term related to the domain wall motion \cite{102, 103}.

\[
T_{ij} = C_{ijkl}^E S_{kl} - e_{ij} E_k
\]
\[
D_i = e_{ik} S_{kl} - e_{ij} E_j
\]
\[
T_{ij} = C_{ijkl}^E S_{kl} - h_{ij} E_k
\]
\[
D_{ij} = C_{ijkl}^E S_{kl} + g_{ij} E_j
\]
\[
h_{ij} = e_{ij} + \frac{1}{2} b_{ij} E_i
\]
\[
g_{ij} = e_{ij} + \frac{1}{2} \chi_{ijk} E_k
\]

In our case, we are interested in evaluating the coefficients of $b$ and $\chi$ tensors in the 3-3 direction because it is the most important one for the AFC use. The coefficients are determined experimentally for the PZT 5A fiber. For $\chi$ coefficients, we will apply an electric field and maintain strain at zero so that we end up with the following relationship:

\[
D_j = \left( e_{ij} + \frac{1}{2} \chi_{ijk} E_k \right) E_j
\]
Experimentally, the fiber was clamped, sinusoidal electric field with 1 Hz frequency and ascending amplitudes (up to 1 MV/m) was applied, finally the polarization was recorded. We plotted the obtained values in Figure 5-2 and fit it to determine the $\chi$ coefficient.

![Graph showing P vs. E for a clamped PZT fiber.](image)

**Figure 5-2**: $P$ vs. $E$ for a clamped PZT fiber.

To determine $b$ coefficients experimentally, stress needs to be measured under clamped conditions while applying an electric field to eliminate the strain in the constitutive equation

$$T_{ij} = -\left(e_{kij} + \frac{1}{2}b_{kij}E_j\right)E_k$$

(5.5)

Due to the complexity of that experimental procedure, various guesses of $b$ coefficients were attempted until the simulation results matched the experimental data. In order to estimate the Rayleigh coefficient of PZT 5A fibers and compare it to the available bulk PZT 5A values in the literature, the constitutive equation (5.6) equivalent to (5.2) in the absence of an applied stress was used.
\[ S_{33} = d_{33}^{\text{initial}} E_3 + \alpha (E_3)^2 = (d_{33}^{\text{initial}} + \alpha E_3) E_3 = d_{33}(E_3) E_3 \quad (5.6) \]

where, \( d_{33}^{\text{initial}} \) is the piezoelectric coefficient at zero electric field, \( d_{33}(E_3) \) is the piezoelectric coefficient as a function of the applied electric field and \( \alpha \) is the Rayleigh coefficient. The initial piezoelectric coefficient and the Rayleigh coefficient were obtained by plotting \( d_{33}(E_3) \) as a function of \( E_3 \) as shown in Figure 5-3. The slope of the plot is the Rayleigh coefficient \( \alpha \), and the y intercept is the piezoelectric coefficient at zero electric field. It is found that \( \alpha = 280 \times 10^{-18} \text{ m}^2/\text{V}^2 \), which is comparable to the values found for bulk PZT 5A varying between 240 and 400 \( 10^{-18} \text{ m}^2/\text{V}^2 \) [102, 104]

![Graph showing the piezoelectric coefficient of PZT fiber as a function of the electric field](image)

**Figure 5-3:** Piezoelectric coefficient of PZT fiber as function of the electric field
5.2.2 Numerical Implementation

The linear behavior model was done in Abaqus, however the non-linear piezoelectric and ferroelectric behaviors are not included in the module of Abaqus. The procedure usually used when a material behavior is not included is to write a user material subroutine (UMAT) in FORTRAN. However, when meshing with piezoelectric elements, the use of UMAT is not possible. The solution is then to write a user element subroutine (UEL) that will take care of the elements and material behavior at the same time, where the previous equations are implemented. The UEL was used to solve for the elements in the PZT fibers, and the Abaqus solver was used for the rest of the elements. The boundary conditions and the materials properties discussed in Chapter 4 were used also in this case. The mesh used with the UEL has to be fine to ensure convergence. When this mesh was used for the four fibers RVE, more than 1 million elements were needed which requires more than 24 hours simulation time. To reduce the number of elements and therefore the simulation time, the RVE was decreased to three fibers instead of four as shown in Figure 5-4.
5.2.3 Actuation response at different electric field magnitudes

The results of the model using the UEL with the constitutive equations (as shown in 5.2.1) are analyzed and compared with experimental results and other models. In Figure 5-6, there are four cases considered: 1) The uniform field/non-linear behavior, which is a quarter fiber model with applied electric potential on the surfaces $z=0$ and $z=l$ where $l$ is the length of the quarter fiber instead of IDE electrodes. 2) The non-uniform field/linear behavior, which is the model discussed in the previous chapter. 3) The non-uniform field/non-linear behavior model, which combines the geometry of several fibers and uses IDE electrodes similar to the one shown in Chapter 4 and the UEL subroutine. 4) The experimental results, which are discussed in Chapter 3.
It can be seen that the uniform field overestimates the strain and the non-uniform field/linear behavior underestimates it. The non-uniform field model shows the dependence on the design parameters like IDE geometry and matrix materials properties. By combining the non-uniformity and non-linearity, the resulting behavior falls between the two, and is closer to the experimental results. Although the model takes into consideration the non-uniformity of the electric field, the poling is assumed to be uniform. The uniform poling assumption does not hold in the region below the electrodes fingers, where the electric field is mainly in the direction of the thickness of AFCs and not in the longitudinal direction as shown in Figure 5-6.

Figure 5-5: Comparing different models with experimental results.
5.3 Time-dependent and polarization switching

The experimental work conducted in Chapter 3 focused on understanding polarization-switching behavior in AFCs, PZT fibers and bulk PZT. Polarization under sinusoidal electric field of magnitudes larger than the coercive field was investigated and showed a non-linear behavior due to polarization switching. Other studies confirmed this behavior for bulk PZT and other piezoelectric ceramics [11, 105]. Our experimental results found in Chapters 2 and 3 as well as other studies [12, 57-59] have shown that mechanical, electrical and electro-mechanical responses of piezoelectric ceramics are time-dependent. PZT fibers and AFCs have shown creep and relaxation responses under different isothermal temperatures. Furthermore, ferroelectric and strain hysteresis loops show
frequency dependence. In this section, we will include this non-linearity and time-
dependence in the finite element model for PZT fibers and AFCs.

5.3.1 Constitutive equations

The poling direction for piezoelectric materials is always chosen as direction 3. In order
to model polarization switching, piezoelectric tensors have to vary with polarization. The
polarization depends on the electric field (magnitude and direction) at the current time \( t \)
and at previous time \( s \). The polarization model is proposed with an analogy to
viscoelastic-plastic modeling [106]. A single integral model with non-linear integrand as
the first approximation for modeling the time-dependent polarization behavior coupled
with residual polarization will be used. The polarization is expressed as summation of a
time-dependent term and a residual term.

\[
P_3[E_3(t-s),t] = R[E_3(t-s),t] + Q[E_3(t-s),t] \\
P[E',t] = R[E',t] + Q[E']
\] (5.7)

\( Q \) can be considered as the residual polarization that is caused by the application of a high
electric field opposite to the poling direction and \( R \) is the time dependent term.

Time dependent term

The time dependent term can be expressed as follows:

\[
R' = R[E_0',t] + \int_{0}^{t} \frac{\delta R[E',t-s]}{\delta E} \frac{dE^s}{ds} ds \\
t \geq 0
\] (5.8)
$R[E^0, t]$ is the polarization at current time $t$ when a constant electric field is applied at initial time $s = 0$. Experimentally, when a DC field is applied to the PZT fibers we noticed a variation of the polarization in creep-like behavior. Theoretically, dielectric materials can be modeled as a combination of resistance and capacitance (RC model) as shown in Figure 5-7. The polarization is the total charge $q$ divided by the cross section of the electrodes. Using the RC model we will solve for $q$ and get $P$ as a result.

![Equivalent RC model](image)

Figure 5-7: Equivalent RC model.

The total charge $q = \sum_{i=0}^{n} q_i$, where $q_0 = U^0 C_0$ and $q_i$ is the solution to the following differential equation:

$$R_i \frac{dq_i}{dt} + C_i q_i = U^0 \quad (5.9)$$

The solution to the equation (5.9) leads to a total charge expressed as:

$$q = U^0 C_0 + \sum_{i=1}^{n} U^0 C_i \left(1 - e^{-\frac{t}{R_i C_i}}\right) \quad (5.10)$$

Based on the total charge result, we can then find the polarization as function of the electric field:
\[ P = \kappa_0 E^0 + \sum_{i=1}^{i=n} \kappa_i E^0 \left( 1 - e^{-\frac{L}{\tau_i}} \right) \] (5.11)

where, \( \kappa_i = \frac{C_i l}{A} \), \( \tau_i = R_i C_i \), \( l \) is the distance between the electrodes and \( A \) is the cross section of the electrodes. In order to identify the different constants, a DC field less than the coercive field (0.75 MV/m) was applied to a PZT fiber and the current density was measured as a function of time. Current density \( \left( \frac{dP}{dt} \right) \) was measured instead of polarization to avoid charge drifts generated from the circuit for long periods of time. An Ammeter was used to measure the current. The current was plotted as a function of time and fitted to (5.12) where summation was stopped at the index two.

\[ I = \frac{dP}{dt} = \frac{\kappa_1}{\tau_1} E e^{-\frac{L}{\tau_1}} + \frac{\kappa_2}{\tau_2} E e^{-\frac{L}{\tau_2}} \] (5.12)

Figure 5-8: Fitting of the electric current with proposed model.
The coefficients obtained from the fitting and $\kappa_0$ obtained from the instantaneous polarization are shown in Table 5-1.

Table 5-1: Fitting coefficients.

<table>
<thead>
<tr>
<th>$\kappa_0$ (F/m)</th>
<th>$\kappa_1$ (F/m)</th>
<th>$\kappa_2$ (F/m)</th>
<th>$\tau_1$ (s)</th>
<th>$\tau_2$ (s)</th>
</tr>
</thead>
<tbody>
<tr>
<td>50 $10^9$</td>
<td>147 $10^9$</td>
<td>21 $10^9$</td>
<td>30.78</td>
<td>1.28</td>
</tr>
</tbody>
</table>

The time dependent term of the polarization can be then written as:

$$R' = \kappa_0 E^0_0 + \kappa_1 E^0_0 \left(1 - e^{-\frac{t}{\tau_1}}\right) + \kappa_2 E^0_0 \left(1 - e^{-\frac{t}{\tau_2}}\right)$$

$$+ \int_0^t \frac{\delta R}{\delta E}[E', t - s] \frac{dE^s}{ds} ds \quad t \geq 0$$

where the partial derivative of $R$ can be expressed as:

$$\frac{\delta R}{\delta E}[E', t - s] = \kappa_0 + \kappa_1 \left(1 - e^{-\frac{t-s}{\tau_1}}\right) + \kappa_2 \left(1 - e^{-\frac{t-s}{\tau_2}}\right)$$

(5.14)

(5.13) can be rewritten then as:

$$R[E, t] = \kappa_0 E_0' - \kappa_1 E_0^0 e^{-\frac{t}{\tau_1}} - \kappa_2 E_0^0 e^{-\frac{t}{\tau_2}} - \int_0^t \left(\kappa_1 e^{-\frac{t-s}{\tau_1}} + \kappa_2 e^{-\frac{t-s}{\tau_2}}\right) \frac{dE^s}{ds} ds$$

(5.15)

**Residual polarization**

The residual polarization can be defined as follows:

$$Q[E'] = \int_0^t \dot{Q} ds = \int_0^t \frac{dQ}{dE} \frac{dE^s}{ds} ds = \int_0^t \frac{dQ}{E} dE^s$$

(5.16)
where $\dot{Q}$ is the rate of residual polarization and $\frac{dQ'}{E}$ can be expressed for an unpoled material as:

$$\frac{dQ'}{dE} = \begin{cases} \lambda_0 \left| \frac{E'}{E_c} \right|^n & 0 \leq |E'| \leq E_c \\ \mu_0 e^{-\omega \left| \frac{E'}{E_c} \right|^n} & |E'| > E_c \end{cases}$$  \quad (5.17)

$\lambda_0$, $n_0$, $\mu_0$ and $\omega_0$ are material parameters that can be obtained from experiments. After the material is poled, an electric field with a magnitude higher than the coercive field and in the opposite direction to the poling is needed to switch the polarization. After poling, $\frac{dQ'}{dE}$ will follow (5.18) and the material parameters will updated to $\lambda$, $n$, $\mu$ and $\omega$ that are shown in Table 5-2.

$$\frac{dQ'}{dE} = \begin{cases} \lambda \left| \frac{E'}{E_c} \right|^n & 0 \leq E' \leq E_c, dE' \geq 0 \\ \lambda \left| \frac{E'}{E_c} \right|^n & -E_c \leq E' \leq 0, dE' \leq 0 \\ \mu \exp \left[-\omega \left( \frac{|E'|}{E_c} - 1 \right) \right] & E_c < E' \leq E_m, \; dE' \geq 0 \\ \mu \exp \left[-\omega \left( \frac{|E'|}{E_c} - 1 \right) \right] & -E_m < E' \leq E_c, \; dE' \leq 0 \\ 0 & 0 \leq E' \leq E_m, \; dE' < 0 \\ 0 & -E_m \leq E' \leq 0, \; dE' > 0 \end{cases}$$  \quad (5.18)

<table>
<thead>
<tr>
<th>$\lambda$ (F/m)</th>
<th>$n$</th>
<th>$\mu$ (F/m)</th>
<th>$\omega$ (s)</th>
</tr>
</thead>
</table>
| Table 5-2: Material parameters.
**Strain response**

The strain in piezoelectric materials can be related to the electric field or the polarization (=electric displacement) using the following constitutive equation:

\[
S_{ij} = s_{ijkl} T_{kl} + g_{ij} D_k
\]

(5.19)

The coefficient \(g\) depends on the polarization at current time using the following equation:

\[
S'_{ij} = g_{ij} \left| \frac{P'}{P_r} \right| \left[ \frac{\sqrt{P'}}{c_j} \right] P_r
\]

(5.20)

The time dependence and non-linearity in the polarization will then affect the strain and we will have similar impact on the strain or stress.

### 5.3.2 Numerical implementation

As stated in 5.2.2, the constitutive equations are implemented in UEL subroutine to be used by the Abaqus model. The integrals in the constitutive equations have been numerically implemented using time increments and electric field increments. The integral in the time dependent polarization can be written as:
The integrals \( I_i \) can be approximated as:

\[
I_i' = e^{-\frac{\Delta t}{\tau_i}} I_i^{-\Delta t} + \left( \kappa_i \frac{\Delta E'}{\Delta t} + \kappa_i e^{-\frac{\Delta t}{\tau_i}} \frac{\Delta E'^{-\Delta t}}{\Delta t} \right) \frac{\Delta t}{2}
\]  

(5.22)

where \( \Delta E' = E' - E'^{-\Delta t} \). The time dependent term of the polarization (5.13) can be written using (5.21) and (5.22) as

\[
R' = \kappa_0 E^0 + \kappa_1 E^0 \left( 1 - e^{-\frac{\Delta t}{\tau_1}} \right) + \kappa_2 E^0 \left( 1 - e^{-\frac{\Delta t}{\tau_2}} \right) + I_1' + I_2'
\]  

(5.23)

The residual polarization term can be written as

\[
Q' = Q'^{-\Delta t} + \Delta Q' = Q'^{-\Delta t} + \frac{dQ}{dE} \Delta E'
\]  

(5.24)

### 5.3.3 Polarization switching and time-dependent response

In this section, we will be showing results for the uniform field model for PZT fibers and for AFCs when the electric field is not uniform.
**PZT fiber with uniform field**

Simulations with a DC electric field and AC electric field have been conducted on a PZT fiber. The geometry was a cylinder and electric potential boundary conditions were applied at \( z=0 \) and \( z=L \) as shown in Figure 5-11.

The DC field was 0.75MV/m and the current and strain were evaluated as a function of time. Figure 5-10 shows the simulation results of strain and current as a function of time and the corresponding experimental results. The current simulation results match the experimental data very well, and the strain simulation results are close to the experimental data. The discrepancy between modeling and experimental results for the strain decreased after 30 seconds. In fact, in this model the strain was proportional to the polarization and time and stress dependence of the mechanical properties were not taken into consideration. These results prove that the model can predict the DC field response.
of PZT fibers. To better test the time dependence in the model, simulations were conducted on PZT fiber under AC electric field at different frequencies as shown in Figure 5-11.

Figure 5-10: PZT fiber response under a DC electric field.

Figure 5-11: PZT fiber modeling results under AC electric field.

The simulations were conducted for frequencies from 0.1Hz to 10Hz. The saturation polarization increases for lower frequencies as shown in experimental results of Chapter three. However, the model did not predict the increase of coercive field for higher frequencies. The model will be a tool to predict the response of PZT for different
frequencies and electric fields and will be used to predict the behavior of AFCs where PZT fibers are embedded.

**AFC response with a non-uniform electric field**

To model the AFC’s behavior, the UEL subroutine was used to model the PZT while the matrix was modeled as a linear elastic material as shown in Figure 5-4. The non-uniform electric field inside the fiber resulted in a large discontinuity in the materials properties (since they depend on the electric field and the polarization) that caused the model to diverge. The model was diverging mainly because there were elements with very high electric field causing high strains and pushing the electric field values to diverge. In order to avoid that, we focused only on solving the case for the polarization and excluded the strain effect. The results of polarization under 1 Hz AC electric field in Figure 5-12 show that the polarization does not match the experimental results. The polarization value is low and the shape of the loop is not similar to the modeling results that considered a uniform field. Moreover the maximum polarization is much smaller than what was observed in the experiments or in the uniform field modeling. This lower value is a result of the lower local electric field inside the fibers as compared to the applied one. The polarization is calculated by summing up the charges at the electrodes nodes and dividing them by the cross section of the fiber and the matrix surrounding it. In order to transfer the polarization to the electrodes and observe a typical ferroelectric hysteresis loop in AFCs, uniform poling assumption should be relaxed and the direction of the electric field should be considered as the 3-direction (in terms of materials property tensors).
In this chapter different constitutive equations of piezoelectric materials were discussed. First, fourth order piezoelectric and dielectric tensors were included to take into consideration the piezoelectric coefficients dependence on the electric field. The constitutive equations were included in a UEL subroutine that was used with ABQUS, and the focus was on three different cases:

- Uniform field and non-linear piezoelectricity
- Non-uniform field and linear piezoelectricity
- Non-uniform field and non-linear piezoelectricity

Figure 5-12: Comparison of AFC’s polarization simulation results for an AC electric field with 1Hz frequency with experimental results and simulation for uniform field.

5.4 Summary

In this chapter different constitutive equations of piezoelectric materials were discussed. First, fourth order piezoelectric and dielectric tensors were included to take into consideration the piezoelectric coefficients dependence on the electric field. The constitutive equations were included in a UEL subroutine that was used with ABQUS, and the focus was on three different cases:
The present model and simulation results can be very effective for accurate simulations when the AFC is an active structure embedded in a host structure. It was shown that taking into consideration the non-uniformity of the electric field and non-linear piezoelectricity is necessary, though it is not sufficient to validate the experimental results.

Second, polarization switching including time dependence constitutive equations was used to estimate materials response to electric fields higher than the coercive field and the response to DC electric field. Most of the material constants and parameters were obtained from experiments on PZT fibers. The equations were implemented in UEL subroutine that was used with Abaqus for PZT fibers and AFCs. For the PZT fibers simulations, the obtained results were in a good agreement with experimental results and showed frequency dependence.

For the AFCs simulations, the obtained results were not in good agreement with experimental results. In the proposed model, we assumed a uniform poling in the longitudinal direction, however the poling should follow the direction of the electric field. In order to match experimental data, non-uniform poling should also be taken into consideration.
Chapter 6

Summary and Future Work

6.1 Summary

AFCs are promising candidates to replace piezoelectric ceramics and 0-3 piezoelectric composites in several applications thanks to their flexibility and lightweight. However, an in-depth review of the research done on AFCs has shown that their piezoelectric performance is still not comparable to that of PZT transducers (effective d_{33} is 120 compared to 400 \times 10^{12} \text{ m/V} for bulk PZTs). In addition, most applications could subject the AFC to simultaneous mechanical loading, extreme environments, and moderate to high electric fields. There is lack of experimental characterization and numerical simulation of AFCs under these conditions and environments. In this dissertation, an experimental characterization (mechanical, electrical and electro-mechanical) of AFCs and their constituents was done for a better understanding of AFCs behavior; in particular, studies focused on exploring the reasons for the low piezoelectric properties compared to PZT and on quantifying the effects of time and temperature on their behavior.

Tensile tests were conducted at three different temperatures and three strain rates. Our findings confirm that the composite properties are time and temperature dependent. At higher temperatures, the modulus and tensile strength decreased for all strain rates while
strain at failure increased. At slow strain rates, the composite showed higher ductility and toughness for the specimens than those from faster strain rates, which resulted in relatively brittle responses. The time and temperature dependence of AFCs is attributed to the viscoelastic nature of the epoxy surrounding the fibers and the weakening of the fiber/epoxy and epoxy/Kapton interfaces. SEM images on fractured samples at higher temperature showed failure at those interfaces. These results were further confirmed by the tensile and DMA experiments conducted on the epoxy. The drop in modulus and strength of AFCs at high temperatures is due to the epoxy’s transition from the glassy state to the rubbery state. The mechanical behavior of PZT fibers was also investigated in depth. Some of the mechanical properties like modulus and strain at failure were found to be different from those of bulk PZT. For the first time, to the best of our knowledge, creep and relaxation experiments were performed for PZT fibers. The study of the response to creep shows a stress and temperature dependence. We conclude that creep and stress relaxation results obtained for AFCs were affected both by the viscoelastic behavior of epoxy and the behavior of PZT fibers. Non-linear mechanical behavior of PZT fibers under tensile testing was investigated with respect to temperature. Furthermore, we were able to quantify the amount of mechanical poling stress using cyclic loading and unloading testing. Some of the properties obtained for epoxy and PZT fibers will be used for the modeling of AFCs. Other results can be used as reference for future modeling of mechanical non-linearity and time dependence of piezoelectric fibers.

An experimental study was also conducted on the impact of time and temperature on the ferroelectric and electro-mechanical response of AFCs, bulk PZT and PZT fibers. It was
found that the remnant and saturation polarizations increase with the decrease of frequency and increase of temperature for AFCs and pure PZT (bulk and fiber) while the coercive field decreased with temperature. The values of polarization in AFCs were smaller than the ones measured for bulk PZT and PZT fibers because of the low local electric field inside the PZT fibers. The non-uniform electric field distribution and the epoxy layer separating the electrodes from the fibers are the reasons for this low electric field. For the same reasons, $d_{33}$ coefficients of AFCs were found to be much lower than those of PZT fibers. The $d_{33}$ coefficients increased with temperature increase and frequency decrease for AFCs and PZT fibers. For AFCs, the frequency and temperature dependence are due to the behavior of the PZT fiber and also to the viscoelastic behavior of the epoxy that is constraining the PZT fibers. The electro-mechanical behavior of the AFCs and PZT fibers was found to be non-linear and depended on the magnitude of the applied electric field even for magnitudes less than the coercive field. The obtained results in this chapter provided a guide for the modeling work conducted on AFCs and PZT fibers. In addition, the experimental procedures discussed in this chapter were used to calibrate some of the materials parameters and properties that were used for the modeling.

A detailed model of AFCs that includes four fibers and three electrode gaps, and taking into consideration the opposite poling in the fibers under the centerline of the electrodes was developed. Another unique feature of the model was the presence of the polymer matrix between fibers and electrodes, whereas in other studies in the literature the electrodes are assumed to be in direct contact with the fibers. Geometric design
parameters and matrix properties were varied and $d_{33}$ was measured. The parametric study provided information related to important geometrical and material parameters with the goal to improve the actuation performance of the AFC; namely, larger electrode gap, wider electrodes, thinner fibers and especially a higher matrix dielectric constant. The biggest parameter impact was increasing the matrix dielectric constant, which is possible by reinforcing the matrix with high dielectric nano-particles. CNFs for example can increase the relative dielectric constant of the matrix from 4 to up to 70 at a relatively low concentration (2wt%). In addition to identifying key parameters and related mechanisms that impact and inform actuation performance, the current model can also be used as a design tool. This parametric study is a good tool to identify an improved geometry and matrix that can be used for the manufacturing of next-generation AFCs with higher performance than what is currently available. However, the initial model considered does not validate the experimental results; the discrepancy is mainly because of the assumption of linear piezoelectric material used in the model.

In order to realize a more realistic behavior of AFCs and use it as an input for models where AFCs are acting on host structures, some of the assumptions were relaxed, and non-linear piezoelectricity and time-dependence are included in the model. Fourth order piezoelectric and dielectric tensors were included to take into consideration the piezoelectric coefficients dependence on the electric field. It was shown that combining the non-uniformity and non-linearity of the piezoelectric coefficients gives a better estimate of the actuation performance of AFCs compared to models where only non-uniformity or non-linearity is taken into consideration. For electric field magnitudes
higher than the coercive field, a polarization-switching model is included which also takes into consideration time-dependence. Materials properties and model parameters were obtained from experiments on PZT fibers. DC and AC simulations conducted on PZT fibers were in a good agreement with experimental results and showed frequency dependence. For the AFCs simulations above the coercive field, the obtained results were not in good agreement with experimental results mainly because of the assumption of uniform poling inside the fibers.

6.2 Contributions of this dissertation

Several conclusions and results were obtained from the current work. In this part we will focus mainly on the most important ones and their impact. Major outputs can be divided into three different fields:

- Experimental characterization and modeling of the PZT fibers
- Development of a genuine FEM model for AFCs
- Overall characterization of AFCs

6.2.1 Experimental characterization and modeling of the PZT fibers

Experimental characterization of the PZT fibers was one of the most challenging tasks in this study mainly because of their brittle behavior, small size and low capacitance. For the mechanical testing, new procedure different from the ASTM standards was used. To
the best of our knowledge, creep and stress relaxation were conducted for the first time on PZT fibers. In addition, mechanical poling of the fibers has been quantified by cyclic mechanical loading under tension, which has not been done before. Mechanical poling was quantified in other studies for bulk PZT and only using compressive stress. The electrical and electro-mechanical tests conducted on PZT fibers generated several properties of the PZT fibers under different loading conditions that can fill the literature gap on the fiber’s properties. The experiments quantified the impact of the use of IDE electrodes on the fiber’s polarization. The experimental setups used for the electrical and electro-mechanical testing of fibers were used to calculate the materials parameters to model polarization switching at different frequencies and DC field response. The modeling results showed a good agreement with the experimental results.

6.2.2 Development of a genuine FEM model for AFCs

The modeling work was not restricted to the PZT fibers only. A FE model was built for AFCs taking into consideration several aspects that were neglected in previous studies like: non-uniform distribution of the electric field, presence of epoxy interface separating the electrodes from the fibers and opposite poling occurring under the electrodes fingers. This model was a tool to identify the most important parameters that control actuation performance of AFCs. The most important parameters are the matrix dielectric constant and the electrodes gap. The model can then be used for similar devices that utilize IDE electrodes. FE modeling of AFCs included also non-linear piezoelectric behavior by adding Rayleigh coefficients to the constitutive equations of PZT in a user element.
subroutine, referred to as UEL in Abaqus. The obtained results showed a good agreement with experimental results. The developed model can be added to a larger one where the AFC is attached on a host structure to estimate the AFC’s performance while in application.

6.2.3 Overall characterization of AFCs

Contribution of AFC’s constituents on its overall behavior has been quantified through experiments. The study also focused on the impact of time and temperature on the properties of the composites. The obtained properties under different loading conditions can serve as a guide to identify the limitations for the use of AFCs in certain applications.

6.3 Future work

From the experimental and numerical simulations conducted on AFCs, we showed that the AFCs are not yet designed with their optimal parameters. AFCs should be able to operate at higher temperatures, which require the use of a matrix with higher $T_g$. A matrix with higher $T_g$ usually has a higher Young’s modulus at room temperature, but according to simulation results, that will not have a big impact on the actuation performance of AFCs. Experimental study needs to be conducted in order to confirm this finding. AFCs suffer from low polarization and electro-mechanical coupling compared to PZT fibers due to low local electric field inside the fibers. Inclusions of low concentrations of CNF
in the matrix can decrease the dielectric mismatch and increase the local electric field inside the fibers. However, the inclusions of CNF might decrease the dielectric breakdown of AFCs. One way to address this challenge is to align the inclusions through the thickness of the sample using AC dielectrophoresis. A preliminary study was conducted to assess feasibility of this approach. The experimental set up used for alignment is shown in Figure 6-1. A PZT fiber is placed in the 1 mm gap between two electrodes then 0.2wt% CNF/resin mixture was added. An AC field with a magnitude of 0.75kV/m and a frequency of 1kHz was applied, and the sample was observed using an inverted optical microscope. The CNF particles aligned on both sides of the PZT as shown in Figure 6-2.

Figure 6-1. Experimental set up used for CNF alignment.

Figure 6-2. Alignment of CNF in an epoxy resin in the presence of a PZT fiber after a) 0 min, b) 15 min and c) 30 min of applying the electric field.
The network of CNF in the epoxy as shown above can create an internal electrode surrounding the fiber and increase the local electric field in the PZT. Although the results of alignment were promising, further investigation is required to study the impact of alignment on dielectric constant, dielectric breakdown and electrical conductivity.

The development and improvement of AFCs will need further simulation to guarantee optimal design. The model should better represent the poling in the direction of the electric field to obtain more accurate simulation results. This can be done by including local orientation for every element in the built mode. Including non-uniform poling with the different material behaviors discussed in Chapter 5 (time-dependent polarization, non-linear piezoelectricity and polarization switching) should allow a better estimate of the behavior of AFC under electrical and mechanical loading.
REFERENCES


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[99] Bent AA. Active fiber composites for structural actuation: Massachusetts Institute of Technology; 1997.


Appendix

Materials properties and tensors

Table 1: Materials properties of epoxy

<table>
<thead>
<tr>
<th>Specification</th>
<th>Value</th>
</tr>
</thead>
<tbody>
<tr>
<td>Specific gravity</td>
<td>1.07</td>
</tr>
<tr>
<td>Maximum use temperature (Specified by manufacturer)</td>
<td>82°C</td>
</tr>
<tr>
<td>Tensile strength</td>
<td>8.27 MPa</td>
</tr>
<tr>
<td>Compressive strength</td>
<td>51.71 MPa</td>
</tr>
<tr>
<td>Elongation at break</td>
<td>51%</td>
</tr>
<tr>
<td>Viscosity</td>
<td>13000 CPS</td>
</tr>
<tr>
<td>Dielectric constant</td>
<td>3.75</td>
</tr>
<tr>
<td>Dielectric strength</td>
<td>490 V/mil</td>
</tr>
</tbody>
</table>

The dielectric, piezoelectric and compliance matrices of PZT 5A are shown below:

\[
\varepsilon^T = \varepsilon_0 \begin{pmatrix} 1750 & 0 & 0 \\ 0 & 1650 & 0 \\ 0 & 0 & 1650 \end{pmatrix} \frac{F}{m}
\]

\[
d = \begin{pmatrix} 0 & 0 & 0 & 0 & 550 & 0 \\ 0 & 0 & 0 & 550 & 0 & 0 \\ -180 & -180 & 400 & 0 & 0 & 0 \end{pmatrix} \times 10^{-12} \frac{m}{V}
\]

\[
s^E = \begin{pmatrix} 16.4 & -5.74 & -7.22 & 0 & 0 & 0 \\ -5.74 & 16.4 & -7.22 & 0 & 0 & 0 \\ -7.22 & -7.22 & 18.8 & 0 & 0 & 0 \\ 0 & 0 & 0 & 47.5 & 0 & 0 \\ 0 & 0 & 0 & 0 & 47.5 & 0 \\ 0 & 0 & 0 & 0 & 0 & 44.3 \end{pmatrix} \times 10^{-12} \frac{m^2}{N}
\]
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Advisor: Zoubeida Ounaies
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Master of Science: Computational Mechanics
Ecole Polytechnique de Tunisie
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Tunis, Tunisia
Dissertation Title: Numerical Simulation of Solar Updraft Tower
(http://www2.esm.vt.edu/~nayfeh/cmgp-tps/index.html)
Advisor: Melik Sahraoui

Bachelor of Science: Mechanics and Structures
Ecole Polytechnique de Tunisie
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Publications