The Pennsylvania State University

The Graduate School

STUDIES OF DIPOLE-DRIVEN ACTIVE MATERIALS: MULTIFIELD PROCESSING SIMULATIONS, MICROSTRUCTURE-BASED CONSTITUTIVE MODELING, AND MACROSCOPIC ACTIVE COMPOSITE OPTIMIZATION

A Dissertation in

Mechanical Engineering

by

Anil Erol

© 2019 Anil Erol

Submitted in Partial Fulfillment of the Requirements for the Degree of

Doctor of Philosophy

August, 2019

The dissertation of Anil Erol was reviewed and approved* by the following:

Paris von Lockette Associate Professor of Mechanical Engineering Dissertation Advisor Chair of Committee

Mary Frecker Professor of Mechanical Engineering and Biomedical Engineering

Zoubeida Ounaies Professor of Mechanical Engineering

Francesco Costanzo Professor of Engineering Science and Mechanics, Mechanical Engineering and Biomedical Engineering Head of the Department or Chair of the Graduate Program

Daniel Haworth Professor of Mechanical Engineering Head of the Department

*Signatures are on file in the Graduate School

ABSTRACT

Dipole-based active materials are a wide-ranging class of materials used in industries from aerospace to biomedicine. The advantages of these materials are their ability to be actuated while untethered via electric and/or magnetic fields, and that the interactions of the embedded dipoles can be manipulated on a micro- or even nano-scale. While dipole-based materials are becoming more widely used, there is a lack of rigorous modeling techniques that could help characterize, design, and implement them in applications. This work aims to address the challenges that arise when studying the materials at each length-scale: micro, meso, and macro.

At the micro-scale, this work studied the effects of electric and magnetic fields on the alignment and ordering of electromagnetically susceptible particles suspended in an elastomeric fluid medium. While past studies have shown that multiple fields can assemble particles in a fluid in multiple directions, ferrohydrodynamic particle simulations in this study demonstrated that the simultaneous application of electric and magnetic fields on hard magnetic particles with geometric anisotropy can create a hierarchy of structures at different length scales, which can be used to achieve a wider range of structure and properties. The simulation methods included permanent magnetic dipoles and induced electric dipoles, yielding magneto- and dielectrophoretic effects, and electric and magnetic torques acting against hydrodynamic drag forces.

At the meso-scale, the mechanics of a class of dipole-based semicrystalline electro-active polymers (EAPs) called relaxor ferroelectric polymers were modeled to understand how tailored dipole-based microstructures may affect bulk electromechanical response. To fill the gap in EAP modeling literature, this work developed a modeling framework for dipole-based EAPs by utilizing the micromechanics of semicrystalline polymer network model, ascribing dipole-dipole energies to crystalline regions and the eight-chain hyper-elastic model to amorphous regions. Finding the equilibrium of the network model yielded a relationship between the spatial arrangements of dipolar regions and the electromechanical coupling of the bulk material. The findings revealed that the anisotropy of dipole-dipole interactions cause particle arrangements parallel to an external field to generate more electromechanical coupling than all other arrangements, implicating a microstructure that could maximize EAP performance. Results of an analysis of the energies also shows that adjusting particle arrangements with respect to the field can either promote or inhibit instabilities, which can either cause material failure or be harvested for larger deformations depending on boundary conditions.

At the macro-scale, a numerical model was developed for arbitrarily segmented and multi-layered beams utilizing EAPs and MAEs. The model was employed in a multi-objective design optimization problem to minimize shape error and cost, while maximizing work output. Optimization results emphasized the importance of gaps between MAE patches and the uniformity and symmetry in their magnetization for matching symmetric shapes with ideal folds. Furthermore, for greatest work performance, some patches yielded greater sensitivity than others, offering a trade-off between work and shape approximation. While these results could be improved with a larger and more diverse initial population, the methodology demonstrated the ability to quickly achieve near-optimal designs with a wide selection of designs based on application priorities.

LIST OF TABLESxiii ACKNOWLEDGEMENTSxiv CHAPTER 1	LIST OF FIGURES	vii
ACKNOWLEDGEMENTS xiv CHAPTER 1 1 1.1 Impacts of Dipole-Driven Materials 1 CHAPTER 2 11 2.1 Background on Multi-Field Processing 11 2.1 Background on Multi-Field Processing 11 2.1 Dipole-Driven Materials 15 2.2 Dynamic Particle Simulation Methods 15 2.2.1 Magnetic force and torque 16 2.2.2 Southeof Volume and Coordinates 16 2.2.3 Control Volume and Coordinates 16 2.2.4 Equations for Case of BHF Particles 18 2.2.5 Hydrodynamic Forces and Torques 22 2.2.6 Equations of Motion 23 2.7 Temporal Discretization 24 2.2.8 Boundary Conditions 25 2.3 Multi-Field Processing Conditions 28 2.3.1 Distance-Based Minimum Spanning Tree Analysis 35 2.3.2 Dispersions of Particle, Stack, and Chain Orientations for Distance-Based Spanning Trees 55 2.4. Conclusions 56 CHAPTER 3 59 3.1 Literature Review on EAP Mechanics 59 3.1.1 Microstructure-property relationship in PVDF-based RFE polymers60 3.1.3 Overview of Literature on EAP Modeling	LIST OF TABLES	xiii
CHAPTER 1 1 1.1 Impacts of Dipole-Driven Materials 1 CHAPTER 2 11 2.1 Background on Multi-Field Processing 11 2.2 Dynamic Particle Simulation Methods 15 2.2 Dynamic Particle Simulation Methods 15 2.2 Dynamic Particle Simulation Methods 16 2.2 Dynamic Forces and Torques 16 2.2 S Hydrodynamic Forces and Torques 22 2.2 A Equations for Case of BHF Particles 18 2.2.7 Temporal Discretization 24 2.2.8 Boundary Conditions 23 2.7 Temporal Discretization 24 2.3.1 Distance-Based Minimum Spanning Tree Analysis 35 2.3.1 Distance-Based Minimum Spanning Tree Analysis 40 2.3.2 Dispersions of Particle, Stack, and Chain Orientations for Distance- Based Spanning Trees 55 2.4. Conclusions 56 CHAPTER 3 59 3.1 Literature Review on EAP Mechanics 59 3.1.1 Microstructure-property relation	ACKNOWLEDGEMENTS	xiv
1.1 Impacts of Dipole-Driven Materials. 1 CHAPTER 2 11 2.1 Background on Multi-Field Processing. 11 2.2 Dynamic Particle Simulation Methods. 15 2.2.1 Magnetic force and torque 15 2.2.2 Dielectrophoresis and electric torques. 16 2.2.3 Control Volume and Coordinates. 16 2.2.4 Equations for Case of BHF Particles. 18 2.2.5 Hydrodynamic Forces and Torques. 22 2.2.6 Equations of Motion 23 2.7.7 Temporal Discretization 24 2.8 Boundary Conditions. 25 2.3 Multi-Field Processing Results and Conclusions 26 2.3.1 Distance-Based Minimum Spanning Tree Analysis. 35 2.3.2 Dispersions of Particle, Stack, and Chain Orientations for Distance-Based Spanning Trees. 37 2.3.3 Dipole-Dipole Energy-Based Minimum Spanning Tree Analysis. 40 2.3.4 Dispersions of Particle, Stack, and Chain Orientations for Distance-Based Spanning Trees. 59 3.1.1 Microstructure-property relationship in PVDF-based RFE polymers60 31.1 Distrostructure-property relationship in PVDF-based RFE polymers60 3.1.2 Background on Hyperelastic Modeling. 62 31.3 Overview of Literature on EAP modeling. 65	CHAPTER 1	1
CHAPTER 2 11 2.1 Background on Multi-Field Processing 11 2.2 Dynamic Particle Simulation Methods 15 2.2.1 Magnetic force and torque 15 2.2.2 Dielectrophoresis and electric torques 16 2.3 Control Volume and Coordinates 16 2.4 Equations for Case of BHF Particles 18 2.2.5 Hydrodynamic Forces and Torques 22 2.6 Equations of Motion 23 2.7 Temporal Discretization 24 2.8 Boundary Conditions 25 2.3 Multi-Field Processing Results and Conclusions 26 2.3.1 Five Cases of Processing Conditions 28 2.3.2 Dispersions of Particle, Stack, and Chain Orientations for Distance- Based Spanning Trees 37 2.3.3 Dipole-Dipole Energy-Based Minimum Spanning Tree Analysis 40 2.3 4 Dispersions of Particle, Stack, and Chain Orientations for Distance- Based Spanning Trees Based Spanning Trees 55 2.4 Conclusions 59 3.1 Literature Review on EAP Mechanics 59 3.1.1 Microstructure-property relationship in PVDF-based RFE polymers60 31.2 Background on Hyperelastic Modeling 3.1.2 Background on Hyperelastic Modeling	1.1 Impacts of Dipole-Driven Materials	1
2.1 Background on Multi-Field Processing 11 2.2 Dynamic Particle Simulation Methods 15 2.2.1 Magnetic force and torque 15 2.2.2 Dielectrophoresis and electric torques 16 2.2.3 Control Volume and Coordinates 16 2.2.4 Equations for Case of BHF Particles 18 2.2.5 Hydrodynamic Forces and Torques 22 2.2.6 Equations of Motion 23 2.2.7 Temporal Discretization 24 2.2.8 Boundary Conditions 26 2.3.1 Five Cases of Processing Conditions 28 2.3.1 Distance-Based Minimum Spanning Tree Analysis 35 2.3.2 Dispersions of Particle, Stack, and Chain Orientations for Distance- Based Spanning Trees 37 2.3.3 Dipole-Dipole Energy-Based Minimum Spanning Tree Analysis 40 2.3.4 Dispersions of Particle, Stack, and Chain Orientations for Distance- Based Spanning Trees Based Spanning Trees 55 2.4. Conclusions 55 2.5. ChAPTER 3 59 3.1.1 Microstructure-property relationship in PVDF-based RFE polymers60 3.1.2 Background on Hyperelastic Modeling 3.1.3 Overview of Literature on EAP modeling 62 3.1.4 Classification of Sele	CHAPTER 2	
2.2 Dynamic Particle Simulation Methods 15 2.2.1 Magnetic force and torque 15 2.2.2 Dielectrophoresis and electric torques 16 2.2.3 Control Volume and Coordinates 16 2.2.4 Equations for Case of BHF Particles 18 2.2.5 Hydrodynamic Forces and Torques 22 2.2.6 Equations of Motion 23 2.7 Temporal Discretization 24 2.2.8 Boundary Conditions 25 2.3 Multi-Field Processing Results and Conclusions 28 2.3.1 Five Cases of Processing Conditions 28 2.3.1 Distance-Based Minimum Spanning Tree Analysis 35 2.3.2 Dispersions of Particle, Stack, and Chain Orientations for Distance- Based Spanning Trees 37 2.3.3 Dipole-Dipole Energy-Based Minimum Spanning Tree Analysis 40 2.3.4 Dispersions of Particle, Stack, and Chain Orientations for Distance- Based Spanning Trees Based Spanning Trees 55 2.4. Conclusions 56 CHAPTER 3 59 3.1.1 Microstructure-property relationship in PVDF-based RFE polymers60 31.2 Background on Hyperelastic Modeling 3.1.3 Overview of Literature on EAP modeling 62 3.1.4 Classification of	2.1 Background on Multi-Field Processing	
2.2.1 Magnetic force and torque 15 2.2.2 Dielectrophoresis and electric torques 16 2.2.3 Control Volume and Coordinates 16 2.2.4 Equations for Case of BHF Particles 18 2.2.5 Hydrodynamic Forces and Torques 22 2.2.6 Equations of Motion 23 2.7 Temporal Discretization 24 2.2.8 Boundary Conditions 25 2.3 Multi-Field Processing Results and Conclusions 26 2.3.1 Five Cases of Processing Conditions 28 2.3.1 Distance-Based Minimum Spanning Tree Analysis 35 2.3.2 Dispersions of Particle, Stack, and Chain Orientations for Distance- Based Spanning Trees 37 2.3.3 Dipole-Dipole Energy-Based Minimum Spanning Tree Analysis 40 2.3.4 Dispersions of Particle, Stack, and Chain Orientations for Distance- Based Spanning Trees Based Spanning Trees 55 2.4. Conclusions 59 3.1 Literature Review on EAP Mechanics 59 3.1.1 Microstructure-property relationship in PVDF-based RFE polymers60 3.1.2 Background on Hyperelastic Modeling 62 3.1.3 Overview of Literature on EAP modeling 65 3.1.4 Classification of Selected Models	2.2 Dynamic Particle Simulation Methods	
2.2.2 Dielectrophoresis and electric torques 16 2.2.3 Control Volume and Coordinates 16 2.2.4 Equations for Case of BHF Particles 18 2.2.5 Hydrodynamic Forces and Torques 22 2.2.6 Equations of Motion 23 2.2.7 Temporal Discretization 24 2.2.8 Boundary Conditions 25 2.3 Multi-Field Processing Results and Conclusions 26 2.3.1 Five Cases of Processing Conditions 28 2.3.1 Distance-Based Minimum Spanning Tree Analysis 35 2.3.2 Dispersions of Particle, Stack, and Chain Orientations for Distance- Based Spanning Trees 37 2.3.4 Dispersions of Particle, Stack, and Chain Orientations for Distance- Based Spanning Trees 55 2.4. Conclusions 55 2.4. Conclusions 59 3.1 Literature Review on EAP Mechanics 59 3.1.1 Microstructure-property relationship in PVDF-based RFE polymers60 3.1.2 Background on Hyperelastic Modeling 3.1.3 Overview of Literature on EAP modeling 62 3.1.4 Classification of Selected Models 68 3.1.5 Summary of Literature 77 3.2 EAP Modeling framework 77 3.	2.2.1 Magnetic force and torque	15
2.2.3 Control Volume and Coordinates 16 2.2.4 Equations for Case of BHF Particles 18 2.2.5 Hydrodynamic Forces and Torques 22 2.2.6 Equations of Motion 23 2.7 Temporal Discretization 24 2.8 Boundary Conditions 25 2.3 Multi-Field Processing Results and Conclusions 26 2.3.1 Five Cases of Processing Conditions 28 2.3.1 Distance-Based Minimum Spanning Tree Analysis 35 2.3.2 Dispersions of Particle, Stack, and Chain Orientations for Distance- Based Spanning Trees 37 2.3.3 Dipole-Dipole Energy-Based Minimum Spanning Tree Analysis 40 2.3.4 Dispersions of Particle, Stack, and Chain Orientations for Distance- Based Spanning Trees 55 2.4. Conclusions 56 CHAPTER 3 59 3.1 Literature Review on EAP Mechanics 59 3.1.1 Microstructure-property relationship in PVDF-based RFE polymers60 51.2 Background on Hyperelastic Modeling 3.1.2 Background on Hyperelastic Modeling 62 3.1.3 Overview of Literature 77 3.2 EAP Modeling framework 77 3.2.1 Kinematics of a semicrystalline, electrostrictive body 78 <	2.2.2 Dielectrophoresis and electric torques	16
2.2.4 Equations for Case of BHF Particles 18 2.2.5 Hydrodynamic Forces and Torques 22 2.2.6 Equations of Motion 23 2.2.7 Temporal Discretization 24 2.2.8 Boundary Conditions 25 2.3 Multi-Field Processing Results and Conclusions 26 2.3.1 Five Cases of Processing Conditions 28 2.3.1 Distance-Based Minimum Spanning Tree Analysis 35 2.3.2 Dispersions of Particle, Stack, and Chain Orientations for Distance-Based Spanning Trees 37 2.3.3 Dipole-Dipole Energy-Based Minimum Spanning Tree Analysis 40 2.3.4 Dispersions of Particle, Stack, and Chain Orientations for Distance-Based Spanning Trees 55 2.4. Conclusions 55 2.4. Conclusions 56 CHAPTER 3 59 3.1.1 Microstructure-property relationship in PVDF-based RFE polymers60 3.1.2 Background on Hyperelastic Modeling 3.1.3 Overview of Literature on EAP modeling 65 3.1.4 Classification of Selected Models 68 3.1.5 Summary of Literature 77 3.2 EAP Modeling framework 77 3.2.1 Kinematics of a semicrystalline, electrostrictive body 78 3.2.2 Traction-free equilibrium 84	2.2.3 Control Volume and Coordinates	
2.2.5 Hydrodynamic Forces and Torques 22 2.2.6 Equations of Motion 23 2.2.7 Temporal Discretization 24 2.2.8 Boundary Conditions 25 2.3 Multi-Field Processing Results and Conclusions 26 2.3.1 Five Cases of Processing Conditions 28 2.3.1 Distance-Based Minimum Spanning Tree Analysis 35 2.3.2 Dispersions of Particle, Stack, and Chain Orientations for Distance- 37 Based Spanning Trees 37 2.3.3 Dipole-Dipole Energy-Based Minimum Spanning Tree Analysis 40 2.3.4 Dispersions of Particle, Stack, and Chain Orientations for Distance- 55 Based Spanning Trees 55 2.4. Conclusions 56 CHAPTER 3 59 3.1.1 Microstructure-property relationship in PVDF-based RFE polymers60 51 3.1.2 Background on Hyperelastic Modeling 62 3.1.3 Overview of Literature on EAP modeling 65 3.1.4 Classification of Selected Models 68 3.1.5 Summary of Literature 77 3.2 EAP Modeling framework 77 3.2.1 Kinematics of a semicrystalline, electrostrictive body 78 3.2.2 Traction-free equilibrium 84 <	2.2.4 Equations for Case of BHF Particles	
2.2.6 Equations of Motion 23 2.2.7 Temporal Discretization 24 2.2.8 Boundary Conditions. 25 2.3 Multi-Field Processing Results and Conclusions 26 2.3.1 Five Cases of Processing Conditions. 28 2.3.1 Distance-Based Minimum Spanning Tree Analysis 35 2.3.2 Dispersions of Particle, Stack, and Chain Orientations for Distance- Based Spanning Trees 37 2.3.3 Dipole-Dipole Energy-Based Minimum Spanning Tree Analysis 40 2.3.4 Dispersions of Particle, Stack, and Chain Orientations for Distance- Based Spanning Trees Based Spanning Trees 55 2.4. Conclusions 56 CHAPTER 3 59 3.1.1 Microstructure-property relationship in PVDF-based RFE polymers60 31.2 Background on Hyperelastic Modeling 3.1.3 Overview of Literature on EAP modeling 62 3.1.4 Classification of Selected Models 68 3.1.5 Summary of Literature 77 3.2 EAP Modeling framework 77 3.2 Electromechanical response of a hyperelastic biphasic body with dipole-dipole interactions 3.2.4 Orientation dependence of polarization of crystalline domains 90 3.2.5 Formulation of electrostrictive model for diffe	2.2.5 Hydrodynamic Forces and Torques	
2.2.7 Temporal Discretization 24 2.2.8 Boundary Conditions 25 2.3 Multi-Field Processing Results and Conclusions 26 2.3.1 Five Cases of Processing Conditions 28 2.3.1 Distance-Based Minimum Spanning Tree Analysis 35 2.3.2 Dispersions of Particle, Stack, and Chain Orientations for Distance- 37 Based Spanning Trees 37 2.3.3 Dipole-Dipole Energy-Based Minimum Spanning Tree Analysis 40 2.3.4 Dispersions of Particle, Stack, and Chain Orientations for Distance- 38 Based Spanning Trees 55 2.4. Conclusions 56 CHAPTER 3 59 3.1.1 Microstructure-property relationship in PVDF-based RFE polymers60 31.2 Background on Hyperelastic Modeling 3.1.3 Overview of Literature on EAP modeling 62 3.1.4 Classification of Selected Models 68 3.1.5 Summary of Literature 77 3.2 EAP Modeling framework 77 3.2 Electromechanical response of a hyperelastic biphasic body with 40 dipole-dipole interactions 85 3.2.4 Orientation dependence of polarization of crystalline domains 90 3.2.5 Formulation of electrostrictive model for different network models	2.2.6 Equations of Motion	
2.2.8 Boundary Conditions 25 2.3 Multi-Field Processing Results and Conclusions 26 2.3.1 Five Cases of Processing Conditions 28 2.3.1 Distance-Based Minimum Spanning Tree Analysis 35 2.3.2 Dispersions of Particle, Stack, and Chain Orientations for Distance-Based Spanning Trees 37 2.3.3 Dipole-Dipole Energy-Based Minimum Spanning Tree Analysis 40 2.3.4 Dispersions of Particle, Stack, and Chain Orientations for Distance-Based Spanning Trees 55 2.4. Conclusions 55 2.4. Conclusions 56 CHAPTER 3 59 3.1 Literature Review on EAP Mechanics 59 3.1.1 Microstructure-property relationship in PVDF-based RFE polymers60 51.2 3.1.2 Background on Hyperelastic Modeling 62 3.1.3 Overview of Literature on EAP modeling 62 3.1.4 Classification of Selected Models 68 3.1.5 Summary of Literature 77 3.2 EAP Modeling framework 77 3.2.1 Kinematics of a semicrystalline, electrostrictive body 78 3.2.2 Traction-free equilibrium 84 3.2.3 Electromechanical response of a hyperelastic biphasic body with 61 dipole-dipole interactions 85 <td>2.2.7 Temporal Discretization</td> <td></td>	2.2.7 Temporal Discretization	
2.3 Multi-Field Processing Results and Conclusions 26 2.3.1 Five Cases of Processing Conditions 28 2.3.1 Distance-Based Minimum Spanning Tree Analysis 35 2.3.2 Dispersions of Particle, Stack, and Chain Orientations for Distance-Based Spanning Trees 37 2.3.3 Dipole-Dipole Energy-Based Minimum Spanning Tree Analysis 40 2.3.4 Dispersions of Particle, Stack, and Chain Orientations for Distance-Based Spanning Trees 55 2.4. Conclusions 55 2.4. Conclusions 56 CHAPTER 3 59 3.1 Literature Review on EAP Mechanics 59 3.1.1 Microstructure-property relationship in PVDF-based RFE polymers60 51.2 Background on Hyperelastic Modeling 62 3.1.3 Overview of Literature on EAP modeling 65 68 68 3.1.5 Summary of Literature 77 3.2 EAP Modeling framework 77 3.2.1 Kinematics of a semicrystalline, electrostrictive body 78 3.2.2 Traction-free equilibrium 84 3.2.3 Electromechanical response of a hyperelastic biphasic body with 40 32.5 Formulation dependence of polarization of crystalline domains 90 3.2.4 Orientation dependence of polarization of crystalline domains 90 3.2.5 Formulation delectrostrictive model for different net	2.2.8 Boundary Conditions	
2.3.1 Five Cases of Processing Conditions. 28 2.3.1 Distance-Based Minimum Spanning Tree Analysis. 35 2.3.2 Dispersions of Particle, Stack, and Chain Orientations for Distance-Based Spanning Trees 37 2.3.3 Dipole-Dipole Energy-Based Minimum Spanning Tree Analysis 40 2.3.4 Dispersions of Particle, Stack, and Chain Orientations for Distance-Based Spanning Trees 55 2.4. Conclusions 55 2.4. Conclusions 55 2.4. Conclusions 59 3.1 Literature Review on EAP Mechanics 59 3.1.1 Microstructure-property relationship in PVDF-based RFE polymers60 51.2 Background on Hyperelastic Modeling 3.1.2 Background on Hyperelastic Modeling 62 3.1.3 Overview of Literature on EAP modeling 65 3.1.4 Classification of Selected Models 68 3.1.5 Summary of Literature 77 3.2.1 Kinematics of a semicrystalline, electrostrictive body 78 3.2.2 Traction-free equilibrium 84 3.2.3 Electromechanical response of a hyperelastic biphasic body with 41 dipole-dipole interactions 85 3.2.4 Orientation dependence of polarization of crystalline domains 90 3.2.5 Formulation delectrostrictive model for different network	2.3 Multi-Field Processing Results and Conclusions	
2.3.1 Distance-Based Minimum Spanning Tree Analysis 35 2.3.2 Dispersions of Particle, Stack, and Chain Orientations for Distance-Based Spanning Trees 37 2.3.3 Dipole-Dipole Energy-Based Minimum Spanning Tree Analysis 40 2.3.4 Dispersions of Particle, Stack, and Chain Orientations for Distance-Based Spanning Trees 55 2.4. Conclusions of Particle, Stack, and Chain Orientations for Distance-Based Spanning Trees 55 2.4. Conclusions 56 CHAPTER 3 59 3.1.1 Microstructure-property relationship in PVDF-based RFE polymers60 51.2 3.1.2 Background on Hyperelastic Modeling 62 3.1.3 Overview of Literature on EAP modeling 65 3.1.4 Classification of Selected Models 68 3.1.5 Summary of Literature 77 3.2 EAP Modeling framework 77 3.2.1 Kinematics of a semicrystalline, electrostrictive body 78 3.2.2 Traction-free equilibrium 84 3.2.3 Electromechanical response of a hyperelastic biphasic body with 40 dipole interactions 85 3.2.4 Orientation dependence of polarization of crystalline domains 90 3.2.5 Formulation of electrostrictive model for different network models 95 3.2.6 Deriving The Permittivity of	2.3.1 Five Cases of Processing Conditions	
2.3.2 Dispersions of Particle, Stack, and Chain Orientations for Distance- Based Spanning Trees 37 2.3.3 Dipole-Dipole Energy-Based Minimum Spanning Tree Analysis 40 2.3.4 Dispersions of Particle, Stack, and Chain Orientations for Distance- Based Spanning Trees Based Spanning Trees 55 2.4. Conclusions 55 2.4. Conclusions 56 CHAPTER 3 59 3.1 Literature Review on EAP Mechanics 59 3.1.1 Microstructure-property relationship in PVDF-based RFE polymers60 51.2 3.1.2 Background on Hyperelastic Modeling 62 3.1.3 Overview of Literature on EAP modeling 62 3.1.4 Classification of Selected Models 68 3.1.5 Summary of Literature 77 3.2 EAP Modeling framework 77 3.2.1 Kinematics of a semicrystalline, electrostrictive body 78 3.2.2 Traction-free equilibrium 84 3.2.3 Electromechanical response of a hyperelastic biphasic body with 85 3.2.4 Orientation dependence of polarization of crystalline domains 90 3.2.5 Formulation of electrostrictive model for different network models 95 3.2.6 Deriving The Permittivity of the Amorphous Phase 114 <td>2.3.1 Distance-Based Minimum Spanning Tree Analysis</td> <td></td>	2.3.1 Distance-Based Minimum Spanning Tree Analysis	
Based Spanning Trees 37 2.3.3 Dipole-Dipole Energy-Based Minimum Spanning Tree Analysis 40 2.3.4 Dispersions of Particle, Stack, and Chain Orientations for Distance-Based Spanning Trees 55 2.4. Conclusions 55 2.4. Conclusions 56 CHAPTER 3 59 3.1 Literature Review on EAP Mechanics 59 3.1.1 Microstructure-property relationship in PVDF-based RFE polymers60 51.2 Background on Hyperelastic Modeling 62 3.1.3 Overview of Literature on EAP modeling 62 3.1.4 Classification of Selected Models 68 68 3.1.5 Summary of Literature 77 3.2 EAP Modeling framework 77 3.2.1 Kinematics of a semicrystalline, electrostrictive body 78 3.2.2 Traction-free equilibrium 84 3.2.3 Electromechanical response of a hyperelastic biphasic body with 61 dipole-dipole interactions 85 3.2.4 Orientation dependence of polarization of crystalline domains 90 3.2.5 Formulation of electrostrictive model for different network models 95 3.2.6 Deriving The Permittivity of the Amorphous Phase	2.3.2 Dispersions of Particle, Stack, and Chain Orientations for Dista	ance-
2.3.3 Dipole-Dipole Energy-Based Minimum Spanning Tree Analysis 40 2.3.4 Dispersions of Particle, Stack, and Chain Orientations for Distance- Based Spanning Trees 55 2.4. Conclusions 56 CHAPTER 3 59 3.1 Literature Review on EAP Mechanics 59 3.1.1 Microstructure-property relationship in PVDF-based RFE polymers60 51 3.1.2 Background on Hyperelastic Modeling 62 3.1.3 Overview of Literature on EAP modeling 65 3.1.4 Classification of Selected Models 68 3.1.5 Summary of Literature 77 3.2 EAP Modeling framework 77 3.2.1 Kinematics of a semicrystalline, electrostrictive body 78 3.2.2 Traction-free equilibrium 84 3.2.3 Electromechanical response of a hyperelastic biphasic body with 85 3.2.4 Orientation dependence of polarization of crystalline domains 90 3.2.5 Formulation of electrostrictive model for different network models 95 32.6 Deriving The Permitivity of the Amorphous Phase	Based Spanning Trees	
2.3.4 Dispersions of Particle, Stack, and Chain Orientations for Distance-Based Spanning Trees 55 2.4. Conclusions 56 CHAPTER 3 59 3.1 Literature Review on EAP Mechanics 59 3.1.1 Microstructure-property relationship in PVDF-based RFE polymers60 51 3.1.2 Background on Hyperelastic Modeling 62 3.1.3 Overview of Literature on EAP modeling 65 3.1.4 Classification of Selected Models 68 3.1.5 Summary of Literature 77 3.2 EAP Modeling framework 77 3.2.1 Kinematics of a semicrystalline, electrostrictive body 78 3.2.2 Traction-free equilibrium 84 3.2.3 Electromechanical response of a hyperelastic biphasic body with 85 3.2.4 Orientation dependence of polarization of crystalline domains 90 3.2.5 Formulation of electrostrictive model for different network models 95 3.2.6 Deriving The Permittivity of the Amorphous Phase	2.3.3 Dipole-Dipole Energy-Based Minimum Spanning Tree Analys	is40
Based Spanning Trees 55 2.4. Conclusions 56 CHAPTER 3 59 3.1 Literature Review on EAP Mechanics 59 3.1.1 Microstructure-property relationship in PVDF-based RFE polymers60 51 3.1.2 Background on Hyperelastic Modeling 62 3.1.3 Overview of Literature on EAP modeling 65 3.1.4 Classification of Selected Models 68 3.1.5 Summary of Literature 77 3.2 EAP Modeling framework 77 3.2.1 Kinematics of a semicrystalline, electrostrictive body 78 3.2.2 Traction-free equilibrium 84 3.2.3 Electromechanical response of a hyperelastic biphasic body with 85 3.2.4 Orientation dependence of polarization of crystalline domains 90 3.2.5 Formulation of electrostrictive model for different network models 95 3.2.6 Deriving The Permittivity of the Amorphous Phase 114	2.3.4 Dispersions of Particle. Stack, and Chain Orientations for Dist	ance-
2.4. Conclusions 56 2.4. Conclusions 56 CHAPTER 3 59 3.1 Literature Review on EAP Mechanics 59 3.1.1 Microstructure-property relationship in PVDF-based RFE polymers60 51.2 Background on Hyperelastic Modeling 62 3.1.3 Overview of Literature on EAP modeling 62 3.1.4 Classification of Selected Models 68 3.1.5 Summary of Literature 77 3.2 EAP Modeling framework 77 3.2.1 Kinematics of a semicrystalline, electrostrictive body 78 3.2.2 Traction-free equilibrium 84 3.2.3 Electromechanical response of a hyperelastic biphasic body with 85 dipole-dipole interactions 85 3.2.4 Orientation dependence of polarization of crystalline domains 90 3.2.5 Formulation of electrostrictive model for different network models 95 32.6 Deriving The Permittivity of the Amorphous Phase	Based Snanning Trees	55
CHAPTER 3593.1 Literature Review on EAP Mechanics593.1.1 Microstructure-property relationship in PVDF-based RFE polymers603.1.2 Background on Hyperelastic Modeling623.1.3 Overview of Literature on EAP modeling653.1.4 Classification of Selected Models683.1.5 Summary of Literature773.2 EAP Modeling framework773.2.1 Kinematics of a semicrystalline, electrostrictive body783.2.2 Traction-free equilibrium843.2.3 Electromechanical response of a hyperelastic biphasic body with85dipole-dipole interactions853.2.4 Orientation dependence of polarization of crystalline domains903.2.5 Formulation of electrostrictive model for different network models953.2.6 Deriving The Permittivity of the Amorphous Phase114	2.4. Conclusions	
3.1 Literature Review on EAP Mechanics593.1.1 Microstructure-property relationship in PVDF-based RFE polymers603.1.2 Background on Hyperelastic Modeling623.1.3 Overview of Literature on EAP modeling653.1.4 Classification of Selected Models683.1.5 Summary of Literature773.2 EAP Modeling framework773.2.1 Kinematics of a semicrystalline, electrostrictive body783.2.2 Traction-free equilibrium843.2.3 Electromechanical response of a hyperelastic biphasic body with85dipole-dipole interactions853.2.4 Orientation dependence of polarization of crystalline domains903.2.5 Formulation of electrostrictive model for different network models 95114	CHAPTER 3	59
3.1.1 Microstructure-property relationship in PVDF-based RFE polymers60 3.1.2 Background on Hyperelastic Modeling	3.1 Literature Review on EAP Mechanics	
3.1.2 Background on Hyperelastic Modeling	3.1.1 Microstructure-property relationship in PVDF-based RFE poly	mers60
3.1.3 Overview of Literature on EAP modeling	3.1.2 Background on Hyperelastic Modeling	
3.1.4 Classification of Selected Models 68 3.1.5 Summary of Literature 77 3.2 EAP Modeling framework 77 3.2.1 Kinematics of a semicrystalline, electrostrictive body 78 3.2.2 Traction-free equilibrium 84 3.2.3 Electromechanical response of a hyperelastic biphasic body with 85 3.2.4 Orientation dependence of polarization of crystalline domains 90 3.2.5 Formulation of electrostrictive model for different network models 95 3.2.6 Deriving The Permittivity of the Amorphous Phase 114	3.1.3 Overview of Literature on EAP modeling	
3.1.5 Summary of Literature 77 3.2 EAP Modeling framework 77 3.2.1 Kinematics of a semicrystalline, electrostrictive body 78 3.2.2 Traction-free equilibrium 84 3.2.3 Electromechanical response of a hyperelastic biphasic body with dipole-dipole interactions 85 3.2.4 Orientation dependence of polarization of crystalline domains 90 3.2.5 Formulation of electrostrictive model for different network models 95 3.2.6 Deriving The Permittivity of the Amorphous Phase 114	3.1.4 Classification of Selected Models	
3.2 EAP Modeling framework 77 3.2.1 Kinematics of a semicrystalline, electrostrictive body 78 3.2.2 Traction-free equilibrium 84 3.2.3 Electromechanical response of a hyperelastic biphasic body with 85 3.2.4 Orientation dependence of polarization of crystalline domains 90 3.2.5 Formulation of electrostrictive model for different network models 95 3.2.6 Deriving The Permittivity of the Amorphous Phase 114	3.1.5 Summary of Literature	
3.2.1 Kinematics of a semicrystalline, electrostrictive body 78 3.2.2 Traction-free equilibrium 84 3.2.3 Electromechanical response of a hyperelastic biphasic body with 84 dipole-dipole interactions 85 3.2.4 Orientation dependence of polarization of crystalline domains 90 3.2.5 Formulation of electrostrictive model for different network models 95 3.2.6 Deriving The Permittivity of the Amorphous Phase 114	3.2 EAP Modeling framework	
3.2.2 Traction-free equilibrium	3.2.1 Kinematics of a semicrystalline electrostrictive body	78
3.2.3 Electromechanical response of a hyperelastic biphasic body with dipole-dipole interactions	3.2.2.7 Traction-free equilibrium	84
dipole-dipole interactions	3.2.2 Fluction free equilibrium sectors of a hyperelastic hiphasic body wi	th
3.2.4 Orientation dependence of polarization of crystalline domains 90 3.2.5 Formulation of electrostrictive model for different network models 95 3.2.6 Deriving The Permittivity of the Amorphous Phase	dipole-dipole interactions	
3.2.5 Formulation of electrostrictive model for different network models 95 3.2.6 Deriving The Permittivity of the Amorphous Phase	3.2.4 Orientation dependence of polarization of crystalline domains	90
3.2.6 Deriving The Permittivity of the Amorphous Phase	3.2.5 Formulation of electrostrictive model for different network mo	dels 95
	3.2.5 1 Ormanation of electrostrictive model for american herework mo	114
3 3 Results and Discussions of Microstructure-Based Model 117	3 3 Results and Discussions of Microstructure-Rased Model	117
3.3.1 Comparisons of network model responses to experimental data 117	3.3.1 Comparisons of network model responses to experimental data	117

TABLE OF CONTENTS

3.4 Conclusions		124
CHAPTER 4		126
4.1 Introduction to Self-Actuated Origami-Inspired Devices		126
4.1.1 Self-Folding Structures	127	
4.1.2 Compliant Mechanisms	127	
4.1.3 Magneto-Active Structures	128	
4.1.4 Electro-Active Structures	129	
4.1.5 Multi-Field Actuated Structures	130	
4.1.6 Multi-field Bimorph Design	131	
4.2 Modeling Multi-Laver Multi-Segment Composites Methodology		131
4.2.1 Model Framework	132	
4.2.2 Model Validation for Basic Configurations	137	
4.2.3 Objective Functions	141	
4.2.4 Formal Optimization Setup	143	
4.2.5 Case Study for Parameter Study and Optimization	144	
4.2.6 Chosen Parameters	145	
4.2.7 Parameter Study Formulation and Results	146	
4.2.8 Formal Optimization Problem Results and Discussions	151	
4.3 Conclusions on Self-Folding Composite Optimization		156
CHAPTER 5		159
5.1 Conclusions and Future Works		150
5.3 Funding Acknowledgements		157
5.5 I ununig Acknowledgements		102
REFERENCES		163
APPENDIX A		170
APPENDIX B		173
B.1 Permittivity of the Amorphous Phase		173
B.2. Obtaining the saturation polarization parameters for the dipoles	••••••	177
APPENDIX C		180
C.1 Mathematical Formulation: Part I – Constitutive Response		180
APPENDIX D		188
APPENDIX E		192

LIST OF FIGURES

Figure 1-1. Modeling dipole-driven materials can be viewed as modeling several types of materials that can be actuated based on electric or magnetic dipoles. Grand challenges of engineering, such as enhancing virtual reality, producing renewable energy, and improving infrastructure, can be addressed by modeling electro-active polymers (EAPs), magneto-active elastomers (MAEs) and other analogous systems with the same modeling tools utilizing dipole physics. Examples include the Wolverine virtual reality interface for grasping virtual objects, vibrating electro-active energy harvesters, 3D printed magnetic structures capable of delivering drugs, and magnetically controlled micro-robots able to perform targeted drug delivery in mice... 3

Figure 1-2. Examples of self-folding active structures employing dipole-driven materials are shown. A magnetically actuated box-design is shown in (a) (Ahmed, Lauff et al.); an electrically actuated origami-inspired "barking dog" design is shown in (b) (Bowen, Springsteen, Ahmed, et al.); and an electrically actuated biomorphic origami-inspired device is shown in (c) (Okuzaki et al.). The actuation mechanisms of the active materials used in these examples are all dipole-driven.

Figure 2-1. Barium hexaferrite particles are typically in the shape of hexagonal platelets. The permanent magnetic moment follows the easy axis as shown in (a), and the approximate size of micro-BHF is 1000 nm by 150 nm as indicated. When a magnetic field H is experienced, as shown in (b), the dipole moment m generates a torque to align itself with the direction of the field. When an electric field E is experienced, an electric dipole moment *pind* is induced in the plane, and subsequently transverse to m. Similarly, *pind* generates a torque to align itself with E...... 13

Figure 2-2. Randomly oriented hard magnetic particles are embedded in a polymer matrix. Applying **H0** while the matrix is a liquid during curing yields chain formations and particle orientations both in the direction of **H0**. Applying **H0** and **E0** simultaneously in transverse directions yields chain formations in the direction of **E0** and particle orientations aligned with **H0**.

Figure 2-5. Nearest neighbor periodic boundary condition presented in 2D. Control volume is highlighted in bolded rectangular outline in the center of the grid. Particles leaving the CV are

assumed to enter a neighboring region as shown and enter the CV from the opposite edge due to the periodic boundary condition
Figure 2-6. Visual representation of starting positions of BHF particles in the control volume. Arrows indicate directions of magnetic dipole moments of each particle
Figure 2-7. Three possible levels of structure with the base unit of a BHF particle, represented by the ellipsoids. Net magnetization of stacks are indicated by the red arrows. The smallest structure is (a) a stack. The medium structure is (b) a micro-aggregate consisting of multiple stacks. The largest structure is (c) a macro-chain consisting of micro-aggregates
Figure 2-8. The five cases are shown with simplified 2D schematics illustrating particles represented by ellipsoids and chains indicated by outlines of connected particles
Figure 2-9. Simulation results for Case 1, the no-field case. Particles are represented by ellipsoids, with magnetic dipole moments signaled by the blue arrows
Figure 2-10. Simulation results for Case 2, the electric field only case. Particles are represented by ellipsoids, with magnetic dipole moments signaled by the blue arrows
Figure 2-11. Simulation results for Case 3, the magnetic field only case. Particles are represented by ellipsoids, with magnetic dipole moments signaled by the blue arrows
Figure 2-12. Simulation results for Case 4, the electric and magnetic fields in transverse case. Particles are represented by ellipsoids, with magnetic dipole moments signaled by the blue arrows and electric dipole moments by red arrows
Figure 2-13. Simulation results for Case 5, the electric and magnetic fields in parallel case. Particles are represented by ellipsoids, with magnetic dipole moments signaled by the blue arrows and electric dipole moments by red arrows
Figure 2-14. The simulation results for $N = 200$ particles are shown under (a) magnetic field only, and (b) electric and magnetic fields. Each ellipsoid represents a BHF particle, and the arrows indicate their dipole moment orientations (blue is magnetic and red is electric)
Figure 2-15. The number of trees is plotted versus the range <i>dtree/Req</i> used in calculating the minimum spanning trees for Case 5. The connections within the trees are shown inside the boxes, color coded with the plateau ranges
Figure 2-16. Angle of a particle and the angles in the edges within a tree are defined as shown 38
Figure 3-1. Polarization curves for a ferroelectric material and a relaxor ferroelectric material 61
Figure 3-2. Two-dimensional schematic of a biphasic body (i.e., a body with inclusions) undergoing deformation. The left body is in the undeformed configuration and the body on the right is in the deformed configuration. Two dipolar regions are highlighted with electric dipole moments $p1$ and $p2$ with a vector R between them in the undeformed configuration and r in the deformed configuration

Figure 3-3. The representative volume element contains a spherical crystalline domain, surrounded by a concentric amorphous domain that is initially spherical and deforms into an ellipsoidal shape after deformation
Figure 3-4. The deformation of a segmented element consisting of crystalline and amorphous phases is presented
Figure 3-5. Variables defining the orientations and separation of two dipole moments with respect to the basis vector $e3$. The angles calculated in equation (3-84) are visually depicted at relevant locations; $\alpha 1$ is drawn separately for simplicity
Figure 3-6. Relationship between ϕp and E as calculated by (3-67)
Figure 3-7. A von Mises distribution function is chosen as the PDF of the electric dipoles. With the implementation of $b\phi p = b\phi p(E)$ solved in (3-67), the model represents a set of dipoles progressively aligning with the field direction ($\phi p = 0$) as <i>E</i> is increased
Figure 3-8. The RVE is represented by a sphere annotated with quantities with a subscript of 1, while a neighboring RVE is indicated by the second sphere annotated with quantities with subscript 2. Depiction of spheres in (a) represent the undeformed shapes of the RVE and neighboring body with no field present, at an initial relative orientation of crystallites defined, and (b) illustrates the deformed shapes of the RVE and neighboring body under an applied field along <i>e</i> 3, creating nominal <i>e</i> 3-alignment and dipole-dipole forces, and subsequent uniaxial compression of the RVE. Please note that the neighboring RVE display does not represent a spherical contact problem
Figure 3-9. The discrete network model is shown for two specific cases. The parallel case is shown in (a), where the vector to nearest neighboring crystalline domain, \boldsymbol{r} , is parallel to the external field, while in the perpendicular case in (b) \boldsymbol{r} is perpendicular to the external field 101
Figure 3-10. The non-dimensionalized energies (a) and the corresponding forces (b) of the two cases of the single chain network model, parallel and perpendicular, are plotted versus the stretch in the direction of the applied electric field. The comparisons highlight key differences in the strengths of the interactive energies/forces between the parallel and perpendicular cases
Figure 3-11. The left- and right-hand sides of (3-98) are plotted versus λ for the extreme cases, (a) parallel single chain, and (b) perpendicular single chain. The blue line is the stress difference associated with the dipolar energy. The intersections of the curves indicate the equilibrium locations.
Figure 3-12. The discretized distribution function in (3-105) is evaluated at $WC\Phi ri = 1$ for varying values of $N\Phi r$
Figure 3-13. The von Mises PDF is graphically represented by a scatter plot of randomly generated points based on the distribution evaluated at varying values of κ . The PDF is centered either at $\mu \Phi r = 0$ and $\mu \Phi r = \pi/2$ as shown, where these distributions move toward approximating the parallel and perpendicular discrete single chain models

Figure 3-14. Comparison of the two extreme cases with PDFs in prediction of electrostriction. As $\kappa \to 1/3$, both PDF-based responses converge toward the discrete network responses, which is why the discrete network models are placed at the low end of the κ range. Due to theoretical material failure, electrostriction could not be determined at field strengths above $E *= 15$ for $\mu \Phi r = 0$ as $\kappa \to 0$; as a result, saturation is not observed at (c)
Figure 3-15. The single-phase polarization model is fitted to the polarization responses of three data sets. The fitted parameters <i>Ps</i> and <i>Es</i> (in legend) are also listed in Table 3-3
Figure 3-16. Tensile test data for the CTFE 7.8% terpolymer, and two best-fit models: modified 8- chain hyperelastic model $W8ch(I1A)$ with crystallinity $vc = 0$ and $vc = 0.36$
Figure 3-17. Comparison of the electrostriction model evaluated at varying distribution parameters with experiments for P(VDF-TrFE-CTFE) 7.8%
Figure 3-18. Comparison of the electrostriction model evaluated at $0 < \kappa < 1/3$ for $\mu \Phi = 0$ and $0.05 < \kappa < 0.3$ for $\mu \Phi = \pi/2$ with experiments for P(VDF-TrFE-CTFE) 9.1%
Figure 3-19. Comparison of the electrostriction model evaluated at varying distribution parameters with experiments for P(VDF-TrFE-CFE) 3.8%
Figure 4-1. A magneto-active body is represented by a rectangle in (a), with its magnetization M indicated by the red arrows. An external field H induces a torque T in the body, as shown in (b). The body's Zeeman energy is minimized by aligning M with H as shown in (c)
Figure 4-2. Bimorph composed of an EAP and MAE layer. Magnetic actuation yields bending towards the left, while electric actuation yields bending towards the right
Figure 4-3. In (a), a schematic of a sample multi-field composite beam with an arbitrary number of segments, each consisting of an arbitrary number of layers, is shown. Each layer of each segment can have an arbitrary material type, denoted by the color code. In (b), a differential element of the composite beam at an arbitrary location along its length is provided, with cut faces subject to normal stresses and shear forces
Figure 4-4. A tip force is applied to one end of the beam. The beam is cut at a location to show the angle of the location and the normal and shear forces acting at the cut surface
Figure 4-5. Resultant, R, is measured as the tip displacement from before and after actuation 137
Figure 4-6. Experimental and modeling results for the EAP-based (terpolymer) unimorph. The figures in (a) and (b) present a deformed shape comparison at 60 MV/m, while (c) is the layer configuration of the unimorph, and (d) shows the tip displacement results. R is the resultant tip displacement and L is the original length when the beam is flat
Figure 4-7. Model and experiments compared for the bimorph with two separate fields turned on. (a) compares the tip displacements for the electrical actuation of the bimorph while (b) is for the magnetic actuation. 139

Figure 4-8. Simulations of the bimorph under (a) constant electric field with varying magnetic fields, and (b) constant magnetic field with varying electric fields
Figure 4-9. Target shape drawn as a line through the structure's neutral axis. Segments are identified between points along the shape's path
Figure 4-10. The location of the tip force is shown on the structure schematic
Figure 4-11. Schematic of a simple bimorph composed of eight segments with j layers, fixed on the left end. An external magnetic field H is applied upward. Layer 1 is an EAP (green); layer 2 is a passive material (blue); even numbered segments contain a layer 3, which is an MAE (magnetization directions signaled by yellow arrows). Dimensions are not to scale
Figure 4-12: Chosen length parameter is visualized on a sample configuration. Red layers are MAEs, blue layers are passive, and green layers are EAPs. The yellow arrows on the MAE patches indicate direction of magnetization. From Y1 to Y5, segments containing MAEs increase in length, while the remaining segments reduce in length
Figure 4-13. The performance space is shown. Each dot represents a simulation of a different combination of M , YP , and Υ . The length ratio, Υ , is indicated by color. The colored arrows on the bottom left indicate the best of the feasible designs
Figure 4-14. Deformed shapes of the three best designs with overlays of the target shape (red lines). From left to right: L2-M5-Y1, L3-M4-Y1, L4-M3-Y1
Figure 4-15 . The Pareto front for the three objective functions is shown in a 3D plot, as obtained from the genetic algorithm. Each point represents an optimal individual based on the objective functions
Figure 5-1 . A full-scale optimization problem may only require material set properties and structure configuration such that the only variables can be processing conditions, and the outputs structural response relating to multiple objectives. This way, an indirect link between processing conditions and structural response can be obtained
Figure 5-2. The relative permittivity (dielectric constant) is plotted versus electric field. The derivative approximation is obtained from the discretized points from the data set in Ref [2], and the linear dielectric approximation is the set of calculated constants based on the linear dielectric energy density equation. The derivative curve fit is the least-error biphasic model to predict the high field permittivity values based on the derivative approximation
Figure 5-3. The dielectric constant versus electric field response is extracted from the polarization responses found in (a) Ref [20] and (b) Ref [16]. The high field dielectric constant is 7.7 for (a) and 11.7 for (b)
Figure 5-4. The dielectric constant for P(VDF-TrFE-CTFE) 61.8/30.4/7.8%, as calculated by applying the derivative method to experimentally measured values of polarization versus electric field. The amorphous phase's relative permittivity, εa , is 1

xi

Figure 5-5.The dielectric constant for P(VDF-TrFE-CTFE) 61.8/30.4/7.8%, as calculated by applying the derivative method to experimentally measured values of polarization versus electric field for only the high field response. The biphasic model is fitted to both one standard deviation above and below the average experimental values and fitted again to the average measurements.

Figure 5-6. The polarization response and the fitted single phase polarization model are compared for (a) P(VDF-TrFE-CTFE) 61.8/30.4/7.8%, (b) P(VDF-TrFE-CFE) 62/38/4%, and (c) P(VDF-
TrFE-CTFE) 65/35/10%. The optimal parameters are listed in Table 5-2
Figure 5-7. The procedures of the iterative solution method for the curvatures with varying angles.

Figure 5-8. An infinitesimal segment along the ar	c, which produces a linear relation between dx
and dz	

LIST OF TABLES

Table 2-1. Comparative degree of orientation of BHF stack and micro-chains. Dispersion factors, (κ) are provided for the stack range, small chain range, and large chain range for each case 39
Table 3-1. The model types and their definitions based on the parameters modeled. 69
Table 3-2. Comparisons of the various models discussed with respect to the relevant criteria75
Table 3-3. Terpolymers are listed with chemical composition and processing details. The chemical compositions may differ from the literature because they have been normalized here to add up to 100%.
Table 3-4. Parameters used for the simulations in Figure 15
Table 5-1. Material parameters. 138
Table 5-2. All possible geometric quantities and material properties for all layers in each segmentfor the chosen four MAE patch bimorph configuration.147
Table 5-3. Material and layer properties for MAE, EAP, and the passive layer
Table 5-4. Objective function evaluations and magnetizations for the highlighted in designs in Figure 11. 153
Table 7-1.Terpolymer high field permittivities are recorded based on least-squaring of the biphasic model to the derivative approximation data
Table 7-2. The optimized parameters from the single phase model for polarization

ACKNOWLEDGEMENTS

First and foremost, I would like to thank my advisor, **Dr. Paris von Lockette**, who provided invaluable guidance, patience, and support throughout my studies. I am also grateful for his mentoring when it came to important decisions in life, whether for a career in academia or beyond it.

Secondly, I would like to thank all of my committee members for their support and availability that helped me make progress on my thesis; **Dr. Zoubeida Ounaies** for her constant feedback in weekly meetings, and expert knowledge on active materials; **Dr. Mary Frecker** for her insightful comments in weekly meetings, and expert knowledge on optimization; **Dr. Francesco Costanzo** for offering the best courses I have ever taken, providing the level of rigor an engineering student studying mechanics could only wish for.

I am grateful and privileged to have parents, **Mediha and Erdal Erol**, who have been immensely supporting and caring throughout my life, to this day. They have not only shown moral and financial support during my graduate studies, but also served as primary role models for me, motivating me to improve myself both as a professional and as a person. Additionally, I am grateful to have my brother, **Akin**, who has been incredibly supportive and motivated me to keep going even during the most challenging times.

I would also like to thank my friends and colleagues; particularly my lab-mates, **Corey Breznak**, **Manuel Rodriguez**, **Brett Cowan**, **Kevin Li**, as well as my colleagues from other labs including Saad Ahmed, **Abdullah Masud**, **Landen Bowen**, and all the other members of the **EFRI group** at Penn State.

Finally, I could never thank **Deniz** enough for her constant moral support and help with my research, especially her willingness to proofread my writing despite how long it sometimes was. Without her, I would not have been able to finish my research with a sound mind.

CHAPTER 1

Motivation, Goals, and Hypotheses

The first chapter of this document provides context and motivation for this work, focusing on examples of how this work can impact the world through addressing grand challenges *via* applications. Various hypotheses are introduced, along with corresponding objectives, aiming to make progress towards potential impacts. The remaining chapters are associated with different objectives testing the hypotheses, each assigned a literature review chapter relevant to the hypothesis. Thus, the literature reviews for specific methodologies are not included in this chapter, and instead spared for other chapters.

This chapter is divided into four subsections. The first subsection discusses the motivation, highlighting the significance of studying dipole-driven materials and self-actuated Origami-inspired devices. Specifically, it provides the impacts of studying active materials and structures on different industries, with a focus on electro-active polymers and other dipole-driven materials. The second subsection briefly summarizes the goals for advancing the field of dipole-driven active materials and structures. The third subsection presents a set of hypotheses that can be tested by a set of objectives. Answering the hypotheses will expand our knowledge on the field of dipole-driven active materials and self-folding devices. The fourth subsection is a summary.

1.1 Impacts of Dipole-Driven Materials

Dipoles are found throughout nature as pairs of oppositely charged poles on various molecules and substances. Each pole exerts a field on their surroundings, and when observed from a far enough distance, the sum of the poles can be viewed as a single object (i.e. a dipole). One form of the field given by a dipole is

$$\Xi = \frac{1}{4\pi c_{\Xi} r^3} [3(\boldsymbol{m} \cdot \hat{\boldsymbol{r}}) \hat{\boldsymbol{r}} - \boldsymbol{m}], \qquad (2-1)$$

where \hat{r} is the unit vector of a distance vector r from the center of the dipole, r is the magnitude of r, m is the moment of a dipole, and C_{Ξ} is the constant associated with the type of field and moment (e.g., electric or magnetic). The forces and torques a dipole experiences are based on the field it is experiencing. The forms of the force, F_{Ξ} , and torque, T_{Ξ} , are

$$F_{\Xi} = (\boldsymbol{m} \cdot \boldsymbol{\nabla})\boldsymbol{\Xi}, \tag{2-2}$$

$$T_{\Xi} = m \times \Xi, \tag{2-3}$$

Dipole-driven materials are active materials whose behaviors are driven by the interactions of dipoles resulting from (2-2) and (2-3). While this broad definition is not widely or explicitly used in literature, it is an important classification, because the similarities between the physics of different types of dipoles (e.g. electric versus magnetic dipoles) means that understanding one type of dipole-driven material can aid in the understanding of other dipole-driven materials, lending broad applicability to results developed in this area.

Another justification for this broad material classification is the substantial amount of research that has been invested into the development of various types of dipole-driven materials, whose applications range from actuators to energy harvesting devices, as shown in Figure 2-1. Due to the breadth of applications, studying dipole-driven materials can impact several grand challenges, such as producing renewable energy, enhancing virtual reality, improving infrastructure, and engineering better medicines (Choi et al. 2016; Erturk 2011; R. Zhao et al. 2019; Alapan et al. 2019). Figure 2-1 highlights examples of how modeling dipole-driven materials such as electro-active polymers (EAPs) and magneto-active elastomers (MAEs) can aid in understanding of various material systems. Predicting each material system will provide insight into the various problems that will help us solve the related engineering challenges. For example, predicting the actuation force of EAPs can aid in controlling the "Wolverine" virtual reality (Choi et al. 2016). Similarly, predicting the electromechanical response of EAPs can help

understand the data from damage sensing applications, where the relationship between the generated stress and the bending or vibration of EAPs must be precisely known (Erturk 2011). Additionally, magnetic dipole based models have contributed to our understanding of hard magnetic composite behavior, which have been controlled by external magnets to navigate through mice to deliver drugs to target regions (Alapan et al. 2019).



Figure 2-1. Modeling dipole-driven materials can be viewed as modeling several types of materials that can be actuated based on electric or magnetic dipoles. Grand challenges of engineering, such as enhancing virtual reality, producing renewable energy, and improving infrastructure, can be addressed by modeling electro-active polymers (EAPs), magneto-active elastomers (MAEs) and other analogous systems with the same modeling tools utilizing dipole physics. Examples include the Wolverine virtual reality interface for grasping virtual objects, vibrating electro-active energy harvesters, 3D printed magnetic structures capable of delivering drugs, and magnetically controlled micro-robots able to perform targeted drug delivery in mice. The borrowed images are from the following references (in top to bottom order): (Choi et al. 2016; Erturk 2011; R. Zhao et al. 2019; Alapan et al. 2019)

Dipole-driven materials, particularly EAPs and MAEs, have shown promise specifically

in the field of self-folded devices. Examples shown in Figure 2-2 include electric (EAP) and

magnetic (MAE) dipole based materials, and utilize designs inspired by Origami and Kirigami

patterns, which are part of an ancient Japanese tradition of folding paper. The placement and

orientations of MAEs (represented by the black rectangular patches) in the structure in Figure 2-2.a influence the generated torques that lift the four arms (Ahmed et al. 2013). Likewise, the placement of the EAP in the Origami-inspired structure in Figure 2-2.b can generate the type of bending needed to close the annotated angle (Bowen et al. 2017). The example in Figure 2-2.c is bio- and Origami-inspired, utilizing an EAP to fold the segments of its body to generate locomotion (Okuzaki et al. 2008). Taking advantage of Origami patterns, as in the case of the electrically actuated biomorph in Figure 2-2 allows for efficient use of space and the ability to morph into a complex shape.



Figure 2-2. Examples of self-folding active structures employing dipole-driven materials are shown. A magnetically actuated box-design is shown in (a) (Ahmed et al. 2013); an electrically actuated origami-inspired "barking dog" design is shown in (b) (Bowen et al. 2017); and an electrically actuated biomorphic origami-inspired device is shown in (c) (Okuzaki et al. 2008). The actuation mechanisms of the active materials used in these examples are all dipole-driven.

1.2 Goals and Scope of Study

This work aims to develop a modeling framework for dipole-driven materials that can be used for finding feasible designs employing dipole-driven materials for some of the applications presented in Figure 2-1 in the direction of Origami-inspired self-folding designs (Figure 2-2). To test the methodologies, the modeling methods will be used to predict self-folding devices that can achieve a desired shape through actuation and generate work output.

Achieving this goal requires understanding and controlling an active structure's design process from material fabrication to structural testing. As a result, the steps between fabricating materials and testing a composite design must be identified. This study selects three links (Figure 2-3) that can be understood with the current knowledge and technology available: The relationships between

(I) <u>Processing and Microstructure</u>: During processing, small electromagnetically sensitive particulates are shown to develop order in the presence of an external field, forming aggregates with internal order, and varying degrees of order across aggregates. The relationship between how the external field contributes to the order and forming of aggregates is the studied link between processing and microstructure. Figure 2-3 illustrates the link with a block containing particulates with direction, initially randomly oriented and dispersed, and a second block containing aggregates of particles aligned in the direction of the applied field.

(II) <u>Microstructure and Material Response</u>: The ordering of aggregates effectively alters the properties of the composite and can be actuated via an external field. The actuation is a result of the interactions between particles within aggregates and interactions across aggregates, based on the fundamental dipole-based forces and torques provided by (2-1) and (2-2). The aggregates are shown as blue brick-like structures in Figure 2-3. The ordering of aggregates can form chain-like structures, which can be actuated by an external field to induce interactions between them. At this level, the medium is assumed to be a solid, elastic material, which can deform under the interactions of aggregates, yielding macro-scale deformation across the bulk material. (III) <u>Material Response and Structural Deformation</u>: The active properties induced by aggregate ordering in a material can be harnessed in a composite structure. Link 3 seeks to learn how the the material response resulting from Link 2 can influence the behavior of a composite structure containing the aggregate-based material. Thus, Figure 2-3 contains an image of a composite for Link 3, containing the aggregate-based material, along with other possibly active or passive material layers. The flat, multi-layer composite is connected to images of a multi-layer bending device employing EAPs, as an example of the type of structure that can be studied.

Since each link is dependent on numerous factors, the studies will be reduced to understanding the relationships between selected variables that are currently not understood and/or may provide a high level of control in the design process.



Figure 2-3. The links connecting material processing techniques and structural actuation are shown explicitly. Link I relates a processing on a mixture of particles and elastomer substrate with an ordered microstructure. Link II relates an ordered microstructure with an actuation response. Link III relates material responses to structural deformation of an active composite. Note: Iconography in images change for consistency with the types of material studied in different chapters of this document. Image of unimorph bender is from (W. Zhang, Ahmed, et al. 2018).

Since the goal of this study is to develop a framework for dipole-driven materials that

spans processing to structural response, understanding the driving mechanism for actuation

comprises a large portion of this work. Specifically, it is crucial that the coupling behavior in a dipole-driven material, such as an EAP, is understood well enough to accurately predict its electromechanical response—its ability to produce mechanical strain from an applied electric field—given any feasible microstructure. In fact, the electromechanical response of EAPs has been the focus of numerous studies because it is directly utilized in sensors/actuators and energy harvesting devices. If EAPs can be modeled with physics-based approaches that can link microstructure to response, then researchers can gain significant insights into how the materials behave and how their performances can be improved. The details on the gaps and the proposed methods are in Sections 2.1 and 3.1.

Microstructure-based modeling of the mechanics of EAPs is at the meso-scale, linking micro-scale information to macro-scale response. Thus, the remainder of this work is on understanding how processing conditions may influence the micro-scale properties of a composite/semicrystalline material, and how to find an optimal design of a structure employing active materials. The next subsection identifies hypotheses that can help reach these goals.

1.3 Goals and Hypotheses

The three links in this work serve as three goals: to find the relationships related to each length-scale,

- **MICRO** (Processing to Microstructure)
- **MESO** (Microstructure to Material Response)
- MACRO (Material Response to Structural Response)

Six hypotheses are formed and divided into groups based on which goal they are related to. Additionally, each hypothesis is accompanied by objectives that can be carried out with the aim of testing the hypothesis.

The first hypothesis aims to make progress toward the first goal (processing-structure relationship). The second, third, and fourth hypotheses are directed towards microstructure-based

modeling. The fifth and sixth hypotheses serve to answer questions about composite structure optimization.

Hypothesis 1: Particle simulations utilizing ferrohydrodynamics could provide evidence for whether applying magnetic and electric fields to particles suspended in a fluidic polymer matrix yields a microstructure with multiple levels of hierarchy.

Objective 1: Develop a dynamic particle simulation model that can accommodate particle motions and orientations under forces and torques induced by external electric and magnetic fields. For the analysis, identify an objective metric that can be used to assess and compare key aspects of the microstructure across different length-scales, and between the simulations and experimental results.

Hypothesis 2: Network models of semicrystalline hyperelasticity accounting for microstructure by representing different relative locations of crystalline regions can form an effective framework for modeling electrostriction while incorporating microstructural features such that experimental data for electrostriction can be found within the range of model responses by varying characteristics of microstructure.

Objective 2.1: Define a base network model that accommodates aspects of the microstructure such as semicrystallinity and spatial arrangements of dipolar (or crystalline) regions. The network model should be expandable to distributions of spatial arrangements of crystalline regions, offering higher fidelity modeling.

Objective 2.2: Calculate the dipole-dipole interaction energy by randomly generating orientations based on a von Mises orientation distribution function.

Objective 2.3: Extend the network model developed by addressing Objective 1 to allow for a probability density distribution (PDF) function to represent spatial arrangements of polar regions and compare each model's response to data.Objective 2.4: Select spatial PDFs that are experimentally feasible and compare them to experimental data in terms of electrostrictive response.

Hypothesis 3: An analytical model for multi-layer, segmented composite beams incorporating the EAP constitutive model and magnetic torques can accurately predict multi-field device response, and can be used for the design optimization for a target shape containing ideal folds.

Objective 3.1: Derive an analytical model for the bending of an arbitrarily segmented, multi-layered, multi-field actuating composite beam that accounts for large displacements, dipole-based electrostrictive constitutive relations of EAPs, and magnetic dipoles of MAEs.

Objective 3.2: Define and execute a multi-objective design optimization problem on a case study with preset material types and fixed number of segments for work, shape, and cost. The shape function must contain ideal folds, and the parameters must include segment lengths and the Young's modulus of at least one material. The results of the multi-objective optimization will determine how well folding can be achieved by segment length variation.

1.4 Outline of Document

The three hypotheses are associated with the three links shown in Figure 2-3, each corresponding to a chapter in this document in the same order. Chapter 2 answers hypothesis 1 by developing simulation methodologies to study the effects of multi-field processing on the microstructure of an embedded composite material, which is Link 1. The literature review for Link 1 can be found in Chapter 2, Section 2.1. Chapter 3 answers hypothesis 2 by developing and validating a model for the electromechanical response of dipolar EAPs that is based on mechanisms found in the microstructure of the material (i.e. Link 2). The literature review for Link 2 can be found in Chapter 3, Section 3.1. Chapter 4 answers hypothesis 3 by modeling and optimizing multi-field actuated, multi-layered composites, which incorporate electrically and magnetically actuated materials, with the aim to build devices that can achieve complex shapes and folding to be implemented into Origami-inspired designs. By focusing on how dipole-based

materials such as EAPs can generate deformation in a structure, Chapter 4 is on Link 3. The literature review for Link 3 can be found in Chapter 4, Section 4.1.

Additional content can be found in the Appendices, including scripts used to evaluate the models, intermediary calculations/derivations, and methodologies for extracting parameters from experiments.

CHAPTER 2

Link Between Processing Conditions and Microstructure

This chapter is on studying the effects of multi-field processing on microstructural evolution of composites. The chapter begins with a review of the significance of multi-field processing by highlighting its impacts on developing a universal 3D printer, and a literature review on past works aimed to manipulate microstructures of composites with various processing methods. Next, a modeling technique based on dynamic particle simulations is selected and formulated for a generalized composite structure exposed to arbitrary fields. The model is reduced to a few specific cases for microstructure evolution simulations and the results are analyzed via image processing for comparison to experimental data.

The first chapter aims to address Hypothesis 1 by comparing different network models and incorporating a distribution function for both the spatial and polarization-related orientations of dipole moments. Note: Portions of this document are borrowed from the author's contributions to a 2019 SPIE Smart Structures and Non-destructive Evaluation conference proceeding (M. Al Masud et al. 2019).

2.1 Background on Multi-Field Processing

Composite materials have been extensively studied because their material properties can be manipulated by selecting different combinations of constituent sets, varying constituent ratios, and varying spatial arrangements of constituents (Stankovich et al. 2006; Chawla 2012). The advent of additive manufacturing has enabled more diverse designs and a convenient method of manipulating filler material (X. Wang et al. 2017). For instance, filler material such as carbon-fiber can be aligned and oriented within a geometry by utilizing the printing direction, which leads to a change in bulk material properties such as the Young's modulus (Mahajan and Cormier 2015; Pierson et al. 2019).

In addition to print direction, particles can also be aligned using external fields. For example, magnetic fields can be used in multi-material 3D printers to align particles in a specific direction, achieving macroscopic arrangements inside a polymetric matrix for soft magnetic particles with anisotropic geometries and hard magnetic particles (Kokkinis, Schaffner, and Studart 2015; R. Zhao et al. 2019). Electric fields have also been used in particle alignment or assembly due to dielectrophoretic interactions between particles and between particles and field (Velev and Bhatt 2006). One study found that the combined effects of electric and magnetic fields on micro- and nano- dipolar particles can assemble particles in multiple directions due to the dipole-dipole interactions between micro- and nano-particles (Bharti et al. 2016). These methods rely on applying the field to the composite while the polymer matrix is in liquid phase such that the particles are free to move and rotate and possibly develop micro-archtectures.

Manipulating particles in a composite material is technologically important because changes in microstructure can alter the properties of the bulk material. For example, the formation of chains via particle assembly under a magnetic field can yield anisotropy in the composite's elastic modulus (Varga, Filipcsei, and Zrínyi 2006), electrical conductivity (Jin, Tiefel, and Wolfe 1992; Leng et al. 2008), and magnetic polarization (Breznak and Lockette 2019). Yet these examples are a small group of possible properties that can be varied. Ultimately, a universal 3D printer with the ability to program a wide range of properties such as high to low compliance or insulative to conductive is one of the goals of additive manufacturing. Currently, this is accomplished using multi-material printers having multiple dispensers or drawing from multiple reservoirs per material or phase. This work seeks to develop simulation methods aiding the additive manufacturing technologies capable of producing components with dichotomous properties solely by varying processing conditions on raw material drawn from reservoir having a single constituent set. Process variations are intended to lead to varying micro-architectures from which differentiation of properties is derived. Such a 3D printer could tailor multiple material properties to varying degrees by manipulating the microstructure of a single constituent set, which can be optimized for the applications mentioned in Chapter 1.

A recent experimental study made progress toward a universal 3D printer by demonstrating that the simultaneous application of electric and magnetic fields to a polymer matrix containing Barium Hexaferrite (BHF) particles can create a hierarchy of structure that could potentially influence multiple bulk properties of the composite (M. Al Masud et al. 2019). Barium hexaferrite particles are nominally hexagonal flat plates that are hard magnetic, exhibiting a permanent magnetic dipole moment m, as shown in Figure 3-1. When the particle experiences a magnetic field H, the moment m generates a torque to align with the field. Furthermore, while the particle does not exhibit a permanent electric dipole moment, an electric dipole moment can be induced with the application of an external field E due to the geometry.



Figure 3-1. Barium hexaferrite particles are typically in the shape of hexagonal platelets. The permanent magnetic moment follows the easy axis as shown in (a), and the approximate size of micro-BHF is 1000 nm by 150 nm as indicated. When a magnetic field *H* is experienced, as shown in (b), the dipole moment *m* generates a torque to align itself with the direction of the field. When an electric field *E* is experienced, an electric dipole moment *p_{ind}* is induced in the plane, and subsequently transverse to *m*. Similarly, *p_{ind}* generates a torque to align itself with *E*.

Since p_{ind} is orthogonal to m, electric and magnetic fields can be simultaneously applied to control particle orientations about two orthogonal axes. Herein, this type of manipulation is referred to as orthogonal control. In addition, the fields produced by particles also contribute to the local fields, which effectively produce interactions between particles. The local interactions of particles and the simultaneous application of fields can lead to a hierarchy of structural arrangements. For instance, Figure 3-2 shows two cases of processing fields for the same constituent set: an elastomeric matrix (PDMS) and magnetic particles (BHF). The initial microstructure is assumed to contain randomly dispersed and oriented BHF particles¹. When a magnetic field H_0 is applied, the particles align with the field, forming chains that also propagate in the direction of the field. When an electric field E_0 is applied in addition to H_0 , BHF particles align their easy axes (out of plane) with H_0 , while chains form in the direction of E_0 along a the induced electric dipole axes (an in-plane axis), as shown in Figure 3-2 (M. A. Al Masud, Ounaies, and von Lockette 2018; M. Al Masud et al. 2019).



Figure 3-2. Randomly oriented hard magnetic particles are embedded in a polymer matrix. Applying H_0 while the matrix is a liquid during curing yields chain formations and particle orientations both in the direction of H_0 . Applying H_0 and E_0 simultaneously in transverse directions yields chain formations in the direction of E_0 and particle orientations aligned with H_0 .

To tailor properties of composites, the relationship between starting properties and proportions of the constituents, processing conditions and the resulting filler micro-structure must be understood in terms of the underlying physics. While some studies have experimentally investigated aspects of microstructure control via processing conditions (Bharti and Velev 2015; Kokkinis, Schaffner, and Studart 2015; Faik et al. 2016), none have attempted to provide a thorough understanding of the relationship. Similarly, some studies have simulated microstructure evolution under single-field conditions, such as magnetic or electric fields only, but none have imposed

¹ This assumption is made to simplify the example shown in Figure 3-2. In reality, the initial dispersion of particles may not be uniformly random.

simultaneous fields². Therefore, it is appropriate to formulate a solution—in the context of this work, a numerical simulation—that can help understand how the governing physical principles such as fluid dynamics, dipole-dipole interactions drive the microstructure evolution of micro-composites. The details of the methodologies are given in Chapter 3.

2.2 Dynamic Particle Simulation Methods

The selected processing conditions, electric and/or magnetic fields, influence the arrangements and orientations of the particles, which are mobile during the curing process when the polymer matrix is not completely solid. Thus, the evolution of the microstructure of the composite during such processing conditions will be simulated *via* particle dynamics simulations under ferrohydrodynamic principles, following a procedure similar to a previous model (Rodriguez and von Lockette 2017).

2.2.1 Magnetic force and torque

The aim of this subsection is to provide forms of functions for the magnetic forces and torques acting on any particle, such that, for example, the magnetic force acting on a particle is in the form $F_i^{mag} = F_i^{mag}(H_0, \{M\})$, where H_0 is the external field and $\{M\}$ is the set of all particles' magnetic moments. The derivation begins with a physical definition of F_i^{mag} , which is

$$F_i^{mag} = \nabla U_n, \tag{3-1}$$

where U_n is the energy due to the Zeeman potential at a particle location, defined by

$$U_n = -\mu_0 M V(\boldsymbol{H}_i \cdot \boldsymbol{d}). \tag{3-2}$$

The field H_i is the local magnetic field, which is a summation of the external field and the fields induced by other dipoles, H_j , namely

$$H_i = H_0 + \sum_{j=1}^{N-1} H_j.$$
(3-3)

 $^{^{2}}$ The assertions that these types of studies (i.e. experimental and numerical works on multi-field processing) are absent in the literature are to the best of the author's knowledge.

The field induced by a magnetic dipole, given the form of (2-1), can be substituted into (3-3) to obtain an expression for the total local magnetic field at particle *i*,

$$H_{i} = H_{0} + \sum_{m=1}^{N-1} \frac{1}{4\pi\mu_{0}r_{j}^{3}} \left[3(\boldsymbol{m}_{j} \cdot \hat{\boldsymbol{r}}_{j}) \hat{\boldsymbol{r}}_{j} - \boldsymbol{m}_{j} \right],$$
(3-4)

where m_j is the magnetic dipole moment of particle *j*, and \hat{r}_j is the unit vector of r_j , which is the vector from particle *j* to particle *i*.

The magnetic torque acting on a particle can be found by the cross product,

$$T_i^{mag} = (m_i \times H_i), \tag{3-5}$$

where H_i is the field local to a dipole.

2.2.2 Dielectrophoresis and electric torques

The particles are also assumed to have a dielectric response, which yields dielectric forces and torques. The dielectrophoretic force experienced by particle i is

$$F_i^{DEP} = (p_i \cdot \nabla) E_i, \tag{3-6}$$

where E_i , like H_i , is defined as the sum of all field contributions of other particles and the external field,

$$\boldsymbol{E}_{i} = \boldsymbol{E}_{0} + \sum_{m=1}^{N} \frac{1}{4\pi\varepsilon_{0}r_{j}^{3}} [3(\boldsymbol{p}_{j}\cdot\hat{\boldsymbol{r}}_{j})\hat{\boldsymbol{r}}_{j} - \boldsymbol{p}_{j}].$$
(3-7)

The electric torque T_i^e experienced by particle *i* is defined as

$$T_i^e = p_i \times E_i \tag{3-8}$$

2.2.3 Control Volume and Coordinates

Consider an ensemble of *N* ellipsoidal particles, existing in a volume of space *V*. The space containing the particles serves as the control volume of the representative volume element of the microstructure and is assumed to be a subset of Euclidean space \mathbb{R}^3 . A typical three-dimensional Cartesian coordinate system, $\{x, y, z\}$, is adopted, and an orthonormal basis $\{\hat{i}, \hat{j}, \hat{k}\}$ spanning \mathbb{R}^3 is

chosen. Since the control volume does not deform and is described by an orthonormal basis, the same coordinate system and basis are used for both the initial and current configurations for convenience. The volume, basis, and coordinates are shown in Figure *3-3*.



Figure 3-3. The control volume is shown with N particles, along with a basis and coordinate system in both the reference and current configurations. Particle positions (X, and x) and orientations (D and d) are also shown.

The volume of the space, V, can be calculated based on the volume fraction of particles f, defined by

$$V = \frac{NV_p}{f},\tag{3-9}$$

where V_p is the volume of a single ellipsoidal particle.

Particle positions and orientations in the current configuration are represented by $x_i = x_i(t)$ and $d_i = d_i(t)$, respectively, at any time t. The initial states of particle positions and orientations in the reference configuration, $X_i = x_i(t = 0)$ and $D_i = d_i(t = 0)$, respectively, are assumed uniformly random. Any vector r_j from particle j to particle i can be defined as $r_j = x_j - x_i$ and its magnitude $r_j = ||r_j||$. The motions and rotations of particles are assumed to be only dependent on the following set of physical phenomena: magnetic and dielectrophoretic forces and torques; and linear and angular drag effects of the matrix fluid. The particles are assumed to have

both magnetic and dielectric properties, and as a result, their behaviors are driven by the interactions induced by an external magnetic field H_0 and electric field E_0 .

2.2.4 Equations for Case of BHF Particles

In Sections 2.2.1 and 2.2.2, the forces and torques experienced by particles are described as generally as possible without providing explicit forms of the dipole moments. Additionally, many of the equations in Sections 2.2.1 and 2.2.2 are written with minimal reliance on coordinates and basis vectors to emphasize the invariance of the forces and torques. In this section, the force and torque equations are restricted to a set of assumptions made for BHF particles.

Ellipsoids are chosen for particle geometry because the analytical forms of their electric and magnetic interactions have been solved (Jones 1995), and can be used as reasonable approximations of hexagonal platelets with preserved volume and aspect ratios (see Appendix F for details on supporting study *via* finite element analysis). The ellipsoid dimensions are defined by the principal semi-axis lengths *a*, *b*, *c* as shown in Figure 3-4 relative to the local basis. The shape of the ellipsoids can be restricted to a class of ellipsoids called oblate spheroids, defined by the geometric relationships $c \ll a = b$.



Figure 3-4. An ellipsoidal particle's dimensions and local bases are shown.

The particles are assumed to have ideal hard magnetic behavior, with a constant, average permanent dipole moment of magnitude *m* across all particles. Due to the constant, ideal dipole behavior, particles are assumed to not experience fields surpassing their coercive field in any given orientation. The orientation of the magnetic dipole moment of particle *i* is given by $m_i = m\hat{e}_i^3$, where $\{\hat{e}_i^1, \hat{e}_i^2, \hat{e}_i^3\}$ is a basis local to the particle and relative to the global basis as shown in Figure 3-4, and spans \mathbb{R}^3 . The magnetic moment *m* can be determined from the magnetization, or magnetic dipole density, *M*, of the composite by the relation $m = MV_p$, where V_p is the volume of a particle. While there are also induced dipole moments due to the anisotropic shapes of the particles, their effects are neglected under the assumption that the hard-magnetic response dominate the magnetic force of each particle.

With the substitution of $m = MV_p$, the potential of a particle experiencing field H_i is

$$U_i = -MV \left(\boldsymbol{H}_i \cdot \hat{\boldsymbol{e}}_i^3 \right), \tag{3-10}$$

and the field H_i is

$$\boldsymbol{H}_{i} = \boldsymbol{H}_{0} + \sum_{j=1}^{N-1} \frac{M V_{p}}{4\pi\mu_{0} r_{j}^{3}} \left[3 \left(\hat{\boldsymbol{e}}_{j}^{3} \cdot \hat{\boldsymbol{r}}_{j} \right) \hat{\boldsymbol{r}}_{j} - \hat{\boldsymbol{e}}_{j}^{3} \right].$$
(3-11)

Substituting (3-10) and (3-11) into (3-1) yields

$$F_{i}^{mag} = \sum_{j=1}^{N-1} \frac{(MV_{p})^{2}}{4\pi\mu_{0}r_{j}^{4}} [\hat{r}_{j}(\hat{e}_{j}^{3}\cdot\hat{e}_{i}^{3}) - 5\hat{r}_{j}(\hat{e}_{j}^{3}\cdot\hat{r}_{j})(\hat{e}_{i}^{3}\cdot\hat{r}_{j}) + \hat{e}_{i}^{3}(\hat{e}_{i}^{3}\cdot\hat{r}_{j}) + \hat{e}_{j}^{3}(\hat{e}_{i}^{3}\cdot\hat{r}_{j})] + \hat{e}_{j}^{3}(\hat{e}_{i}^{3}\cdot\hat{r}_{j})].$$
(3-12)

The magnetic torque can be described by

$$\boldsymbol{T}_{i}^{mag} = -\mu_{0} M V (\hat{\boldsymbol{e}}_{i}^{3} \times \boldsymbol{H}_{i}), \qquad (3-13)$$

which can be expanded with the substitution of (3-11),

$$T_{i}^{mag} = -\mu_{0}MV \left(\left(\hat{e}_{i}^{3} \times H_{0} \right) + \sum_{j=1}^{N-1} \frac{MV_{p}}{4\pi\mu_{0}r_{j}^{3}} \left[3\left(\hat{e}_{j}^{3} \cdot \hat{r}_{j} \right) \left(\hat{e}_{i}^{3} \times \hat{r}_{j} \right) - \left(\hat{e}_{i}^{3} \times \hat{e}_{j}^{3} \right) \right] \right).$$

$$(3-14)$$

While it may not be clear in the derivations (3-9)–(3-13), $\hat{r}_j = \hat{r}_j (x_i(t), x_j(t))$, and $\hat{e}_i^3 = \hat{e}_i^3 (x_i(t), x_j(t))$.

The electric analogs of (3-12) and (3-13) can be derived similarly. The biggest difference is that BHF particles do not exhibit a permanent electric dipole. Instead, the external field induces an effective electric dipole moment p_i for a particle *i*, which can be expressed by

$$\boldsymbol{p}_{\boldsymbol{i}} = V_p \left(\varepsilon_p - \varepsilon_m \right) \left(\frac{E_0 \cdot \hat{\boldsymbol{e}}_i^1}{1 + \left(\frac{\varepsilon_p - \varepsilon_m}{\varepsilon_m}\right) L_1} \hat{\boldsymbol{e}}_{\boldsymbol{i}}^1 + \frac{E_0 \cdot \hat{\boldsymbol{e}}_i^2}{1 + \left(\frac{\varepsilon_p - \varepsilon_m}{\varepsilon_m}\right) L_2} \hat{\boldsymbol{e}}_{\boldsymbol{i}}^2 + \frac{E_0 \cdot \hat{\boldsymbol{e}}_i^3}{1 + \left(\frac{\varepsilon_p - \varepsilon_m}{\varepsilon_m}\right) L_3} \hat{\boldsymbol{e}}_{\boldsymbol{i}}^3 \right).$$
(3-15)

The form of (3-15) is borrowed from literature (Jones 1995), and it is dependent on depolarization factors L_i , the electric permittivities of the medium and particles, ε_m and ε_p , respectively, and the components of the external field E_0 with respect to orthonormal bases $\{\hat{e}_i^1, \hat{e}_i^2, \hat{e}_i^3\}$ local to the particle (see Figure 3-4). Within the form of (3-15), the following assumptions are made: the particles are lossless (in electric conductivity), dielectric, isotropic (in polarizability), and homogeneous such that they experience uniform polarization. The depolarization factors L_i also correspond to the directions of \hat{e}_i^k (for k = 1,2,3).

With a change of bases, p_i can be written as a function of the global coordinates³ {x, y, z} based on the orthonormal basis vectors $\hat{i}, \hat{j}, \hat{k}$.

$$p_{ix} = \boldsymbol{p}_i \cdot \boldsymbol{\hat{i}},\tag{3-16}$$

$$p_{iy} = \boldsymbol{p}_i \cdot \hat{\boldsymbol{j}},\tag{3-17}$$

³ Since the control volume does not deform, the formulation can be simplified by using a single set of coordinates for both the reference and current configurations.

$$p_{iz} = \boldsymbol{p}_i \cdot \hat{\boldsymbol{k}}. \tag{3-18}$$

The forms of L_k (for k = 1,2,3) are dependent on the shape of the polarizable body. In this case, ellipsoidal depolarization factors are used, assuming the form

$$L_k = \frac{abc}{2} \int_0^\infty \frac{ds}{(s+i^2)R_s},$$
 (for $k = 1,2,3$) (3-19)

$$R_s = \sqrt{(s+a^2)(s+b^2)(s+c^2)}.$$
(3-20)

A simpler form of (3-19) for oblate spheroids is

$$L_1 = L_2 = \frac{c}{2a} \left[\frac{\pi}{2} - \frac{c}{a} \right], \tag{3-21}$$

and by substituting the definition, $L_1 + L_2 + L_3 = 1$, L_3 is obtained,

$$L_3 = 1 - \frac{c}{a} \left[\frac{\pi}{2} - \frac{c}{a} \right].$$
(3-22)

The next derivation is for expressing (3-15) in terms of the global basis. Let particles' orientation \hat{e}_i^3 be described by spherical coordinates $\{1, \theta, \phi\}^4$, related to $\{x, y, z\}$ through

$$x = \sin\theta\cos\phi,\tag{3-23}$$

$$y = \sin\theta \sin\phi, \tag{3-24}$$

$$z = \cos\phi, \tag{3-25}$$

such that $\hat{e}_i^3 = \sin\theta\cos\phi\hat{i} + \sin\theta\sin\phi\hat{j} + \cos\phi\hat{k}$.

Next, (3-16)–(3-25) are substituted into (3-15) for the following form of the induced electric moment's z-component,

$$(p_i)_z = V_p \left(\varepsilon_p - \varepsilon_m\right) \left(\frac{E_{0x} \sin\theta \cos\phi + E_{0y} \sin\theta \sin\phi + E_{0z} \cos\phi}{1 + \left(\frac{\varepsilon_p - \varepsilon_m}{\varepsilon_m}\right) \left(1 - \frac{c}{a} \left[\frac{\pi}{2} - \frac{c}{a}\right]\right)}\right),\tag{3-26}$$

where E_{0i} are components of E_0 in the x, y, and z-directions. Substituting (3-26) into (3-6) gives a form of the component of F_i^{DEP} on particle *i* in the z-direction,

⁴ Radial coordinate, typically represented by *r*, is 1 since \hat{e}_i^3 is a unit vector.

$$(F_{i}^{DEP})_{z} = V_{p} (\varepsilon_{p} - \varepsilon_{m}) \left(\frac{E_{0x} sin\theta cos\phi + E_{0y} sin\theta sin\phi + E_{0z} cos\phi}{1 + \left(\frac{\varepsilon_{p} - \varepsilon_{m}}{\varepsilon_{m}}\right) \left(1 - \frac{c}{a} \left[\frac{\pi}{2} - \frac{c}{a}\right]\right)} \right) \sum_{j=1}^{N-1} \frac{1}{4\pi\varepsilon_{0} r_{j}^{4}} \left[(\hat{r}_{j} \cdot \hat{k}) (\hat{e}_{j}^{3} \cdot \hat{r}_{j}) (\hat{e}_{i}^{3} \cdot \hat{r}_{j}) + (\hat{e}_{i}^{3} \cdot \hat{k}) (\hat{e}_{i}^{3} \cdot \hat{r}_{j}) + (\hat{e}_{j}^{3} \cdot \hat{k}) (\hat{e}_{i}^{3} \cdot \hat{r}_{j}) + (\hat{e}_{j}^{3} \cdot \hat{k}) (\hat{e}_{i}^{3} \cdot \hat{r}_{j}) \right]$$

$$(3-27)$$

$$(\hat{r}_{j}) \left[\frac{1}{2} \left[\frac{1}{2} - \frac{c}{a} \right] + \frac{1}{2} \left[\frac{1}{2} - \frac{c}{a} \right] + \frac{1}{2} \left[\frac{1}{2} - \frac{c}{a} \right] \right] \right]$$

Since the particles are assumed to be rigid bodies, \hat{e}_i^1 and \hat{e}_i^2 rotate by the same angles as \hat{e}_i^3 , meaning the remaining components of p_i can be derived similarly with the use of Rodrigues' rotation method (Palais and Palais 2007), which can be substituted into (3-6) to find the remaining components of F_i^{DEP} as done with the z-component in (3-27).

The total field E_i at any point x_i (occupied by particle *i*) can be described by

$$\boldsymbol{E}_{i} = \boldsymbol{E}_{0} + \sum_{j=1}^{N-1} \frac{p_{i}}{r_{ij}^{3} \varepsilon_{0}} \left[3 \left(\hat{\boldsymbol{e}}_{j}^{3} \cdot \hat{\boldsymbol{r}}_{j} \right) \hat{\boldsymbol{r}}_{j} - \hat{\boldsymbol{e}}_{j}^{3} \right],$$
(3-28)

where $p_i = |p_i|$.

Note that while induced magnetic and permanent electric dipoles are not accounted for, these missing terms can be derived from their magnetic or electric counterparts through analogy. A permanent electric dipole moment p_i^{per} with constant magnitude p_i^{per} across all particles, assumed to behave ideally, can be added as $p_i^{per} = p^{per}d_i$, and an induced magnetic dipole moment m_i^{ind} can be added in a similar form as (3-15). The ability to write these additional terms allows for a generality in the formulation such that any arbitrary combination of electric and magnetic properties of particles can be modeled, assuming spheroidal particles with homogeneous, lossless dielectric/magnetic properties.

2.2.5 Hydrodynamic Forces and Torques

In addition to the magnetic and DEP forces, there are also hydrodynamic forces and torques as a result of the fluid medium in which the particles are suspended. The drag force on particle i is

$$F_i^{drag} = -D_s \dot{x}_i, \tag{3-29}$$
where D_s is the drag coeffcient, which is assumed $D_s = 6\pi\eta R_{eq}$, with R_{eq} being the radius of a sphere of equivalent volume, and η is the dynamic viscosity (Stokes 1901). The fluid also exerts a torque against the particles as they try to rotate, which is given by

$$T_i^{drag} = C_s \boldsymbol{\omega}_i, \tag{3-30}$$

where ω_i is the angular velocity of a particle and $C_s = 8\pi\eta R_{eq}^3$ (Guazzelli and Morris 2011). Again, it should be noted that the drag and repulsive terms are assumed spherical particles due to computational costs and the ease of implementation.

Besides fluid forces and torques, a repulsive force proportional to the magnetic and electric forces is added to prevent overlapping of particles. The repulsive force is an exponential function used in hard sphere contact problems, and for the dynamic simulations in this study, takes the form of

$$F_{i}^{rep} = \sum_{j=1}^{N} \left(e^{-\beta \left(\frac{r_{m}}{2R} - 1\right)} \right) \left(\frac{\mu_{0}(MV_{p})^{2} H_{0}^{2}}{4\pi} \right) \hat{r}_{j},$$
(3-31)

in which, *R* is an equivalent radius of the particles, and β is a constant. Since a hard sphere repulsive term is used, the ellipsoidal particles are expected to not make contact when their magnetic dipole moments align (i.e. in the direction of their shortest axis), and possibly overlap when they are aligned along the in-plane direction. A repulsive force term for ellipsoids is not yet included in the formulation due to a much greater computational cost of implementing an algorithm that considers orientations of all neighboring particles to adjust the repulsive term in every calculation. Likewise, a more complex algorithm is necessary for implementing ellipsoidal forms of the drag terms.

2.2.6 Equations of Motion

The sum of forces and moments for each particle are given by

$$m\boldsymbol{a}_{i} = \boldsymbol{F}_{i}^{mag} + \boldsymbol{F}_{i}^{DEP} + \boldsymbol{F}_{i}^{rep} + \boldsymbol{F}_{i}^{drag}, \qquad (3-32)$$

$$J(\boldsymbol{d}_{i} \times \boldsymbol{\alpha}_{i}) = \boldsymbol{T}_{i}^{mag} + \boldsymbol{T}_{i}^{e} + \boldsymbol{T}_{i}^{drag}, \qquad (3-33)$$

where a_i is the linear acceleration and α_i is the angular acceleration of particle *i*. The equations can also be written in terms of state variables x_i and d_i by adopting the following relations: $v_i = \dot{x}_i$ (velocity), $a_i = \ddot{x}_i$ (acceleration), $\omega_i = \dot{d}_i$ (angular velocity), and $\alpha_i = \ddot{x}_i$ (angular acceleration). Thus, the forces and torques are summed by

$$m\ddot{x}_i = F_i^{mag} + F_i^{DEP} + F_i^{rep} + F_i^{drag}, \qquad (3-34)$$

$$J(\boldsymbol{d}_i \times \boldsymbol{\ddot{d}}_i) = \boldsymbol{T}_i^{mag} + \boldsymbol{T}_i^e + \boldsymbol{T}_i^{drag}.$$
(3-35)

With some algebraic manipulation, and substitution of (3-29) and (3-30),

$$\ddot{x}_i = \frac{1}{m} \left(F_i^{mag} + F_i^{DEP} + F_i^{rep} \right) - \frac{D_s}{m} \dot{x}_i,$$
(3-36)

$$\ddot{d}_i = \frac{1}{J} (\boldsymbol{m}_i \times \boldsymbol{H}_i + \boldsymbol{p}_i \times \boldsymbol{E}_i) - \frac{c_s}{J} \dot{d}_i.$$
(3-37)

2.2.7 Temporal Discretization

Since the system of equations presented in (3-36) and (3-37) are difficult to solve in their current forms, a few assumptions can be made to simplify them to approximate their solutions. For instance, assume that the system is quasi-equilibrium, $\ddot{x}_i \approx 0$ and $\ddot{d}_i \approx 0$, which yields

$$\dot{x}_{i} = \frac{1}{D_{s}} \left(F_{i}^{mag} + F_{i}^{DEP} + F_{i}^{rep} \right),$$
(3-38)

$$\dot{d}_i = \frac{1}{c_s} [(\boldsymbol{m}_i \times \boldsymbol{H}_i) + (\boldsymbol{p}_i \times \boldsymbol{E}_i)].$$
(3-39)

The time domain can be discretized such that the equations can be calculated at $t = t_0 + h$ for any initial time t_0 and small time interval h. This assumption allows for the substitution of the linear relations $\dot{x}_i = (x_i - x_{0i})/h$ and $\dot{d}_i = (d_i - d_{0i})/h$, where $x_{0i} = x_i(t_0)$ and $d_{0i} = d_i(t_0)$, yielding

$$x_{i}(t_{0}+h) = x_{0i} + \frac{h}{D_{s}} \left(F_{i}^{mag} + F_{i}^{DEP} + F_{i}^{rep} \right),$$
(3-40)

$$\boldsymbol{d}_{i}(t_{0}+h) = \boldsymbol{d}_{0i} + \frac{h}{c_{s}} \big((\boldsymbol{m}_{i} \times \boldsymbol{H}_{i}) + (\boldsymbol{p}_{i} \times \boldsymbol{E}_{i}) \big).$$
(3-41)

The assumption that the linear and angular accelerations are zero implies that particles are moving slow enough that the inertial effects of changes in speed are neglected.

2.2.8 Boundary Conditions

A nearest-neighbor periodic distribution is assumed as the boundary condition, as shown in Figure 3-5. This boundary condition plays two significant roles in the simulations: (1) it maintains a constant number of particles in the control volume and (2) reduces edge effects of the control volume by introducing particle field effects from neighboring regions.



Figure 3-5. Nearest neighbor periodic boundary condition presented in 2D. Control volume is highlighted in bolded rectangular outline in the center of the grid. Particles leaving the CV are assumed to enter a neighboring region as shown and enter the CV from the opposite edge due to the periodic boundary condition.

2.3 Multi-Field Processing Results and Conclusions

This section utilizes the modeling techniques developed earlier to simulate microstructure evolution under different processing conditions based on combinations of electric and magnetic fields. Furthermore, the simulation results are compared with experimental results obtained by colleague M. Abdullah Masud at Penn State to validate model assumptions and verify Hypothesis 1. As such, the simulations aid in advancing toward a universal printer by offering a method to predict formation of micro-architectures based on multiple processing conditions. The join work with experiments have been published in the SPIE conference proceeding (REF). A more generalized modeling formulation and its detailed results are in preparation for Journal submission.

The of all the simulations set parameters used in is $\{N, M, \varepsilon_m^*, \varepsilon_p^*, f, H_0, E_0, \eta, a, b, c, R_{eq}, \beta\}$. Some parameters, such as H_0, η, β , and R_{eq} are chosen based on a previous study on a single-field simulation of BHF particles in a fluid medium (Rodriguez and von Lockette 2017). The external field H_0 is chosen relative to M such that $H_0 =$ 5*M*, and M = 380 kA/m. The dynamic viscosity is $\eta = 3.5 Pa * s$, and the equivalent radius is $R_{eq} = 0.5 \ \mu m$. The external field E_0 is chosen to be 100 MV/m. The volume fraction f is 1%, which matches the experimental value (M. A. Al Masud, Ounaies, and von Lockette 2018). The relative electric permittivities of the elastomeric matrix and the particles are $\varepsilon_m^* = 2$, and $\varepsilon_p^* = 10$, respectively, which also matches theoretical values reporting a ratio of $\varepsilon_p^*/\varepsilon_m^* = 5$. The particle shapes' aspect ratios are known to be approximately 7, based on experimental imaging methods, for the hexagonal plates (M. Al Masud et al. 2019). The aspect ratio of a hexagonal plate is measured by the side length of the hexagon divided by the thickness of the plate. As a result, the ellipsoidal dimensions have a relation of c = a/3.5 (b = a). The initial positions and orientations of particles are assumed uniformly random (generated via MATLAB's random number generator). An example of an initial starting point of the microstructure for N = 100 particles is shown in Figure 3-6. The magnetic dipole moments of each particle are represented by the blue arrows. Electric dipole moments are not shown since they depend on the direction of the applied field, whereas magnetic dipole moments are fixed to the local axes on each particle.



Figure 3-6. Visual representation of starting positions of BHF particles in the control volume. Arrows indicate directions of magnetic dipole moments of each particle.

To analyze simulation results, three types of aggregate structures are defined: stacks, microchains, and macro-chains. Stacks are a short grouping of particles aligned in the same direction, yielding a net magnetization direction as shown in Figure *3-7*.a. Micro-chains are groups of stacks that may or may not be ordered (shown randomly oriented in the micro-chain example in Figure *3-7*.b). Macro-chains are strings of micro-chains stretching across the material, as shown in Figure *3-7*.c. These definitions are partly based on experimental results found in a join study (M. Al Masud et al. 2019).



Figure 3-7. Three possible levels of structure with the base unit of a BHF particle, represented by the ellipses. Net magnetization of stacks are indicated by the red arrows. The smallest structure is (a) a stack. The medium structure is (b) a micro-chain consisting of multiple stacks. The largest structure is (c) a macro-chain consisting of micro-chains.

2.3.1 Five Cases of Processing Conditions

Prior to comparing simulation results to experiments, five cases are simulated to test the simulation results versus expectations. The cases represent basic conditions utilizing a few of the different ways interactions between magnetic dipoles and electric dipoles can align particles and form stacks, micro-chains, and macro-chains. The expected outcomes of the cases are shown in Figure 3-8. Case 1 is the no-field case, in which there are no external fields present. Particles in Case 1, represented by the blue ellipses in Figure 3-8, are expected to form local micro-chains of stacks, where some may contain randomly aligned stacks while others may contain macro-chains forming loops minimizing the local energy. However, macro-chains covering relatively significant portions of the control volume are not expected in Case 1 since the system is not receiving additional energy that could decrease the entropy of the system. In Case 2, particles are subjected to an electric field in the z-direction, $E_0 = E_0 \hat{k}$. The electric field is expected to induce an electric dipole moment in the z-direction for Case 2, such that micro-chains align with the field with stacks having no order as shown in Figure 3-8.b. In Case 3, particles are subjected to a magnetic field in the z-direction, $H_0 = H_0 \hat{k}$. The magnetic dipoles in Case 3 are expected to form stacks that align with the direction of the field to reduce the Zeeman energy, and the resulting interactions should form macro-chains also in the z-direction. In Case 4, particles are subjected to both fields in transverse configuration such that $H_0 = H_0 \hat{k}$ and $E_0 = E_0 \hat{i}$. The particles stacks' magnetic moments are expected to align with H_0 but form macro-chains in the direction of E_0 . In Case 5, the particles are subjected to both fields in parallel such that $H_0 = H_0 \hat{k}$ and $E_0 = E_0 \hat{i}$. The stacks are expected to align and form macro-chains in the direction of the fields, as shown in Figure 3-8.e. Similar to the stack, microchains, and macro-chain definitions, the expectations of these cases are partly based on prior experimental results (M. A. Al Masud, Ounaies, and von Lockette 2018; M. Al Masud et al. 2019).



Figure 3-8. The five cases are shown with simplified 2D schematics illustrating particles represented by ellipsoids and chains indicated by outlines of connected particles.

The differences between some of the cases indicate a hierarchy of structure across length scales. For instance, the differences between Cases 2, 4, and 5 are the internal structuring of stacks within chains. In Case 5, both stacks and chains are aligned in the same direction, which could yield bulk properties such as magnetic and elastic responses that are anisotropic in the same direction. Meanwhile, Case 4 also contains chains, but stack orientations are transverse to chain directions. A Case 4 microstructure may yield magnetic and elastic bulk properties that are transverse to each other, which is different from Case 5. Case 2, by contrast, does not have any order at the stack level, which would produce isotropic magnetic property. This kind of ability to manipulate hierarchical organization in a microstructure can allow for means of optimizing properties such as magnetic torque in a material used for actuator applications. Optimization of torque via hierarchical organization could lead to smaller and fewer actuators, which could reduce costs and increase the efficiency of the devices or systems utilizing the actuators

Similarly, the difference between Cases 3 and 5 are subtle but yield equally different bulk properties. Case 5 is expected to form chains longer than those in Case 3 due to the combined effects of both fields in the same direction, which produce coinciding electric and magnetic dipole-dipole interactions. Even though both cases have chains and stacks aligned in the same directions, longer chains (increasing probability of percolation) in one case over another could be the difference needed for high electrical conductivity versus almost no electrical conductivity, assuming particles exhibit high electrical conductivity and the medium is nearly an ideal electrical insulator.

The simulation results for each case are shown in Figure 3-9 through Figure 3-13. For each case, the simulations are run for at least 0.5 seconds of simulation time, which is adequate for convergence based on a past study that measured a convergence metric versus simulation time (Rodriguez and von Lockette 2017). The results of the cases are shown in Figure 3-13. Simulation results for cCse 1 show chains and stacks of particles of various lengths with no visible pattern in terms of chain orientations (i.e. chain orientations are random). These simulation results match with expected results shown in Figure 3-8 for Case 1, with the notable exception of a lack of a third level

of hierarchy in the simulations, which are micro-aggregates. That is, the simulations contain chains consisting of stacks of particles. This discrepancy may be a product of the various assumptions buried in the methodologies, primarily a lack of friction between particles, which could yield the development of aggregates composed of smaller stacks. Nonetheless, the particle positions and orientations from simulation results for Case 1 in Figure *3-9* do confirm the expected "random" orientations of chains with some ordering at the stack level.



Figure 3-9. Simulation results for Case 1, the no-field case. Particles are represented by ellipsoids, with magnetic dipole moments signaled by the blue arrows.

Simulation results for Case 2 are shown in Figure 3-10. The particles are oriented such that their long axes are aligned with E_0 while forming chains along the same direction. Induced electric dipole moments are indicated by red arrows, all aligned with E_0 . However, on the y-z plane, particle orientations appear randomly distributed, as seen by the blue arrows indicating magnetic moments. Again, micro-aggregates do not form in simulations, but the particles can also be treated as aggregates whose geometric anisotropy induces electric dipole moments aligning them with the field.



Figure 3-10. Simulation results for Case 2, the electric field only case. Particles are represented by ellipsoids, with magnetic dipole moments signaled by the blue arrows.
Simulation results for Case 3 are shown in *Figure 3-11*. The particles form stacks in the direction of *H*₀ some large enough to be classified as chains. There is a distribution of stack and chain lengths, which are likely due to the random nature of the initial starting positions.
Nonetheless, these simulation results agree with expected results shown in Figure 3-8.



Figure 3-11. Simulation results for Case 3, the magnetic field only case. Particles are represented by ellipsoids, with magnetic dipole moments signaled by the blue arrows.

Simulation results for Case 4 are shown in Figure 3-12. The particles form stacks in the direction of H_0 , which coincides with \hat{k} , and a combination for particles and stacks form macrochains in the direction E_0 , which coincides with \hat{i} . There is a distribution of stack sizes within the chains, but stay within the range of two to three particles per stack, which is likely due to the relatively small control volume of the simulations (i.e. small population size of 100). Nonetheless, these simulation results agree with expected results shown in Figure 3-8, as they show how two levels of hierarchy, micro-scale stacks in one direction and macro-chains in another direction, can be produced and controlled *via* the application of electric and magnetic fields.



Figure 3-12. Simulation results for Case 4, the electric and magnetic fields in transverse case. Particles are represented by ellipsoids, with magnetic dipole moments signaled by the blue arrows and electric dipole moments by red arrows.

Simulation results for Case 5 are shown in Figure *3-12*. Stacks form in the direction of the fields, and comprise macro-chains in the same direction. Besides the distinct difference from Case 4, i.e., stack-level alignments, Case 5 also differs from Case 3 with much longer chains. These results agree with expected results shown in Figure *3-8*.



Figure 3-13. Simulation results for Case 5, the electric and magnetic fields in parallel case. Particles are represented by ellipsoids, with magnetic dipole moments signaled by the blue arrows and electric dipole moments by red arrows.

The simulations are compared with experiments from a joint study (M. Al Masud et al. 2019). The locations and orientations of particles from a set of simulation results having N = 200 are recorded and plotted as ellipsoids in *Figure 3-14*.a for the case with only magnetic field (Case 3) and *Figure 3-14*.b for both fields in parallel (Case 5). Observations of the organizations of particles in Figure *3-14* indicate that all particles are aligned with the externally applied fields, and each case has significant chain formation. The case with both the magnetic and electric fields appears to have longer chain formation, but the exact differences are hard to distinguish by visual examination. Thus, the next section develops a basic quantitative metric that could aid in comparing simulation and experimental results, as well as learning more about the differences in structural hierarchy between the two cases.



Figure 3-14. The simulation results for N = 200 particles are shown under (a) magnetic field only, and (b) electric and magnetic fields. Each ellipsoid represents a BHF particle, and the arrows indicate their dipole moment orientations (blue is magnetic and red is electric). Scale of center images is 100 microns, with particles of 1 micron in length. Image is from (*M. Al Masud et al.* 2019).

2.3.1 Distance-Based Minimum Spanning Tree Analysis

The simulation results are further analyzed by finding the minimum spanning forest of each graph. The graphs consist of edges between the particles, weighted by the distances between them. A minimum spanning tree is subset of all possible edges such that the chosen edges connect all vertices while minimizing the total edge weight without any circles forming. The final positions and orientations of particles from simulations are used to create a connectivity matrix with a maximum allowed distance d_{tree} . Based on the limit set by d_{tree} , MATLAB's minspantree algorithm is utilized to find minimum spanning trees. The set of minimum spanning trees is referred to as the minimum spanning forest, where each tree is separated by at least a distance of d_{tree} . Thus, varying d_{tree} will change the number of trees, and consequently the number of particles per tree. In terms of the material microstructure, this could mean that a minimum spanning tree could represent a number of structures depending on the scale of d_{tree} . At a low scale of d_{tree} , stacks can be obtained; at a medium scale, aggregates can be obtained; and at a large scale, chains can be obtained. These scales are relative to the dimensions of the particles. Thus, the minimum spanning forest can help objectively identify these structures from a microstructure, whether simulated or experimentally acquired, such that other quantitative analyses can be conducted (e.g. comparing distribution of

orientations). Furthermore, if there are some ranges for which the number of trees does not change, then there may be a prominent level of structural hierarchy in that scale.

Figure 3-15 shows the number of trees versus d_{tree}/R_{eq} , where R_{eq} is the equivalent spherical radius of a particle, for the microstructure simulated using both fields parallel (Case 5). At $\frac{d_{tree}}{R_{eq}} < 2$, spanning trees cannot form since the range is less than the size of a particle. Thus, the total number of trees for $\frac{d_{tree}}{R_{eq}} < 2$ is equivalent to the number of particles. However, at sufficiently large ranges where trees can form, the microstructure analysis reveals plateaus, such as at 2 < $\frac{d_{tree}}{R_{eq}} < 6$. Based on the images of the spanning trees associated with this region, highlighted in red in Figure 3-15, this range is associated with stacks. The next range, at approximately 7 $< \frac{d_{tree}}{R_{eq}} <$ 10, small chain formations exist, and at the range 10 $< \frac{d_{tree}}{R_{eq}} <$ 14, large chains exist which represents the micro-chains similar to experiment.



Figure 3-15. The number of trees is plotted versus the range d_{tree}/R_{eq} used in calculating the minimum spanning trees for Case 5. The connections within the trees are shown inside the boxes, color coded with the plateau ranges.

Additionally, for the same range (stack-level), there are an average of 22 trees for Case 3 (H_0 only), and 17.5 for Case 5 (H_0 and E_0 parallel), which is 20% less. The averages are calculated across three independent simulations per case. Fewer number of trees for the same N is an indication of larger chains forming in Case 5. The longer chain formation may be explained by the effects of combined dipole-dipole interactions due to both electric and magnetic dipoles. When particles are oriented as shown in the simulation microstructure in Figure 3-13, their induced electric dipole moments are almost exactly in the same direction as the field, and thus, they exhibit very weak or no electric torque. Although a small perturbation can create an electric torque and destabilize the particle orientations, their equilibria are maintained by the relatively larger magnetic torques generated by H_0 . Thus, in this orientation, the electric and magnetic fields induced by both electric and magnetic dipoles coincide, and as a result, so do the magnetic and DEP forces that promote chain formation. Consequently, Case 5 develops longer chains since electric and magnetic fields are applied in the same direction.

2.3.2 Dispersions of Particle, Stack, and Chain Orientations for Distance-Based Spanning Trees

For a quantitative analysis of the relative spatial alignments of particles, along with the orientation of each individual particle, the average angle (ϕ) of the connections inside each minimum spanning tree is calculated for two levels of hierarchy: stacks and chains. A distribution of angles for each level of hierarchy is obtained by assessing the average angle of the edges within each tree, and a von Mises distribution function is fitted to each distribution from the simulation and experimental results. The form of the distribution is

$$f_{\rm VM}(\phi|\mu,b) = \frac{e^{b\cos(\phi-\mu)}}{2\pi I_0(b)},$$
(3-42)

where μ is the mean of the distribution, and *b* is a measure of the spread. The average angles are visually defined in Figure 3-16. The experimental results are borrowed from a related published work (M. Al Masud et al. 2019). The von Mises distribution function can also be analyzed by the

dispersion factor, κ , a parameter ranging from 0 (singular point) to 1/3 (uniform distribution) at a mean of μ , unlike *b* which ranges from 0 to infinity. The form of κ is shown later in (4-109) along with a detailed explanation. A smaller κ indicates more alignment. The dispersion factors of the fitted von Mises distributions are listed in Table 3-1. At the particle level, both for simulation ($\kappa =$ 0) and experimental results ($\kappa = 0.093$), dispersions of particle orientations are low, indicating high alignment of the particles with the field directions for both cases. At the stack level, experimental results ($\kappa = 0.26$) show high dispersion compared to simulation results ($\kappa = 0.024$). Microstructures from SEM results showed agglomeration of stacks in few instances which may be the reason for large stack dispersion from experimental results. At the chain level, both cases for experiments ($\kappa = 0.23$) and simulations ($\kappa = 0.25$) show large dispersions close to uniform distributions. This is because with the increase of d_{tree} , side branching occurs perpendicular to the applied field.



Figure 3-16. Angle of a particle and the angles in the edges within a tree are defined as shown.

From Table 3-1, comparison of experimental dispersion factors between the two cases shows that the dispersion factor of particles ($\kappa_{H+E} = 0.079, \kappa_H = 0.093$), stacks ($\kappa_{H+E} = 0.25, \kappa_H = 0.26$) and chains ($\kappa_{H+E} = 0.22, \kappa_H = 0.23$) is slightly lower for the electric and magnetic field processed (Case 5) composites compared to just magnetic field processed (Case 3) composites. This suggests slightly better orientation of BHF at all scales for Case 5 composites. This experimental result validates the simulations, which had predicted that longer chains would form under simultaneously applied fields in the same direction due to coinciding electric and magnetic dipoles, which is unique to that specific case. However, simulation results show that the dispersion factors of BHF stacks ($\kappa_{H+E} = 0.042, \kappa_H = 0.024$) and chains ($\kappa_{H+E} = 0.30, \kappa_H =$ 0.25) are slightly higher for Case 5 composites compared to Case 3 composites. The slight difference in the dispersion factors at the stack level may be an artifact of the methods used to determine the angles of a chain, since stacks in the simulations are already well aligned with respect to the field directions. When calculating the angles of a chain, the angles across stacks are included, which will increase the dispersion factor at the chain level for Case 5 due to longer chains running parallel to each other (i.e. some connections may appear to be side branches of trees), while in Case 3 there are shorter chains or stacks, which can connect more in the direction of the fields than transversely.

	HONLY		H AND E	
	$\operatorname{Exp}\left(\kappa\right)$	$\operatorname{Sim}(\kappa)$	Exp (<i>κ</i>)	$Sim(\kappa)$
PARTICLE (Φ_P)	0.093	0	0.079	0
STACKS (Φ_S)	0.26	0.024	0.25	0.042
CHAINS (Φ_{CH})	0.23	0.25	0.22	0.30

Table 3-1. Comparative degree of orientation of BHF stack and micro-chains. Dispersion factors, (κ) are provided for the stack range, small chain range, and large chain range for each case.

One of the challenges of the spanning tree analysis of the microstructure is that it starts to result in high dispersion at large range when the trees side-branch (e.g. two long chains parallel to each other can only be connected by an edge relatively perpendicular to the field direction). While dispersion factors from spanning tree analysis offer insights into the microstructural hierarchy as a direct result of processing conditions, there are other metrics that can be studied to understand different aspects of the microstructure besides spatial and particle alignments. For example, the study does not consider the moments of inertia of stacks and chains, the average distances between stacks, and the number of stacks within chains, all of which provide extra layers of information regarding the structural properties. It should also be noted that the choice of d_{Tree} strongly influences spanning tree results, and the fact that distance is chosen as the weighting function for the minimum spanning tree. In addition, the simulations can better match experimental conditions by considering contact mechanics, friction between particles, and the limitations of dipole field equations.

2.3.3 Dipole-Dipole Energy-Based Minimum Spanning Tree Analysis

So far, the definitions of stacks, micro-chains, and macro-chains have not been properly quantified, and instead, the comparisons relied on qualitative aspects of each structure. In this section, the goal is to quantitatively distinguish groups of particles as either stacks, micro-chains or macro-chains based on a set of metrics. To begin developing such metrics, the structures for each case are defined by the descriptions shown in Table 2-2. Based on these descriptions, it is notable that the same definition for a structure (e.g. stacks) may not be applicable to all cases. For example, the stacks in Case 2 (electric field only) are based on the anti-parallel alignments of magnetic dipole moments perpendicular to the electric field direction. This definition for stacks is exclusive to case 2, while all other cases can use the two criteria, (1) short distances between particles, and (2) magnetic moments of particles are parallel. Consequently, this table provides a basis for determining the metrics to quantify each structure for all cases.

CASE	STACKS	MICRO-CHAINS	MACRO- CHAINS
1	 Short distances between particles Particles' <i>m</i> in same direction 	 Stacks are close to each other <i>m</i>_{stack} in different directions Stacks form links 	- Limited percolation

Table 3-2. Qualitative descriptions of the structures at each length-scale are given for each processing case.

2	 Short distances between particles Parallel <i>p</i> Anti-parallel m 	 <i>m</i>_{stack} varies within chain Stacks are close together Stack positions along a line 	- High percolation
3	 Short distances between particles Parallel <i>m</i> 	- Low percolation	- Medium percolation
4	 Short dist. Between particles Parallel m 	 Short distances between stacks <i>m</i>_{stack} in same direction <i>p</i>_{stack} in same direction 	- High percolation
5	- Short distances between particles	 Medium percolation Stacks in same direction 	- High Percolation

While the qualitative definitions in Table 2-1 are a necessary step in the process of characterizing the microstructure observed in each processing case, they cannot be solely used to consistently identify stacks, micro-chains or macro-chains from any group of particles representing a microstructure. Furthermore, descriptions such as "short distances between particles," and "parallel magnetic moments of particles" seem obvious when looking at the example microstructures provided in Figures 2-9 through 2-13, but reality can be more ambiguous due to the stochastic nature and numerical approximations of the simulations. There can be cases when particles do not exactly align, or the separation distances may vary in a way that fits a type of statistical distribution. To have a more robust and clear approach to distinguishing these structures, a quantitative approach is necessary.

One method for quantification can be found in graph theory, known as spanning forests. If each microstructure is treated as a graph, with each particle represented by a node, then the nodes can be connected via edges that could be weighted by a function of our choosing. A minimum spanning tree is a subset of all possible edges such that the edges of the subset connect all nodes while minimizing the total edge weight without any circles forming. For this study, one natural weight function to consider is the radial distances between the particles. However, distance-based weights cannot account for some of the most common definitions in Table 2-2, such as particle alignments. Consequently, a spanning tree algorithm using distance-based weights may group particles that may not make sense or represent the conventions proposed in Table 2-2. As an alternative, a weight function based on dipole-dipole interaction energy is proposed, in the form of

$$w_{dip} = \frac{(abc)^{1/3}}{r_j} [3(\boldsymbol{m}_j \cdot \hat{\boldsymbol{r}}_j) \hat{\boldsymbol{r}}_j - \boldsymbol{m}_j].$$
(3-43)

The form of (3-43) is based on the dipole-dipole interaction energy between two magnetic dipoles, normalized by the volume of an ellipsoidal particle. This function can account for both distances between particles and their relative orientations, which can address the definitions in Table 2-1.

With the weights between all pairs of particles, a spanning tree can be constructed for the entire representative microstructure. However, to distinguish between stacks, micro-chains, and macro-chains, an additional parameter must be selected to limit the formations of trees, such that particles can be grouped into multiple trees instead of just one. This parameter can be a maximum allowed weight, d_{tree} , which could be thought of as analogous to a maximum distance if distance-based weights were used.

Based on the limit set by d_{tree} , MATLAB's minspantree algorithm is utilized to find minimum spanning trees for each case. The set of minimum spanning trees is referred to as the minimum spanning forest, where each tree is separated by at least a weight of d_{tree} . Thus, varying d_{tree} will change the number of trees, and consequently the number of particles per tree. In terms of the microstructure, this could mean that a minimum spanning tree could represent a number of structures depending on the scale of d_{tree} . At a low value of d_{tree} , stacks can be obtained; at a medium value, micro-chains can be obtained; and at a large value, macro-chains can be obtained. Thus, the minimum spanning forest can objectively identify these structures from a microstructure, whether simulated or experimentally acquired, such that other quantitative analyses can be conducted (e.g. comparing distribution of orientations).

Two properties are chosen for the analysis in determining d_{tree} for stacks, micro-chains, and macro-chains. The first is the rate of the amount of relative mass added to trees versus d_{tree} . The relative change in mass can help eliminate gradual changes in the number of trees or particles per tree that are due to isolated, single particles slowly joining nearby trees. The second property is the average percolation of trees for a d_{tree} , which is calculated as a percent of the dimension of the control volume.

Each case is simulated five times at N = 200 with the parameters listed in Table 2-2.

Parameter Name	Parameter Name Description		Unit
М	M Magnetization of particles		A/m
ε_{pr}	Relative electric permittivity of particles		N/A
ε_{mr}	ε_{mr} Relative electric permittivity of fluid medium		N/A
H_0	<i>I</i> ₀ External magnetic field		A/m
E_0	External electric field		MV/m
η Dynamic viscosity		3.5	Pa * s
(<i>abc</i>) ^{1/3} Equivalent radius of a particle		0.5	μm
f	<i>f</i> Volume fraction of particles		N/A
a/c (a=b)	=b) Aspect ratio of particles		N/A

Table 3-3. A list of all parameters used in the simulations.

Case 1

The first case is the no-field processing condition, where particles are left to interact solely based on the field contributions from other particles. In this case, the best indicator of chain, microchain, and macro-chain groupings is the change in mass with respect to d_{tree} . Figure 3-17 plots the change in mass versus d_{tree} , with important events annotated. Stack formations occur at $d_{tree} = 0.03$, micro-chains at $d_{tree} = 0.1$, and macro-chains at $d_{tree} = 0.85$. These values are validated by comparing the structures the spanning tree has formed, as shown in Figure 3-19, with the definitions in Table 2-2. For example, the stacks for Case 1 should be groups of particles that are in close-proximity and well aligned with each other. Stacks that are close to each other can be connected to make up the micro-chains, which can be connected to form macro-chains that have limited percolation.

Figure 3-19 shows the trees formed within a Case 1 structure at $d_{tree} = 0.03$, which indeed mostly groups particles that are very close to each other, and stacks that form chains with moderate curvatures are separated (as shown in the red circle). The trees at $d_{tree} = 0.11$ are groups of stacks that are near each other, forming micro-chains that curve in various directions, with average percolation of about 10% (with respect to the control volume's length). The trees at $d_{tree} = 0.85$ are groups of micro-chains that are well-aligned, and have medium to high percolation of over 50%. At $d_{tree} = 0.85$, nearly all particles are grouped into one large tree.



Figure 3-17. The rate of change in relative mass of trees (measured by the number of particles in a tree, N_p) with respect to the dipole-energy-based weights for Case 1.



Figure 3-18. The average percent percolation of trees at different d_{tree} values for Case 1.



Figure 3-19. The trees generated by the spanning tree at different thresholds for the weights are plotted and shown for Case 1. Each dot represents the position of a particle. The blue lines indicate edges and the red arrows indicate magnetic dipole moments.

Case 2

In Case 2, the external field induces electric dipoles in the particles, and forms chains in the same direction. In the plane perpendicular to the field, the magnetic moments do not have any order. From the spanning tree analysis, the micro-chains and macro-chains can be determined by both the change in mass and the level of percolation. The large change in mass around $d_{tree} = 0.1$ can be attributed to the formation of micro-chains (Figure 3-20), which have an average percolation of 30% or higher (Figure 3-21). The large change in mass at $d_{tree} = 1$ can be attributed to the formation of macro-chains (See Figure 3-22), which have an average percolation of 80% (Figure 3-21).



Figure 3-20. The rate of change in relative mass of trees (measured by the number of particles in a tree, N_p) with respect to the dipole-energy-based weights for Case 2.



Figure 3-21. The average percent percolation of trees at different d_{tree} values for Case 2.

The trees representing each structure are shown in Figure *3-22*. The micro-chains are links of stacks (particles in anti-parallel arrangements) in close proximity, while macro-chains connect stacks that are farther in distance, but whose axes align (in the direction of chain formation).



Figure 3-22. The trees generated by the spanning tree at different thresholds for the weights are plotted and shown for Case 2. Each dot represents the position of a particle. The blue lines indicate edges and the red arrows indicate magnetic dipole moments.

Case 3

The formations of stacks, micro-chains and macro-chains for Case 3 can be determined from a combination of percolation and change in mass. The stacks are associated with the first large changes in mass, up to $d_{tree} = 0.11$, as shown in Figure 3-23. The stacks have about 15% percolation (Figure 3-24), and look like particles that are well aligned and close to each other, as shown in Figure X3-3. The micro-chains form at around $d_{tree} = 0.85$, which aligns with the first change in mass of trees after $d_{tree} = 0.11$ (Figure 3-24). Furthermore, at $d_{tree} = 0.85$, percolation goes up to about 30%. Figure 2-28 shows that the trees consist of groups of stacks that are relatively well aligned. Macro-chains occur at the next step, at $d_{tree} = 0.875$, with a large change in mass (Figure 3-23), high percolation of 40% or higher (Figure 3-24), and group multiple micro-chains together (Figure 3-25).

Due to the compactness of these events in the [0.85, 0.9] range and the stochastic nature of the simulations, slight shifts between simulation for the same case (i.e. processing condition) can add noise to the data, which is why there are large standard deviations in this range. Thus, while some simulations may have micro-chains occurring at 0.85 and macro-chains at 0.875, others have the same events occur at 0.875 and 0.9.



Figure 3-23. The rate of change in relative mass of trees (measured by the number of particles in a tree, N_p) with respect to the dipole-energy-based weights for Case 3.



Figure 3-24. The average percent percolation of trees at different d_{tree} values for Case 3.



Figure 3-25. The trees generated by the spanning tree at different thresholds for the weights are plotted and shown for Case 3. Each dot represents the position of a particle. The blue lines indicate edges and the orange arrows indicate magnetic dipole moments.

Case 4

In Case 4, the microstructure is expected to contain stacks aligned in the direction of the magnetic field, and chains formed in the direction of the electric field (perpendicular to the magnetic field). From the dipole-energy-based spanning tree analysis, stacks can be associated with the first relatively large change in mass at $d_{tree} = 0.3$ (Figure 3-26), where short stacks of very low percolation (1%, Figure 3-27) consisting of two particles are formed (Figure 3-28). The next big changes in mass occur at $d_{tree} = 0.8$ and $d_{tree} = 0.85$, which indicate the formation of chains, where multiple stacks are grouped (Figure 3-28). This jump indicates the shift to micro-chains, which have a relatively low percolation of 5-10%. Macro-chains occur at $d_{tree} = 0.875$ at the next increase in mass (Figure 3-26) with much higher percolation of 30-60% (Figure 3-27), where microchains are grouped into larger, macro-chains (Figure 3-28). However, the trees in Figure 3-28 for micro-chain and macro-chain groupings based on the dipole-energy-based spanning trees do not reflect the definitions in Table 2-2. For instance, the micro-chain trees do not connect particles that appear to form links resembling chains. Additionally, the macro-chains connect micro-chains in diagonal paths that follow the magnetic dipole moments of the particles. Since these types of groupings are artefacts of the dipole-energy-based weights, another type of weight for the spanning tree algorithm can be used for the micro-chains and macro-chains. The simplest approach is a distance-based weight function that can connect nearby particles in ways resembling the descriptions for Case 4 in Table 2-2.



Figure 3-26. The rate of change in relative mass of trees (measured by the number of particles in a tree, N_p) with respect to the dipole-energy-based weights for Case 4.



Figure 3-27. The average percent percolation of trees at different d_{tree} values for Case 4.







Figure 3-29. The rate of change in relative mass of trees calculated by a distance-based spanning tree algorithm is potted in (a), with significant peaks circled in red. The average percolation in trees at different values of d_{tree} relative to the size of a particle, $(abc)^{1/3}$ is plotted in (b), with

significant changes in percolation circled. The spanning forests corresponding to $d_{tree} = 2(abc)^{1/3}$ and $d_{tree} = 10(abc)^{1/3}$ are shown in (c), which show groupings that fit the definitions of micro-chains and macro-chains, respectively.

The results from the distance-based spanning tree algorithm in Figure 3-29 can help quantitatively group particles into micro-chains and macro-chains. The rate of change of mass (Figure 3-29) for the distance-based spanning tree algorithm for Case 4 shows high changes at $d_{tree}/(abc)^{1/3} = 2$, 8, and several beyond 12. With this information alone, it is hard to determine which point can be attributed to the formation of micro-chains and macro-chains. Thus, the jumps in percolation (Figure 3-29.b) can help determine which one of these changes in mass is meaningful in terms of the definitions of the structures in Table 2-2. Namely, the micro-chains and macro-chains should be distinctive in terms of percolation. In Figure 3-29.b, there is a jump in percolation at $d_{tree}/(abc)^{1/3} = 2$ to about 60% percolation, and a gradual increase in percolation within the range $d_{tree}/(abc)^{\frac{1}{3}} = [5,10]$ as it plateaus at around 80%. Beyond $d_{tree}/(abc)^{1/3} = 10$, percolation goes up to almost 100%. Given these changes in mass and percolation at various values d_{tree} , the spanning forests confirm that the micro-chains and macrochains occur at $d_{tree}/(abc)^{1/3} = 2$ and $d_{tree}/(abc)^{1/3} = 10$ (Figure 3-29.c).

Case 5

Case 5 is the case with both fields applied in the same direction. As a result, both particles and chains are expected to be oriented in the direction of the fields. The change in mass (Figure 3-30) shows large changes up to $d_{range} = 0.1$, and a few after $d_{range} = 0.8$. The changes in mass up to $d_{range} = 0.1$ can be associated with the formation of either stacks or micro-chains, since at $d_{range} = 0.11$ the trees contain highly-aligned particles in the same direction, with varying degrees of lengths (Figure 3-32). To determine whether these structures are stacks or micro-chains, the percolation is considered (Figure 3-31). After $d_{range} = 0.1$, the percolation is at 30% or higher, which is relatively high for stacks, and thus, these structures would be considered as micro-chains. The next changes in mass at $d_{range} = 0.8, 0.9$ are a reflection of the formation of macrochains, as confirmed by Figure 3-32, which shows that micro-chains that are well aligned are grouped together into macro-chains at $d_{range} = 0.9$ with a high percolation of 90%.



Figure 3-30. The rate of change in relative mass of trees (measured by the number of particles in a tree, N_p) with respect to the dipole-energy-based weights for Case 5.



Figure 3-31. The average percent percolation of trees at different d_{tree} values for Case 2.



Figure 3-32. The trees generated by the spanning tree at different thresholds for the weights are plotted and shown for Case 5. Each dot represents the position of a particle. The blue lines indicate edges and the orange arrows indicate magnetic dipole moments.

2.3.4 Dispersions of Particle, Stack, and Chain Orientations for Distance-Based Spanning Trees

Case 1 shows high dispersion across all levels of structure, with values of κ at 0.23 or higher. This quantitative assessment confirms its relatively random state at all length-scales, since there is no external field being applied. Case 2 also shows high dispersion at the particle level, at $\kappa = 0.32$, but the micro-chains are highly aligned ($\kappa = 0.03$) with a center at $\mu = 2.1$ near the direction of the field $\phi = 1.57$. Macro-chains are also highly aligned with $\kappa = 0.05$, and centered at $\mu = 1.77$, which is even closer to the direction of the field than the micro-chains for Case 2. By contrast, Case 3 highly aligned at all levels of structure, with low dispersion and a center at $\mu = 0$ matching the direction of the magnetic field, $\phi = 0$. Case 4, meanwhile, shows high particle in the direction of the magnetic field, which is to be expected. At the stack-level, there is greater dispersion but the center is still aligned with the magnetic field. However, at the micro-chain level, the dispersion is very low and orientations are aligned with the electric field, with a mean of $\mu =$ 1.5. The macro-chains for Case 4 are similar to the micro-chains. Finally, Case 5 has very low dispersion at all levels, with precise alignment of structures toward the direction of both fields in Case 5, which is $\phi = 0$.

Table 3-4. A summary of the results for all cases and the fitted distributions of the orientations of structures at each level.

CASE	PARTICLES	STACKS	MICRO-CHAINS	MACRO-CHAINS
1	$\mu = 1.5$ $\kappa = 0.26$	$d_{stack} = 0.01$ $\mu = 2.1$ $\kappa = 0.23$	$d_{micro-ch} = 0.1$ $\mu = 2.0$ $\kappa = 0.24$	$d_{macro-ch} = 0.85$ $\mu = 2.4$ $\kappa = 0.25$
2	$\mu = 2.4$ $\kappa = 0.32$	$d_{stack} = N/A$	$d_{micro-ch} = 0.1$ $\mu = 2.1$ $\kappa = 0.03$	$d_{macro-ch} = 1$ $\mu = 1.77$ $\kappa = 0.05$

3	$\mu = 0$	$d_{stack} = 0.125$	$d_{micro-ch} = 0.88$	$d_{macro-ch} = 0.91$
	$\kappa = 0.01$	$\mu = 0$	$\mu = 0$	$\mu = 0$
		$\kappa = 0.02$	$\kappa = 0.03$	$\kappa = 0.08$
4	$\mu = 0$	$d_{stack} = 0.4$	$d_{micro-ch} =$	$d_{macro-ch} =$
	$\kappa = 0.03$	$\mu = 0$	$2^{*}(abc)^{1/3}$	$10^{*}(abc)^{1/3}$ (distance-
		$\kappa = 0.09$	(distance-based)	based)
			$\mu = 1.5$	$\mu = 1.5$
			$\kappa = 0.06$	$\kappa = 0.09$
5	$\mu = 0$	$d_{stack} = N/A$	$d_{micro-ch} = 0.1$	$d_{macro-ch} = 0.95$
	$\kappa = 0.02$	$\mu = N/A$	$\mu = 0$	$\mu = 0$
		$\kappa = N/A$	$\kappa = 0.02$	$\kappa = 0.05$

2.4. Conclusions

In this chapter, the methodologies were aimed to answer Hypothesis 1 and complete the related objectives. Hypothesis 1 states that particle simulations utilizing ferrohydrodynamics could provide evidence for whether applying magnetic and electric fields to particles suspended in a fluidic polymer matrix yields a microstructure with multiple levels of hierarchy. To answer the hypothesis, the hierarchical microarchitecture formation of magnetic particles inside PDMS was computationally studied for five cases of processed composites: (1) no-field or control, (2) electric field only, (3) magnetic field only, (4) both fields in transverse and (5) both fields in parallel.

The simulation results were compared to a set of expected results for the five cases based on experimental data from literature. For Case 1, chain formations were random; for Case 2, particles aligned geometrically with the electric field; for Case 3, particles aligned with the magnetic field forming short chains; for Case 4, particles aligned with the magnetic field but formed chains in the direction of the electric field; and for Case 5, particles aligned and formed chains in the directions of both fields. Thus, the ability of the simulations to replicate the expected trends partly validated the methodologies chosen to simulate the particles' behaviors and interactions in the fluids for both fields (i.e., ferrohydrodynamic principles). Furthermore, the differences between the simulation results and expected microstructures also helped understand the behavior of particles and chain formations under ideal conditions. For instance, particles formed anti-parallel arrangements in simulations for Case 2 (under an electric field), which is likely due to both a random initial microstructure and a lack of friction between particles. These differences are important because they demonstrate the kinds of microstructures that can be achieved with "perfect" dispersion of particles prior to processing, and minimal friction between particles. Better dispersion of particles can be achieved with methods through different stirring methods or additional processing such as probe sonification or hydrothermal synthesis, while friction experienced by particles could be aided by a polymer matrix with lower viscosity. Results also suggest that more realistic initial distributions produce results more in line could affect the final microstructures and possibly with experiments.

Additionally, a spanning tree threshold analysis of the simulated microstructures quantitatively provided evidence for multiple levels of structure present in the microstructure, referred to as stacks, micro-chains, and micro-chains. These structures were measured by metrics such as percolation, defined by the percent length covered by a tree of the dimension of the control volume, as well as the change in mass as the threshold for the spanning tree is increased. The spanning tree algorithm incorporated a dipole-dipole interaction-based weight function, as opposed to distance-based weights. Based on comparisons between the two types of weight functions, the dipole-based weight function was found to be the best approach to analyze the microstructures since it helps with connecting particles whose dipole moments that naturally align.

Changes in metrics such as percolation and change in mass helped determine define structures at each length scale (i.e., stacks, micro-chains, and macro-chains). For instance, for Case 5, typical micro-chains exhibit 30% percolation, while macro-chains are over 70% percolation, while micro-chains in Case 3 are only at about 15% percolation, and macro-chains no more than 30%. The level of percolation can directly influence the electrical conductivity of materials, and the change from 30% percolation to over 70% could make the difference between a near-perfect insulator to a highly conductive material.

A von Mises distribution function fitted to the average angles of stacks, micro-chains and macrochains for each case quantified the orientations of these structures for more exact comparisons. For example, particles in Case 2 were almost randomly distributed in angle distributions ($\kappa = 0.32$) while micro-chains were highly ordered ($\kappa = 0.03$). Thus, the microstructure in Case 2 could yield isotropic magnetic properties with high dielectric or electrically conductive properties in one direction. By contrast, the dispersion of magnetic moments in Cases 3, 4, and 5 were very low in the direction of the magnetic field, which would yield highly anisotropic magnetic properties. However, due to the differences in hierarchical organization in those cases, such as the net magnetic moment of stacks in Case 4 being perpendicular to chain directions, the magnetic moment of the bulk material could be re-oriented with respect to macroscopic chains, which can influence other properties such as elasticity or conductivity. Additionally, the differences in chain percolation between Cases 3 and 5 could lead to two different materials with magnetic anisotropy: one with high electric conductivity and dielectric coefficient, and one with high electric insulation and low dielectric coefficient. These findings ultimately show that it is possible to manipulate microarchitectures on multiple length-scales by varying the processing conditions, and that the range of attainable microstructures is wide. As a result, the range of attainable material properties is also wide.
CHAPTER 3

Link Between Microstructure and Electrostrictive Material Response: Development and Validation of Nonlinear Electrostriction Model

This chapter aims to address Hypothesis 2 by developing a microstructure-based model for electrostriction and comparing different network models incorporating a distribution function for both the spatial and polarization-related orientations of dipole moments.

The beginning of Chapter 3 reviews previous modeling methods of polarization and electrostriction and hyperelastic models. Next, a constitutive modeling approach based on network models is developed, and specific cases of the model are compared to experimental data.

Note: Parts of this document are borrowed from the author's JMPS journal publication in 2019 (Erol et al. 2019), and SMASIS 2017 conference proceeding (Erol et al. 2017).

3.1 Literature Review on EAP Mechanics

The aim of this chapter is to provide a background on Hypothesis 1 and the related objectives, which focus on understanding the link between microstructure and response of a dipole-based material. Hypothesis 1 states that network models accounting for microstructure by representing relative locations of dipolar regions in a hyperelastic medium can form an effective framework for modeling electrostriction. The first subsection discusses a new class of EAPs, relaxor ferroelectrics, that is driven by dipoles in its microstructure. The second section briefly covers hyperelastic models and identifies the various aspects of each model that could be beneficial for modeling dipole-based EAPs. The third subsection is a literature review on EAP modeling, from deformable linear dielectrics to nonlinear hyperelastic models incorporating aspects of microstructure. It is found that no single model contains a framework for dipole-based EAPs, but various aspects of different models can be adopted for a new framework that incorporates microstructural characteristics of interest.

3.1.1 Microstructure-property relationship in PVDF-based RFE polymers

Electro-active polymers (EAPs) have gained significant attention over the years for their high electromechanical responses (Q. M. Zhang 1998; Bar-Cohen and Bar-Cohen 2004). Among EAPs, polyvinylidine fluoride, P(VDF), and its copolymer polyvinylidine-fluoride trifluoroethyline, P(VDF-TrFE), are ferroelectric materials that derive their electromechanical properties from polarizable, ferroelectric domains (Nalwa 1995; Dang, Lin, and Nan 2003). More recently, a bulky third monomer was added to the copolymer to deliberately generate defects in the microstructure, breaking down crystalline phase into nano-polar domains (Xu et al. 2001; Ang and Yu 2004). As a result, new PVDF-based terpolymers with relaxor ferroelectric (RFE) properties were developed. Two types of PVDF-based terpolymers are poly (vinylidene-fluoridetrifluoroethylene-chlorofluoroethylene), and poly (vinylidene-fluoride-trifluoroethylenechlorotrifluoroethylene), also known as P(VDF-TrFE-CFE) and P(VDF-TrFE-CTFE), respectively (Xu et al. 2001; Bauer et al. 2004; Chu, B. et al. 2006). The terpolymers mainly differ from ferroelectric materials by exhibiting a slim hysteresis loop in their polarization response, as shown in dashed line in Figure 4-1, (Xu et al. 2001; Klein et al. 2005), which was ascribed to the semicrystalline structure of PVDF-based RFE polymers containing mobile crystalline phases with dipolar responses within an amorphous phase (Xu et al. 2001; Lu et al. 2008).



Figure 4-1. Polarization curves for a ferroelectric material and a relaxor ferroelectric material. Electrostriction in the PVDF-based polymers is dependent on the presence of both crystalline and amorphous phases. Greater presence of the amorphous phase yields greater strains by reducing the stiffness of the material (Lu et al. 2006), while more crystalline phase content increases the polarization response. Thus, researchers studied the tradeoff between the two phases by varying the chemical composition of PVDF-based terpolymers, aiming to tailor the electromechanical properties in a precise fashion (Klein et al. 2005; Lu et al. 2006; Yang et al. 2013). Specifically, increasing the percentage of CTFE expanded the amorphous phase, and reduced the crystal domain sizes (Xu et al. 2001) and their separation distances (Yang et al. 2013), which provided more room between crystalline domains, allowing domains to rotate with less friction. Moreover, an increase in crystallinity increased the stiffness of the materia (Lu et al. 2006). These findings indicate that by varying just the chemical composition, researchers can affect the microstructure to modify the electrostrictive response of these PVDF-based RFE polymers.

Several studies have shown that processing steps significantly affect the microstructure and thereby bulk material properties of RFE polymers. Studies performed by (Sencadas, Gregorio,

61

and Lanceros-Méndez 2009; Silva et al. 2010) explored how the crystallinity and microstructural variations of PVDF, the primary monomer of the RFE polymers, were affected by temperature, processing methods, and other variables. (Bao et al. 2007) observed that for the same composition of PVDF-based terpolymer, different processing conditions affected the amount of polar nanodomains in the material. Through SEM images, (Smith et al. 2014) discovered that thermal treatments with a slow quenching process allowed for larger crystal domain formation during the crystallization process, with fibrillary shape characteristics, and these changes in the microstructure increased the dielectric constant of the material. (Cho et al. 2016) confirmed these results by comparing fabrication methods with and without thermal treatments above the transition temperature of the terpolymer, which results in the dominance of different types of crystalline phases, such as the α -, β - and γ - phases. The results of (Cho et al. 2016) also showed that thermal treatments produce largely a-phase dominant microstructures, while room temperature crystallization produces the larger β -phase crystallites; the crystallization process is important because the shifting of phases affects the electrostrictive response of the material. Ultimately, these findings imply that the shapes, sizes, and phase type of crystalline domains are all important factors in the overall electromechanical response mechanism of PVDF-based EAPs and, furthermore, may be controlled, providing an avenue to tailored electromechanical response. Thus, a predictive model of the mechanics of these EAPs should be able to incorporate as many of these features as possible for the highest level of efficacy.

3.1.2 Background on Hyperelastic Modeling

The first part of this subsection will consider previous modeling methods for the hyperelastic response of polymers, and the latter part will survey tools and methods for combining hyperelasticity theory with electrodynamics. Later, approaches to incorporate orientation and spatial distribution information will be discussed, leading to the focus of this work, which is to address the orientational polarization of the crystalline domains within a coupled hyperelastic framework.

Polymers can exhibit high strains that reach beyond the limits of classical Hookean mechanics, such that a linear model employing Young's modulus as a constant is not sufficient in predicting the elastic response of the material (Mooney 1940). However, the only extant model that incorporates both non-polar amorphous and polar crystalline phases is based in linear elasticity (Capsal et al. 2012). In many works and herein, the mechanical deformation of polymers is assumed to be governed by a nonlinear model that captures experimentally observed response.

Invariant-based modeling was a method adopted initially to characterize hyperelastic materials. (Rivlin 1948) developed a generalized infinite series to predict the behavior of elastic materials in the form of

$$W = \sum_{i=0}^{\infty} \sum_{j=0}^{\infty} C_{ij} (I_1 - 3)^i (I_2 - 3)^j,$$
(4-1)

in which C_{ij} were constants, and I_1 and I_2 were the principle invariants, defined as functions of the principal invariants of the right Cauchy Green deformation tensor,

$$I_1 = tr(\boldsymbol{F}^T \boldsymbol{F}), \tag{4-2}$$

$$I_2 = \frac{1}{2} [(tr(\mathbf{F}^T \mathbf{F}))^2 - tr((\mathbf{F}^T \mathbf{F})^2)].$$
(4-3)

The deformation gradient F is defined in Section 2.

The model that (Rivlin 1948) proposed was a first order approximation of the infinite series in (4-1). This first-order approximation is commonly known as the Neo-Hookean model, and with the form of

$$W = C_{10}(I_1 - 3). \tag{4-4}$$

To provide a better fit to experimental data, the Mooney-Rivlin model added the i = 0, j = 1 term of (4-1) to (4-4) (Rivlin 1948). Following this trend, later models added terms to (4-4). More recently, (Ogden 1997) developed a constitutive model utilizing an indefinite number

of stretch-based terms to obtain a widely adjustable fit to rubber elasticity response. However, the primary disadvantage to invariant-based and stretch-based modeling was that the coefficients scaling each term in the series did not represent physical quantities; attribution to physical quantities only came with additional assumptions of structure not presented in the original models. We seek a model in which adjustable parameters are tied directly to physical quantities, allowing the model to aid understanding of the relationship between microstructure and electrostrictive response.

In contrast to invariant- and stretch-based hyperelastic modeling, (Treloar 1944) began building the framework for physics-based constitutive modeling by exploring the statistical mechanics of polymer chains for rubber. (Treloar 1943) constructed a strain energy function for the elastic response of rubber, the form of which is shown in (4-5).

$$W = \frac{1}{2}NkT(I_1 - 3). \tag{4-5}$$

In (4-5), *N* is the chain-density, k is Boltzmann's constant, *T* is absolute temperature, and λ_i are the principal stretches for i = 1,2,3 in a principal coordinate space. This model's significance was its ability to capture the behavior of rubbers based on polymer chain mechanics. Furthermore, its form was similar to the Neo-Hookean model and thus offered a physical meaning to the coefficient C_{10} such that $C_{10} = \frac{1}{2}NkT$ was proportional to the initial 'rubbery' modulus. Though the model was based on a statistical description of a physical chain (i.e. Gaussian chains), the model did not account for the locking stretch, and consequently did not provide a link to material structure. While the formulation in (4-5) is for a single polymer chain, the same theoretical methodology has been applied to more complex representations of polymer networks such as the three-chain network (M. C. Wang and Guth 1952) and the four chains (i.e. tetrahedral) network models (Flory and Rehner 1943). In addition to these models, an eight-chain model was developed by (Arruda and Boyce 1993), apportioning the energy of the network, still computed through random walk by statistics, to a deforming unit cube geometry under affine deformation in

the principal space. (Arruda and Boyce 1993) discovered that the additional chains provided a better fit to experimental data across multiple deformation states, arguably because the diagonal orientations of the chains provide wider spatial averaging of polymer network behavior. While the efficacy of wider averaging of the network has been shown for purely elastic behavior, it will be explored in this work with regards to electromechanical coupling, specifically concurrent averaging across spatial and orientation distributions of electric dipoles of the crystalline regions of a PVDF-based RFE polymer.

3.1.3 Overview of Literature on EAP modeling

Modeling electrostriction can be a powerful tool in improving electrically actuated materials by being able to predict actuation based on information about the microstructure of the material. Given enough data, a material can be tailored to fit specific purposes and actuation designs with the aid of a high-fidelity electrostriction model. However, electromechanical coupling is often implemented in a simple way to predict the macroscopic deformation of a material by the relation in (4-6).

$$S_{33} = Q_{33} P(E)^2 \tag{4-6}$$

The most frequently studied electromechanical strain is in the direction of the applied electric field, which is prescribed in the third principle direction, by convention. Thus, (4-6) focuses on strain in the 3-direction. The polarization P is a function of E, and is therefore written as P(E), and it is in a quadratic relationship with the strain. The quadratic relationship is useful for an electrostrictive material that observes a nonlinear response between the electric field, and the strain and polarization responses it induces, because the coefficient Q_{33} then becomes constant. Another relation that is commonly studied is

$$S_{33} = M_{33}E^2 \,, \tag{4-7}$$

which is not linearly related like the one observed in (4-6). Instead, the coefficient M_{33} varies with the electric field as it directly relates the electric field and strain. The advantage of (4-7) is

that it only requires knowing the field strength to provide an output for the strain in the 3direction. However, in (4-6), one must know the relationship between electric field and polarization, and then determine the relationship between polarization and strain.

Thus, the relationship found in (4-7) can be easily implemented into the kinematics of a multi-layered beam to determine the response of a composite by only knowing how M_{33} varies with electric field. For an electrostrictive material that exhibits a nonlinear electromechanical response, this relationship is often hard to determine.

The electromechanical deformation of dielectric materials has been extensively studied since the 1950s. (Toupin 1956) first developed the field equations for deformable dielectric materials, where the energy density of the material, U, was determined to be a function of the relative electric permittivity, ϵ_r , and quadratically related to the electric field E, such that U = $(1/2)\epsilon_r\epsilon_0 E^2$; others improved on the field equations in the following years (Truesdell and Toupin 1960; Eringen 1963). Recent additions have been made to the list of dielectric and other active material models that have each added new concepts or mechanisms, such as deformation dependent permittivities (X. Zhao and Suo 2008; Jiménez and McMeeking 2013); a free energy consisting of multiple components attributed to electrostrictive, elastic, dielectric, and residual dielectric energies (Richards and Odegard 2010); viscoelasticity (Hong 2011; Ask, Menzel, and Ristinmaa 2015); hysteresis of ferroelectrics (Lallart et al. 2016); multiplicative electro-elasticity (Skatulla, Sansour, and Arockiarajan 2012; Zah and Miehe 2015); electro-plasticity (Christian Miehe 1998); variational frameworks that model microstructural evolutions of domains in ferroelectric ceramics (C. Miehe, Zäh, and Rosato 2012); multi-scale methods that account for micro-scale dipole rotations (Cohen and deBotton 2014); and homogenization methods that study the effects of particle sizes and distributions on active material properties (Ponte Castañeda and Siboni 2012; Zäh and Miehe 2013). These models have sought to improve existing electro-elastic theories by adding methods that address various complexities of EAP modeling, but they have not

developed semicrystalline microstructure-based modeling of electrostriction that address microscale spatial orientation, and none have specifically modeled the response of RFE polymers.

Recent work has suggested that microscale spatial organization of polycrystalline polarizable regions within a PVDF-based RFE polymer affects its macroscale electromechanical coupling. (Guan et al. 2010) discussed the importance of interactions between ferroelectric domains within an amorphous matrix for PVDF-based polymers, and developed a rudimentary model to predict the depolarization fields for this type of microstructure. The authors pointed out that there was anisotropy in the interactions of polar domains, based on dipole-dipole interactions, and concluded that the relative locations of domains were an important factor in the polarization of semicrystalline EAPs. However, (Guan et al. 2010) did not analyze the electromechanical coupling of their semicrystalline model. The framework proposed herein will account for the anisotropic behavior of dipole-dipole interactions and their spatial organization when computing electrostriction.

Only recently did models emerge in the field of EAPs that have incorporated physical elements of the microstructure of the material to the deformations observed in experiments, although note these works focused on dielectric elastomers, not RFE polymers (Cohen and deBotton 2014, 2015). (Cohen, Dayal, and deBotton 2016) introduced a method in which the polymer network is defined as a segment of a circular chain composed of rigid rods (unit elements) that represent dipoles—which is an interpretation of the microstructure of dielectric elastomers. The alignment of those dipoles under an electric field subsequently generates a deformation in the chain. This model shows promise due to its ability to consider various modes of deformation of individual unit elements within a network model that ties the mechanics of polymer chains to the electrostriction of the material, thus enabling the model to address information within the microstructure in an electrostriction modeling framework. However, since the model represents dielectric polymers with a single phase, it is not applicable to semicrystalline RFE polymers.

(Zah and Miehe 2015) proposed a model (for graft elastomers) with a semicrystalline physical basis. Their model assumed that graft elastomers generated electromechanical strain through the rotations of crystals that pull together neighboring chains via crosslinks. This mechanism, however, is less relevant for PVDF-based RFE polymers, which do not have crosslinks in their microstructure. Instead, dipole-dipole interactions will drive the electromechanical mechanism for RFE polymers, as discussed by Guan et al. (2010).

(Capsal et al. 2012) developed a model that accounted for the *biphasic* constitution of RFE polymers by splitting the bulk polarization response of the material into separate contributions from the amorphous and crystalline phases. In addition, their model incorporated an averaging method for the orientational distribution of dipoles. However, the model did not address the spatial locations of the crystalline domains, a primary characteristic in determining their interaction. Further, the model calculated electromechanical strain using a linear elasticity. While we similarly propose the use of a biphasic model, our microstructural representation of dipole arrangements will account for spatial location and orientation; we further assume a hyperelastic material response.

3.1.4 Classification of Selected Models

For developing a framework for modeling the mechanics of dipole-based EAPs, the literature review focuses on a few models, which are classified into the following categories: (a) dipole orientation models, (b) polarization models, (c) polarization-electrostriction models, and (d) only electrostriction models. Studying the models from the perspective of these categories can aid in finding solutions for addressing various physical phenomena in the proposed model.

Type (a) models focus on the dipole orientations of materials and how these orientations are affected by an external electric field. Type (b) models describe the average polarization density (macroscopic polarization), while Type (c) models either borrow an already existing macroscopic polarization description or develop their own, and implement it into a constitutive relation with the macroscopic strain or deformation of the material (i.e., electromechanical coupling). Lastly, Type (d) models forego describing polarization on its own, and instead develop an electromechanical constitutive model from other methods. Each of these types are listed in Table 4-1.

Туре	Definition
<i>(a)</i>	Dipole orientation models
<i>(b)</i>	Polarization models
<i>(c)</i>	Polarization based electrostriction models
<i>(d)</i>	Direct electrostriction models

Table 4-1. The model types and their definitions based on the parameters modeled.

Additionally, the following criteria will be considered: (1) the material(s) modeled, (2) the modeled parameter(s), (3) parameter(s) with a physical meaning, (4) modeling methods, and (5) dielectric nonlinearity. These criteria are selected based on their relevance or importance with respect to modeling dipole-based EAPs such as the PVDF-based terpolymer.

3.1.4.1 Type (a) Modeling – Dipole Orientation Models

An example of type (a) models is provided by Ref [23], in which first-principles total energy calculations are utilized to examine the effects of an electric field on dipole alignment at the nano-scale for the ferroelectric polymer P(VDF). The energy calculations incorporate a dipoledipole interaction energy model for an infinite dipole array of P(VDF) chains. From these calculations, the authors obtain simulation data for the total energy for both a mono- and bi-layer slab of P(VDF) as a function of the rotation of dipoles. This provides a strong foundation to build models that can predict macroscopic polarization—at least for P(VDF)—by deriving a relationship between the applied electric field and the orientation of dipoles with the implementation of the dipole-dipole interaction energy.

3.1.4.2 Type (b) Modeling – Polarization Models

Reference [24] is an example of (b)-type modeling papers; a macroscopic polarization model is developed by performing Monte Carlo simulations of dipole-dipole interactions within P(VDF) crystals to study polarization hysteresis and ferroelectric-paraelectric phase transition. This study, along with others [25]-[27], emphasizes the importance of dipole-dipole interactions in the polarization of P(VDF)-based polymers.

Reference [49] is another (b)-type modeling approach, which takes polymer chain structures into account. The paper examines the local behavior of a unit model that represents the structure of a dipole unit and considers multiple modes of deformation, such as uniaxial and transversely isotropic. These element types are evaluated, as an example, in a network of polymer chain arc segments, which is directly a physical interpretation of the microstructure of the material. This network model accounts for the non-electrical, mechanical constraints on the movements of chains caused by structural arrangements, and their contributions to the macrodeformation of the polymer. The major drawback of the model is that it results in a polarization function as a linear function of electric field and the model is inherently composed of a framework for linear dielectrics.

The model in Ref [28] employs the Langevin dipole model, which combines Boltzmann statistics with local field calculations to determine the average polarization response in a ferroelectric material with isolated polar regions. The model accounts for local fields at a dipole location and implements distribution functions for the reorientational component of the random field. The local field E_l is defined as

$$\boldsymbol{E}_{l} = \boldsymbol{e} + \boldsymbol{E}_{0} + \gamma \boldsymbol{P} \tag{4-8}$$

where **e** is the field induced on a microdipole by neighboring structures, E_0 is the external field, **P** is the polarization, and γ is the Lorentz constant. Reference [28] defines a dipole moment μ by the Langevin function in (4-9).

$$\mu = \mu_0 tanh\left(\frac{E_0\mu_0}{3k_BT}\right) \tag{4-9}$$

Here, E_0 is the external field, μ_0 is the dipole moment near absolute zero temperature, k_B is the Boltzmann constant, and T is temperature. Next, the dipole moment is substituted into the equation for polarization, which is also a function of the local field E_l as shown in (4-10).

$$\boldsymbol{P} = n\mu \boldsymbol{E}_l(\boldsymbol{r}) / \boldsymbol{E}_l \tag{4-10}$$

In (4-10), $E_l(r)$ is the local field vector with a magnitude of E_l , and n is the dipole density. By redefining E_l as

$$E_l = \sqrt{E^2 + e^2 + 2Eecos\theta_e} , \qquad (4-11)$$

in which θ_e is the angle between the direction of the external field and the direction of the random field. Reference [28] examines the effects of this orientational parameter by assuming that the random field directions fall under some distribution function, with which the average polarization can be calculated, as in (4-12).

$$P = \frac{\mu n}{2} \int_0^{\pi} p(\theta_e, E) \sin\theta_e d\theta_e$$
(4-12)

Furthermore, Ref [28] predicts the linear relationship between polarization and electric field at low fields, and the quadratic relationship at high fields, by incorporating the reorientational component of the random field.

3.1.4.3. Type (c) Modeling – Polarization-based Electrostriction Models

A good example of a (c)-type model is found in Ref [29] where the authors develop a model that predicts the electrostrictive coefficient of an EAP. As a (c)-type model, Ref [29] builds a polarization model as a function of electric field based on Boltzmann statistics and Debye/Langevin formalism. They define polarization as a function of the dipole moments within a volume, V, as shown below in (4-13).

$$\boldsymbol{P} = n\langle \boldsymbol{\mu} \rangle \tag{4-13}$$

In (4-13), **P** is the macroscopic polarization vector, $\langle \mu \rangle$ is the average dipole moment vector, and n is the dipole density. The average dipole moment is split into an average magnitude and average cosine of the angle between the dipole moment and the external field.

$$\langle \boldsymbol{\mu} \rangle = \mu \langle \cos \theta \rangle \tag{4-14}$$

Most importantly, Boltzmann statistics is applied to (4-14) to describe the average cosine by a Langevin function, which results in the relation,

$$\langle \cos\theta \rangle = \coth\left[\frac{E}{E_{Sat}}\right] - \frac{E_{Sat}}{E},$$
 (4-15)

where E is the magnitude of the external field, and E_{Sat} is the electric field when saturation of the dipoles begins. As a result, the polarization is rewritten as

$$P(E) = N\mu \left[coth \left[\frac{E}{E_{Sat}} \right] - \frac{E_{Sat}}{E} \right].$$
(4-16)

The authors implement (4-16) in a unique way by separating the total macroscopic polarization into two separate terms that represent the two different phases of the semicrystalline material: one term is the polarization response of the crystalline phase (P_1), and the other term is the response of the amorphous phase (P_2).

$$P(E) = P_1(E) + P_2(E)$$
(4-17)

The amorphous phase is treated as a nearly constant permittivity that begins saturating at very high electric fields (around 500 MV/m), whereas the crystalline phase saturates at much lower field strengths (under 50 MV/m), which is explained by the gradual orientation of dipoles. Furthermore, the study considers a polarization model that is a function of the orientation of dipoles within the material, defined by (4-15). Lastly, Ref [29] derives the permittivity of the material from the polarization function by the relation,

$$\varepsilon_r = \frac{1}{\varepsilon_0} \frac{dP}{dE} + 1 \tag{4-18}$$

where ε_r is the relative permittivity of the phase/material, and ε_0 is the permittivity of vacuum. The permittivity is substituted into (4-19) to determine the electromechanical strain.

$$S_{Maxwell} = \frac{\varepsilon^* E^2}{Y}, \qquad S_{33} = \frac{P(E)^2}{Y_{\mathcal{E}}},$$
 (4-19.a,b)

In (4-19.a,b), two separate methods are presented for determining the electromechanical response of the material. The first method is by calculating $S_{Maxwell}$, which is the strain generated by Maxwell forces. This term is a function of the nominal permittivity of the material, ε^* , which is a non-constant term that is related to the relative permittivity by the relation $\varepsilon_r = \frac{\varepsilon^*}{\varepsilon_0} + 1$, and the Young's modulus, Y. The second method calculates the electrostrictive coefficient Q_{33} based on the low-field permittivity of the material, ε , which as a result works well for low field responses. Recall that Q_{33} directly solves for the strain in (4-6) by the quadratic relationship with the polarization P(E). In both cases, although the polarization response is nonlinear, the *elastic* response of the material is treated with a linear expression (Y = constant).

3.1.4.4. Type (d) Modeling – Direct Electrostriction Models

The models mentioned so far focus on the polarization response of the material, and the ones that solve for electromechanical coupling have only done so by assuming a linear elastic response. As a (d) type model, Ref [51] offers the first approach that considers hyperelasticity by postulating a Helmholtz free energy function in the form of

$$W = A_1 + A_2 + A_3 + A_4 , (4-20)$$

where A_i represent a set of contributions to the free energy of the material and the authors of Ref [51] have prescribed them as the elastic, electrostrictive, dielectric, and residual electric displacement responses, respectively. The elastic response of the free energy function is provided

by a modified Mooney-Rivlin model [33[34]. The electrostrictive response function in (4-20) is similar to that of the linear dielectric energy density, with a quadratic relation with E.

$$A_2 = -\frac{1}{2}(\alpha_6 + \alpha_7 T)E_1^2(I_4 - 1)$$
(4-21)

In (4-21), α_6 and α_7 are material parameters, and I_4 is the determinant of the right Cauchy Green deformation tensor. The dielectric response is given by a similar function with a quadratic dependence on the electric field in the three principal axes. As such, the combined effect of the electrostrictive and dielectric response functions describes the general response of a linear dielectric under an arbitrary electric field. The electric displacement is assumed as a function of the residual electric displacement value multiplied by the electric field in the 1direction. Although the study considers multiple cases of boundary conditions to reduce and simplify the total free energy function to model for some applications, there are no comparisons with experimental data, and ultimately the model only partially utilizes variables with a physical basis.

Reference [50] finds a quantum explanation for the negative piezoelectric phenomenon that PVDF experiences by modeling PVDF chains at the nanoscale. Piezoelectric materials observe an induced strain under an electric field, and an induced field under a mechanically generated strain; both effects are linearly related. Unlike most piezoelectrics, however, P(VDF) exhibits contraction, i.e., negative strain, in the direction of the field. The authors apply first principle calculations with quantum-chemical methods (QCM), and analyze the changes of dipole moment orientations and lattice deformations to determine the piezoelectric coefficient d_{33} for P(VDF). In addition to this coefficient, they calculate other properties such as the polarizability, dielectric permittivity ε , electrostrictive coefficient Q, and compare them with experimental results. As a side note, since the model predicts a coupling coefficient without deriving an expression for the polarization, it falls under the (d)-type model category. Most recently, Ref [31] emerged with a more integral approach to the overall modeling of electrostriction. Similar to Ref [51], they utilize the free energy density function of a dielectric material. In addition, a deformation dependent electric permittivity is implemented instead of a constant value. The study follows with the rigorous development of a continuum-based model that considers the effects of cross-linking of the polymer chains on the electromechanical coupling. The work specifically focuses on graft elastomers, which consist of backbone chains connected via flexible side chains that contain crystalline, polar regions, which rotate under an applied electric field. As the crystal units rotate, they pull the surrounding backbone chains together.

The model proposed by Ref [31] agrees well with experiments. However, this type of deformation may only be valid for graft elastomers due to the assumption that cross-links are prevalent in the polymer network. On the other hand, cross-links do not exist in the P(VDF)-based terpolymer, as previously mentioned, and as a result, a major component of the mechanism for electrically-induced deformation is significantly different between the relaxor ferroelectric polymer and the graft elastomer.

model	Modelea	туре	Modeling Methods	Physical	Dielectric
	Material			basis	Nonlinearity
[23]	P(VDF) (ferroelectric)	a	Dipole-dipole interaction model	Yes (dipole moments)	N/A
[24]	P(VDF) (ferroelectric)	b	Monte Carlo simulations of dipole interactions	Yes (dipole moments)	Yes
[50]	P(VDF) (piezoelectric)	b	Quantum-Chemical simulations	Yes (dipole moments)	No
[49]	Dielectrics	b/c	Electromechanical energy density with polymer chain deformation	Yes (dipole moments)	No
[28]	Ferroelectrics with isolated polar regions	с	Langevin dipoles, probability distribution of field orientations	Yes (dipole orientations)	Yes

Table 4-2. Comparisons of the various models discussed with respect to the relevant criteria.

[29]	Electrostrictive polymers	с	Debye/Langevin Formalism	Yes (multi- phase Polarization)	Yes
[24]	P(VDF) ferroelectric	d	Molecular Dynamics	Yes	Yes
[51]	Dielectric	d	Helmholtz free energy; electromechanical terms	No	No
[31]	Graft elastomers	c/d	Non-affine deformation; stretch- dependent permittivity	Yes (rotation of crystal units with cross- linking)	Yes

The comparisons in Table 4-2 highlight some of the advantages and disadvantages of each model, and demonstrates the different approaches taken by each model. The wide range of modeling types (a)-(d) illustrate that many approaches can be taken to modeling electrostriction: type (a) models seek the relationship between electric field E and the rotation of dipoles; type (b) models seek the relationship between E and the macroscopic polarization P; type (c) models seek the relationship between E and the field-induced strain S by first determining the relationship between E and P and then by determining the relationship between P and S; type (d) models directly determine the relationship between E and S by methods such as hyperelasticity. Some of the models in Table 4-2 implement averaging methods based on dipole statistics, or molecular dynamics implementing the dipole-dipole interaction energy [24[28]. Other models utilize some aspects of the microstructure, such as dipole unit shapes, chain orientations and crystal region rotations [31[49]. However, there are no in-depth studies on crystal domain arrangements that incorporate the dipole-dipole interaction energy based on dipole statistics or distributions. As a result, this gap in the literature warrants the formulation of a constitutive model that captures the variations in the microstructure of EAPs in terms of relative polar domain orientations with respect to each other and with respect to the external field by applying deriving the free energy of various network models.

3.1.5 Summary of Literature

In summary, PVDF-based polymers are semicrystalline polymers, requiring models that describe their biphasic, crystalline and amorphous, constitution. The spatial and orientation distributions of these polarizable crystalline regions, which may be controlled through composition and processing, affect electromechanical coupling and hence are important quantities to any modeling attempt. The spatial organization of the crystalline regions formed in these relaxor ferroelectric (RFE) polymers during processing varies, requiring the development of a model capable of capturing what is known about terpolymer morphology. Thus, the main goal of this study is to construct an appropriate network model for a P(VDF)-based relaxor RFE polymer that ties microstructure to electromechanical coupling by incorporating chain kinematics and deformation for polymeric response; spatial and orientation averaging methods of polarizable domains' relative locations and orientations; and a representation of the material's semicrystallinity, i.e. its biphasic constitution. A secondary purpose of this model is to guide future researchers in choosing a target design for the microstructure of a dipole-based material in terms of domain arrangements in order to achieve their desired material properties. Thus, the model will build a foundation for more complex analyses on understanding the relationship between specific parameters within the microstructure and bulk properties of the material.

3.2 EAP Modeling framework

In this section, the mechanics of the dipole-based EAP system is developed. The first subsection outlines the kinematics of a semicrystalline, deformable body consisting of rigid and elastic regions; the second subsection provides the traction-free equilibrium equations with related constitutive laws; the third subsection adds energies for the system; the fourth subsection relates the orientation of dipolar regions to the polarization; the fifth subsection derives the network model incorporating both crystalline and amorphous regions; and the sixth subsection provides methodologies for deriving the permittivity of the amorphous region.

3.2.1 Kinematics of a semicrystalline, electrostrictive body

The discussion begins by defining a body Ω consisting of two phases (see Figure 4-2). It is assumed that there are distinct boundaries at the interfaces between the two phases, such that the two phases are contained in separate domains. An amorphous phase is contained in the nominally contiguous subspace, Ω_0 , and the crystalline phases are a collection of discrete domains within the subspaces, Ω_i (for $i = 1, 2, ..., N_c$). The subspaces are defined such that

$$\Omega = \Omega_0 \cup \Omega_{C_1} \cup \Omega_{C_2} \cup \dots \cup \Omega_{N_c} \,. \tag{4-22}$$

Figure 4-2 illustrates the deformation of Ω , which can be represented by the linear mapping χ between the undeformed body, and the deformed body. The position vectors in the undeformed and the deformed configurations are **X** and **x**, respectively. The deformation χ can be written as a function of the position vectors in the reference configuration, **X**, as well as other parameters that affect the deformation, such that $\chi = \chi(X, E, O)$, where *E* is the applied electric field and $O = \{O_A, O_C\}$ contains sets of parameters relating to the properties of the amorphous and crystalline phases of the material. For simplicity, the bases in both configurations are prescribed as the orthonormal set, $\{\hat{e}_1, \hat{e}_2, \hat{e}_3\}$. The components of the deformation gradient of Ω are defined in the reference configuration by

$$F = \frac{\partial \chi(X, O)}{\partial X} = \nabla_X x.$$
(4-23)

In (4-23), **F** is the deformation gradient, which is a second-order tensor, and ∇ is the gradient operator in a three-dimensional point space. The left and right Cauchy-Green deformation tensors are defined in terms of the deformation gradient by the relations, respectively,

$$\boldsymbol{B} = \boldsymbol{F}\boldsymbol{F}^{T},\tag{4-24}$$



Figure 4-2. Two-dimensional schematic of a biphasic body (i.e., a body with inclusions) undergoing deformation. The left body is in the undeformed configuration and the body on the right is in the deformed configuration. Two dipolar regions are highlighted with electric dipole moments p_1 and p_2 with a vector R between them in the undeformed configuration and r in the deformed configuration.

Expressing any vector **R** between any two crystalline domains Ω_i and Ω_i in the undeformed

configuration in terms of the basis vectors yields

$$\boldsymbol{R} = R_1 \hat{\boldsymbol{e}}_1 + R_2 \hat{\boldsymbol{e}}_2 + R_3 \hat{\boldsymbol{e}}_3. \tag{4-26}$$

where R_k represents the component of **R** in \hat{e}_k for k = 1,2,3. After deformation χ ,

$$\boldsymbol{r} = \boldsymbol{F}\boldsymbol{R}.\tag{4-27}$$

The magnitude of \boldsymbol{r} can be expressed as

$$r^2 = FR \cdot FR. \tag{4-28}$$

We assume Ω undergoes affine deformation, and further that crystalline domains Ω_i are free to rotate. The relatively large stiffness of the crystalline phase with respect to the amorphous phase

(4-25)

strongly apportions stretch to the amorphous phase. Consequently, crystalline regions are assumed to be rigid. Additionally, the rotations of the crystalline regions will have no impact on the elastic or dielectric response of the material. These assumptions follow findings of Yang et al (2013). For the purpose of this study, we choose a representative volume element (RVE) composed of a single spherical crystalline region surrounded by a spherical amorphous region, as shown in Figure 4-3.

Given the assumptions made on the crystalline regions (rigidity and ability to freely rotate), any polarization vector p_i of any domain Ω_i is a function of E and O. This implies that p_i may be assumed to be uninfluenced by mechanical deformation. The field-dependent form of p_i will be later defined by electrodynamics.



Figure 4-3. The representative volume element contains a spherical crystalline domain, surrounded by a concentric amorphous domain that is initially spherical and deforms into an ellipsoidal shape after deformation.

Based on the symmetry of the RVE, a differential element can be drawn similar to a "prototype" semicrystalline chain developed by Nateghi et al. (2018). In this prior work, the crystalline region is represented as a straight, rigid line segment while the amorphous region continues from the end of the crystalline segment along a representative random walk. Considering this type of element will allow us to write the kinematics of the semicrystalline RVE in terms of the microstretch, and relate it to the macro-stretch. The newly defined semicrystalline element in Figure 4-4 occupies an infinitesimal region in space, which is divided among the two phases. In our work, the crystalline region is indicated by ordered, rectilinear chain paths, while the amorphous region contains less ordered paths resembling a random walk. In Figure 4-4, the left element is in the undeformed configuration, and the right element is in the deformed configuration; the configurations are annotated with their respective notations, and each adopt an appropriate spherical coordinate system, as depicted in Figure 4-4.



Figure 4-4. The deformation of a segmented element consisting of crystalline and amorphous phases is presented.

The element contains a rigid crystalline region of length $r_c \equiv R_c$, and a deforming amorphous region of length r_A (see Figure 4-4). The total length of the semicrystalline element in the undeformed state is $R_{sc} = R_c + R_A$, and in the deformed state, $r_{sc} = r_c + r_A$, which can be related to the macroscopic deformation by the relations in (4-27) and (4-28). A spherical coordinate system is adopted for the undeformed and deformed configurations, as depicted in Figure 4-4. The subscript "sc" denotes parameters used for the semicrystalline differential element.

Following Nateghi et al. (2018), although the crystalline region is assumed rigid (relatively), the amorphous region may stretch, yielding the relationship $r_A = \lambda_A R_A$, where λ_A is the stretch of the amorphous region. The stretch along r of the two-segment element, λ , can be defined as,

$$\lambda = \frac{\lambda_A R_A + R_C}{R_A + R_C}.$$
(4-29)

The length of the crystalline segment can be determined if the material's percent crystallinity X_c is known, and defined as

$$V_c = v_c V, \tag{4-30}$$

82

where V_c is the volume occupied by the crystalline phase, and V is the total volume of the RVE. These volumes are spherical, which yields

$$V = \frac{4\pi}{3}R^3,$$
 (4-31)

$$V_c = \frac{4\pi}{3} R_c^{3}.$$
 (4-32)

Equations (4-30)-(4-32) can be combined to determine the initial lengths of the crystalline and amorphous regions,

$$R_{c} = v_{c}^{\frac{1}{3}}R,$$

$$R_{A} = (1 - v_{c}^{\frac{1}{3}})R,$$
(4-33)

which can be substituted into (4-29) to yield

$$\lambda = \lambda_A \left(1 - v_c^{\frac{1}{3}} \right) + v_c^{\frac{1}{3}}.$$
(4-34)

Equation (4-34) relates the semicrystalline element's stretch to the crystallinity of the material and the stretch of the amorphous segment. If the material contains 0% crystalline phase, then $v_c =$ 0, yielding $\lambda = \lambda_A$. Conversely, if the material is 100% crystalline, then $v_c = 1$, and $\lambda = 1$ (completely rigid). This equation is thus a good start for the relation between the total chain stretch and the amorphous segment stretch, but must still be related to macroscopic deformation.

The semicrystalline element's stretch can be related to the macroscopic stretches by writing r_{sc} in terms of its components r_1 , r_2 , and r_3 ,

$$r^2 = r_1^2 + r_2^2 + r_3^2, (4-35)$$

where $r_i = \mathbf{r} \cdot \hat{\mathbf{e}}_i$ for i = 1,2,3. Relating each component to the undeformed total length and macroscopic stretches,

$$(\lambda R)^2 = (\lambda_1 R_1)^2 + (\lambda_2 R_2)^2 + (\lambda_3 R_3)^2, \tag{4-36}$$

By following a procedure similar to Wu and van der Giessen (1992), we relate the components of the semicrystalline element's length to the element's orientation in the undeformed configuration,

$$R_2 = Rsin\Phi sin\Theta,$$

$$R_3 = Rcos\Phi,$$
(4-37)

which yields

 $R_1 = Rsin\Phi cos\Theta$,

$$\lambda = \sqrt{(\sin\phi\cos\theta\lambda_1)^2 + (\sin\phi\sin\theta\lambda_2)^2 + (\cos\phi\lambda_3)^2}.$$
(4-38)

The relation in (4-38) can be combined with (4-34) to write λ_A in terms of the principal

stretches,

$$\lambda_A = \frac{1}{(1-\nu_c)} \sqrt{(\sin\phi\cos\theta\lambda_1)^2 + (\sin\phi\sin\theta\lambda_2)^2 + (\cos\phi\lambda_3)^2} - \frac{\nu_c}{(1-\nu_c)}.$$
(4-39)

Similarly, λ_A may be written in an Eulerian description as λ_A (),

$$\lambda_{A} = \frac{1}{(1-\nu_{c})} \left[(\sin\phi\cos\theta)^{2} \lambda_{2}^{-2} + (\sin\phi\sin\theta)^{2} \lambda_{2}^{-2} + (\cos\phi)^{2} \lambda_{3}^{-2} \right]^{-1/2} - \frac{\nu_{c}}{(1-\nu_{c})}.$$
(4-40)

The derivation of $\lambda(\phi, \theta)$ can be found in Wu and van der Giessen (1992).

For later calculations, it will also be convenient to express the principal stretches of only the amorphous region, which are denoted by λ^{A}_{i} . The relations between the components of r, r_{c} , and r_{A} can be established as

$$r_i = r^c{}_i + r^A{}_i, aga{4-41}$$

where $r_i^c = r_c \cdot \hat{e}_i$ and $r_i^A = r_A \cdot \hat{e}_i$ (superscripts not to be confused with contravariant components). Similar to the procedure used to obtain (4-36), we can write

$$\lambda_i R = R_c + \lambda^A{}_i R_A, \tag{4-42}$$

and with the help of (4-33), substitutions for R_c and R_A can be made. After rearranging (4-41), λ^A_i can be determined,

$$\lambda_i^A = \frac{\lambda_i - \nu_c^{\frac{1}{3}}}{1 - \nu_c^{\frac{1}{3}}}.$$
(4-43)

This relation direction relates the stretch of the amorphous region in the micro-scale semicrystalline chain λ_i^A to the macroscopic stretch λ_i .

3.2.2 Traction-free equilibrium

The electrostrictive material is assumed to have an electromechanical strain energy-density, W. The Cauchy stress, T, can be written as a function of W by a constitutive law between W and Tderived for an isothermal, electromechanical process (developed by Richards, et al. (2010) via the Clausius-Duhem inequality). Thus, the expression for T is given by

$$\boldsymbol{T} = \frac{2}{J} \frac{\partial W}{\partial \boldsymbol{B}} \boldsymbol{B} + q \boldsymbol{I}, \tag{4-44}$$

where q is a Lagrange multiplier enforcing the incompressibility constraint, and I is the second order identity tensor. In describing the behavior of a small section of this electrostrictive material, it is assumed the body is traction-free and its boundary $\partial \Omega$ is not subjected to any constraints on deformation. These assumptions represent traction free self-equilibrium (free deformation), T =**0**.

From (4-44), the forms of each stress are

$$T_{11} = \lambda_1 \frac{\partial W}{\partial \lambda_1} + q, \qquad T_{22} = \lambda_2 \frac{\partial W}{\partial \lambda_2} + q, \qquad T_{33} = \lambda_1 \frac{\partial W}{\partial \lambda_3} + q.$$
(4-45)

The purpose of this model is to determine the electromechanical response of the material operating in a planar electrostriction mode, which is often measured in terms of strain in the 3-direction versus electric field. Thus, the principal stress difference between is considered,

$$T_{33} - T_{11} = \lambda_3 \frac{\partial W}{\partial \lambda_3} - \lambda_1 \frac{\partial W}{\partial \lambda_1}, \tag{4-46}$$

eliminating the unknown hydrostatic stress.

Equations (5) - (14) form the kinematic basis for the model developed in this work. In the next section, the Helmholtz free energy relating elastic to electric energy densities within the EAP is derived such that the Cauchy stresses may be determined.

3.2.3 Electromechanical response of a hyperelastic biphasic body with dipole-dipole interactions

This section outlines the methodologies for including microstructure as defined by averages of crystalline domains' relative spatial locations and orientations in to the electrostriction model. The biphasic aspect of the microstructure of PVDF-based RFE polymers is also considered through free energy contributions of each phase. It is assumed that the amorphous phase provides a hyperelastic and linear dielectric response while the crystalline phases contribute to the free energy solely through their nonlinear electrostatic interactions.

3.2.3.1 Free energy formulation of a biphasic body

The strain energy density of a body composed of a crystalline and an amorphous phase is postulated as

$$W = W_{A,El} + W_{A,LD} + W_C, (4-47)$$

where $W_{A,El}$ and $W_{A,LD}$ are the elastic and electrostatic (linear dielectric) responses of the amorphous phase, respectively, while W_C is the dipolar response of the crystalline phase. This decomposition of the energy density function into the contributions from the amorphous phase and the crystalline phase is similar to the models developed by Richards (2010) and Zah (2014), which implemented multiple contributions into their material energy densities. The following subsections explore the separate contributions of each phase's response to the total energy density, W.

3.2.3.2 Hyperelastic response of the amorphous phase

The relaxational behavior of semicrystalline RFE polymers is due to the behavior of the amorphous phase (Ang et al. 2005; Lu, et al. 2006; Lu, et al. 2008). We assume that the amorphous phase has a hyperelastic response, which can be described by an energy density function $W_{A,El}$. Furthermore, the problem is simplified by assuming that the ellipsoidal amorphous

phase of the material exhibits affine deformation, with stretch apportioning following (4-39), and its elastic response is governed by the eight-chain hyperelastic model by Arruda and Boyce (1992), due to its efficacy modeling polymeric materials in multiple deformation states and physical network basis. The eight-chain model, after approximation, has the form

 $W_{8ch}(I_1,\lambda_m,C_1)$

$$\approx C_{1} \left[\frac{1}{2} (I_{1} - 3) + \frac{1}{20\lambda_{m}^{2}} (I_{1}^{2} - 9) + \frac{11}{1050\lambda_{m}^{4}} (I_{1}^{3} - 27) + \frac{19}{7000\lambda_{m}^{6}} (I_{1}^{4} - 81) + \frac{519}{673750\lambda_{m}^{8}} (I_{1}^{5} - 243) \right],$$

$$(4-48)$$

where I_1 is the first principal invariant as defined in (4-2); C_1 and λ_m are experimentally determined constants; The constant λ_m also has a physical interpretation: it is the number of rigid links in a polymer chain.

The elastic energy must be written for only the amorphous segment of the semicrystalline element (see Figure 4-4), which demands the modification of I_1 such that it represents the stretch of only the amorphous phase: $I_1^A = I_1(\lambda_A)$. As a result, we obtain $W_{8ch}(I_1^A, \lambda_m, C_1)$ for the elastic energy of the amorphous phase.

Additionally, the elastic energy density must be scaled by the volume fraction occupied by the amorphous phase, which is proportional to $(1 - v_c)$,

$$W_{A,El} = (1 - v_c) W_{8ch}(I_1^A, \lambda_m, C_1).$$
(4-49)

Since the elastic energy describes the energy of our chosen differential element, we must average the energy over the entire range of orientations based on a probability density function, $f_{sc}(\phi, \theta)$. The average energy $\langle W_{A,El} \rangle$ is defined by the integral

$$\langle W_{A,El} \rangle = \frac{(1-v_c)}{A_{el}} \iint_{0\ 0}^{\pi\ 2\pi} f_{\rm sc}(\phi,\theta) W_{8ch}(I_1^A(\phi,\theta),\lambda_m,C_1) \sin\phi d\theta d\phi.$$
(4-50)

The constant A_{el} is a normalizer, determined by

$$A_{el} = \iint_{0}^{\pi} \int_{0}^{2\pi} f_{sc}(\phi, \theta) sin\phi d\theta d\phi.$$
(4-51)

In the Lagrangian description, $\langle W_{A,EL} \rangle$ can also be written as (Wu and van der Giessen, 1992)

$$\langle W_{A,El} \rangle = \frac{\left(1 - v_c^{1/3}\right)}{A_{el}} \iint_{0\ 0}^{\pi\ 2\pi} f_{sc}(\Phi, \Theta) W_{8ch}(I_1^A(\Phi, \Theta), \lambda_m, C_1) sin\Phi J^{-1} d\Theta d\Phi.$$
(4-52)

We assume that the chain's orientation distribution is initially uniformly distributed, since the amorphous region exists in the same amount in every direction around the crystalline domain inside the RVE (see Figure 4-3), resulting in $A_{el} = 1/4\pi$. Furthermore, the Lagrangian description of $\langle W_{A,El} \rangle$ will be of more use to us since the initial distribution $f_{sc}(\Phi, \Theta)$ is known. However, due to the difficulty in obtaining an analytical solution to (4-52), we consider a discretized approach to calculating the integral in (4-52) numerically,

$$\langle W_{A,El} \rangle = n (1 - v_c^{1/3}) \sum_{i}^{N_{\Theta}} \sum_{j}^{N_{\Phi}} f_{sc}(\Phi_j, \Theta_i) W_{8ch}(I_1^A(\Phi_j, \Theta_i), \lambda_m, C_1) sin\Phi J^{-1} \Delta \Theta \Delta \Phi,$$

$$(4-53)$$

where $\Delta\Theta$ and $\Delta\Phi$ are step sizes for the angles, and N_{Θ} and N_{Φ} are the total number of steps, which are related to the step sizes by $N_{\Theta}\Delta\Theta = 2\pi$ and $N_{\Theta}\Delta\Phi = \pi$.

3.2.3.3 Linear dielectric response of the amorphous phase

Following Capsal et al. 2012, the amorphous phase comprises chain structures that generate a linear dielectric response. Its saturation field is very high, but its dielectric constant (approx. 1 to 5) is significantly lower than the effective dielectric constant produced by the crystalline phases (approx. 50 or greater), as discussed in Capsal et al. 2012.

The linear dielectric energy density of the amorphous phase can be written as

$$W_{A,LD} = (1 - v_c) \frac{J}{2} \epsilon_r \epsilon_0 (\boldsymbol{E} \cdot \boldsymbol{E}), \qquad (4-54)$$

where ϵ_r is the relative electric permittivity of the amorphous phase, ϵ_0 is the electric permittivity of free space, and **E** is the applied field. The form of (4-53) is a common representation of a linear dielectric material response, scaled by the volume fraction of the amorphous phase, $(1 - v_c)$.

3.2.3.4 Dipole-dipole interactions of crystalline phases

As noted in Section 1.2, relative spatial and orientation characteristics of dipolar domains are important to their energetic contributions and subsequent electro-mechanical coupling. Consequently, the work herein seeks to develop a model framework that incorporates those dependencies. The model framework begins with a statement of the interaction energy (Duan et al. 2004) between two crystalline domains, treated as nominally point dipoles of dipole moments p_1 and p_2 , as shown in Figure 4-5. The energy is in the form of

$$U_d = \frac{1}{4\pi\epsilon} \left[\frac{\boldsymbol{p}_1 \cdot \boldsymbol{p}_2}{r^3} - \frac{3(\boldsymbol{p}_1 \cdot \boldsymbol{r})(\boldsymbol{p}_2 \cdot \boldsymbol{r})}{r^5} \right].$$
(4-55)

where r is the dipole-dipole separation vector with a magnitude of r, and ϵ is the electric permittivity of the medium that contains the dipoles. This permittivity, ϵ , should not be confused with the overall permittivity of the polymer; it is strictly the permittivity of the space—or medium—between the dipoles.

The free energy of the crystalline domains is assumed comprised wholly of the potential between dipoles. Consequently, the energy density of the crystalline domain W_c can be written as

$$W_C = \frac{1}{V_c} U_d.$$
(4-56)

where the volume of the crystalline phase is $V_c = v_c V_{tot}$. Specifically, (4-56) characterizes the attractive or repulsive potentials between a pair of crystalline domains, associated with Ω_{C_i} . This dipole energy will be coupled to the elastic energy, which characterizes the elastic potential of the amorphous domains, Ω_A , in a representative volume element to derive the total free energy density.



Figure 4-5. Variables defining the orientations and separation of two dipole moments with respect to the basis vector $\hat{\boldsymbol{e}}_3$. The angles calculated in equation (4-84) are visually depicted at relevant locations; α_1 is drawn separately for simplicity.

For a convenient assessment of the electromechanical strains, the dipolar energy density can be written in terms of F by substituting the kinematic relationships in (4-27) and (4-28) and the definition of the interaction energy in (4-55), into (4-56), yielding

$$\widetilde{W}_{C} = \frac{1}{4\pi X_{c} V_{tot} \epsilon} \left[\frac{\boldsymbol{p}_{1} \cdot \boldsymbol{p}_{2}}{[\boldsymbol{F}\boldsymbol{R} \cdot \boldsymbol{F}\boldsymbol{R}]^{3/2}} - \frac{3(\boldsymbol{p}_{1} \cdot \boldsymbol{F}\boldsymbol{R})(\boldsymbol{p}_{2} \cdot \boldsymbol{F}\boldsymbol{R})}{[\boldsymbol{F}\boldsymbol{R} \cdot \boldsymbol{F}\boldsymbol{R}]^{5/2}} \right].$$
(4-57)

Finally, the total energy density of the material is rewritten by combining (4-53) and (4-57),

$$\widetilde{W} = \frac{\left(1 - X_{C}^{1/3}\right)}{A_{el}} \sum_{i}^{N_{\Phi}} \sum_{j}^{N_{\Phi}} f_{sc}(\Phi_{j}, \Theta_{i}) W_{8ch}(I_{1}^{A}(\Phi_{j}, \Theta_{i}), \lambda_{m}, C_{1}) sin\Phi J^{-1} \Delta \Theta \Delta \Phi$$

$$+ \left(1 - X_{C}\right) \frac{J}{2} \epsilon_{r} \epsilon_{0}(\boldsymbol{E} \cdot \boldsymbol{E})$$

$$+ \frac{1}{4\pi X_{c} V_{tot} \epsilon} \left[\frac{\boldsymbol{p}_{1} \cdot \boldsymbol{p}_{2}}{[\boldsymbol{F}\boldsymbol{R} \cdot \boldsymbol{F}\boldsymbol{R}]^{3/2}} - \frac{3(\boldsymbol{p}_{1} \cdot \boldsymbol{F}\boldsymbol{R})(\boldsymbol{p}_{2} \cdot \boldsymbol{F}\boldsymbol{R})}{[\boldsymbol{F}\boldsymbol{R} \cdot \boldsymbol{F}\boldsymbol{R}]^{5/2}} \right].$$

$$(4-58)$$

3.2.4 Orientation dependence of polarization of crystalline domains

3.2.4.1 Determining magnitude of the average dipole moment

When subject to an external field, the crystalline domains, Ω_i , comprise collinear electric dipoles resulting in a net dipole moment p per crystalline domain. We assume that each crystalline domain exhibits an average dipole moment strength, p. It is convenient to write the dipole moment of a crystalline domain as

$$\boldsymbol{p} = p\widehat{\boldsymbol{p}},\tag{4-59}$$

where the dipole moment vector is split into its magnitude p and the orientation component, \hat{p} , which is the unit vector defined as

$$\hat{\boldsymbol{p}} = \cos\theta_p \sin\phi_p \,\hat{\boldsymbol{e}}_1 + \sin\,\theta_p \sin\phi_p \,\hat{\boldsymbol{e}}_2 + \cos\phi_p \,\hat{\boldsymbol{e}}_3,\tag{4-60}$$

where θ_p and ϕ_p are new polar and azimuthal angles defined for the dipole moment vector with respect to the orthonormal basis vectors, as shown in Figure 4-5.

When considering the alignment of dipoles, the angle ϕ_p is of interest, as it defines the alignment of the dipole with respect to the direction of the applied field, prescribed as \hat{e}_3 . This angle can be better visualized in Figure 4-5, which depicts two dipoles separated by some vector r, each assigned ϕ_{p_i} .

The average dipole moment magnitude p can be determined by assessing the polarization of a representative volume element (RVE) under the saturation condition, $E \rightarrow \infty$, which implies $\lim_{E\to\infty} P \rightarrow P_{sat}$, where P_{sat} is the saturated electric polarization density. Assuming all crystalline domains will perfectly align at some saturation field strength, at saturation the dipole moment magnitude p can be nominally expressed as

$$p = \frac{V_{tot}P_{sat}}{N},\tag{4-61}$$

in which *N* is the number of dipoles (i.e., crystalline domains) within the RVE of volume V_{tot} . Note that P_{sat} is a directly measurable quantity. Substitution of (4-60) and (4-61) in (4-59) yields,

$$\boldsymbol{p} = \frac{V_{tot} P_{sat}}{N} \left(\cos \theta_p \sin \phi_p \, \hat{\boldsymbol{e}}_1 + \sin \, \theta_p \sin \phi_p \, \hat{\boldsymbol{e}}_2 + \cos \phi_p \, \hat{\boldsymbol{e}}_3 \right). \tag{4-62}$$

3.2.4.2 Dipole moment orientations based on a probability distribution function

This subsection introduces the concept of polar domain orientations as a function of electric field. This relationship is characterized by a von Mises distribution function incorporating previous descriptions of the average orientation of dipoles to derive an expression for a probability distribution function (PDF) of polar domain orientations that is a function of the applied electric field, *E*.

The dipole-dipole energy in (4-55) is influenced by crystalline domains alignments with respect to the field, and with respect to each other. The alignment of a set of crystalline domains can be described by a distribution function, denoted by $f_{\phi p}$, through which we can determine the relationship between the applied electric field and the average alignment of crystalline domains. The alignment is quantified by ω , as defined in Figure 4-5, which assumes rotational symmetry about \hat{e}_3 (an artifact of assuming in-plane isotropy). The angle is defined in this manner for convenience in later calculations.

We consider the collection of crystalline domains with net dipoles p, of uniform strength, p, with varying orientations about the axis \hat{e}_3 , defined by ϕ_p in (4-60) (see Figure 4-2 and Figure 4-5). For the set of dipoles, the average dipole moment can be expressed as

$$\langle \boldsymbol{p} \rangle = \langle \boldsymbol{p} \, \widehat{\boldsymbol{p}} \rangle. \tag{4-63}$$

In (4-63), the brackets $\langle \cdot \rangle$ denote the average over all dipoles.

The average orientation of the dipoles with respect to \hat{e}_3 based on a probability distribution function is adopted as

$$\langle \cos\phi_p \rangle = \frac{1}{A_{\phi_p}} \int_{-\pi}^{\pi} f_{\phi_p} \left(\phi_p \mid \mu, b_{\phi_p} \right) \cos\phi_p d\phi_p, \tag{4-64}$$

where $f_{\Phi_p}\left(\Phi_p \mid \mu, b_{\Phi_p}\right)$ denotes a von Mises (wrapped normal) distribution, with the form

$$f_{\phi_p}\left(\phi_p \mid \mu, b_{\phi_p}\right) = \frac{e^{b_{\phi_p} cos\left(\phi_p - \mu_{\phi_p}\right)}}{2\pi I_0\left(b_{\phi_p}\right)},\tag{4-65}$$

in which $\phi_p \in [-\pi, +\pi]$, while μ_{ϕ_p} is the mean of the distribution, I_0 is the modified Bessel function of the first kind with order 0, and b_{ϕ_p} is the concentration parameter that determines the width of the distribution of dipole orientations about the mean μ_{ϕ_p} . The term A_{ϕ_p} normalizes the distribution and can be computed by equating the integral of $f_{\phi_p} \left(\phi_p \mid \mu, b_{\phi_p} \right) d\phi_p$ over the domain of interest to unity. In this context, we emphasize that this distribution of dipole orientations refers to the *distribution of dipole orientations of the crystalline domains*, Ω_i . Each crystalline domain is treated as a single dipole. With this distinction in mind, the dipoles—or crystal domains—are assumed to perfectly align with the field along \hat{e}_3 (given $E = E\hat{e}_3$) as $E \rightarrow \infty$, so the mean of the distribution in (4-65) is set to $\mu_{\phi_p} = 0$. It is possible that variable processing or loading conditions may in the future warrant $\mu_{\phi_p} \neq 0$.

The utility of the distribution function lies in its statistical representation of a set of domains that gradually align with an external field. The distribution's concentration, $b_{\Phi p}$, couples the effect of the external field to the change in alignment of the crystalline domains. Without influence of an electric field, it is assumed that crystalline domains exist in a randomly oriented state characterized by $b_{\Phi p} = 0$. A uniform distribution of dipole orientations falls in a full circular range, such that there is an equal probability of a dipole within the collection to be at any orientation between $\phi_p = -\pi$ and $\phi_p = +\pi$. By contrast, a higher value such as $b_{\Phi p} = 5$ means there is a much greater probability of dipole orientations near the mean alignment $\mu_{\Phi p}$. To determine the relationship between electric field and the concentration parameter, we consider the averaging method for dipole orientations found in the appendix of Capsal et al, 2012, in which the average dipole orientation $\langle \cos \phi_p \rangle$ is expressed as

$$\langle \cos \phi_p \rangle = \coth \left[\frac{E}{E_s} \right] - \frac{E_s}{E}.$$
 (4-66)

Thus, combining (4-64) - (4-66) yields

$$\operatorname{coth}\left[\frac{E}{E_{s}}\right] - \frac{E_{s}}{E} = \frac{1}{A_{f}} \int_{0}^{\pi} \frac{e^{b_{\phi_{p}} \cos(\phi_{p} - \mu_{\phi_{p}})}}{2\pi I_{0}\left(b_{\phi_{p}}\right)} \cos\phi_{p} d\phi_{p}, \tag{4-67}$$

which can be used to evaluate b_{ϕ_p} as a function of the experimentally applied electric field strength *E* by seeking successive $[E, b_{\phi_p}]$ pairs that satisfy (4-67), to within a given tolerance, for an experimentally determined constant, E_s . (Note: E_s is not the field at which saturation occurs, but the field when the polarization response breaks from a linear regime. Consequently, a typical RFE polymer will not saturate until far beyond E_s . This notation is adopted to stay consistent with Capsal et al, 2012.) The correlation between *E* and b_{ϕ_p} is shown in Figure 4-6.



Figure 4-6. Relationship between ϕ_p and E as calculated by (4-67).

Figure 4-7 illustrates the relationship between *E* and b_{ϕ_p} with a plot of the distribution in (4-65) at varying electric field strengths. As *E* increases, the probability of alignment increases near $\phi_p = 0$, the direction of *E*.

The purpose of deriving the $[E, b_{\phi_p}]$ pairs is to utilize them in assessing the average of the dot products, $\langle p_1 \cdot p_2 \rangle$, in the interaction energy, (4-57). One approach to calculating $\langle p_1 \cdot p_2 \rangle$ is by

determining the average angle of the dipoles $\langle \phi_p \rangle$ with respect to the field such that the average dipole moment unit vector $\langle \hat{p} \rangle$ is written as

$$\langle \hat{\boldsymbol{p}} \rangle = \cos(\theta_p) \sin(\phi_p) \, \hat{\boldsymbol{e}}_1 + \sin(\theta_p) \sin(\phi_p) \, \hat{\boldsymbol{e}}_2 + \cos(\phi_p) \, \hat{\boldsymbol{e}}_3, \tag{4-68}$$

where $\langle \theta_p \rangle$ is the average angle based on a uniform distribution $f_{\theta_p} \left(\theta_p \mid \mu_{\theta_p} = 0, b_{\theta_p} = 0 \right)$, and $\langle \phi_p \rangle$ is calculated by applying the $[E, b_{\phi_p}]$ pairs determined by (4-67) into

$$\langle \Phi_p \rangle = \frac{1}{A_{\Phi p}} \int_0^{\pi} f_{\Phi p} \left(\Phi_p \left| \mu_{\Phi p}, b_{\Phi p} \left(E \right) \right) \Phi_p d\Phi_p, \tag{4-69}$$

and similarly

$$\langle \theta_p \rangle = \frac{1}{A_{\theta_p}} \int_0^{\pi} f_{\theta_p} \left(\theta_p \mid \mu_{\theta_p} = 0, b_{\theta_p} = 0 \right) \theta_p \, d\theta_p, \tag{4-70}$$

in which A_{ϕ_p} and A_{θ_p} normalize the integrals over the domain.



Figure 4-7. A von Mises distribution function is chosen as the PDF of the electric dipoles. With the implementation of $b_{\phi_p} = b_{\phi_p}(E)$ solved in (4-67), the model represents a set of dipoles progressively aligning with the field direction ($\phi_p = 0$) as *E* is increased.

The form of $\langle \hat{p} \rangle$ incorporated into the first term of (4-57) reduces to $\langle \hat{p} \rangle \cdot \langle \hat{p} \rangle = 1$, which may not always be true, because crystalline domains are not assumed collinear with each other. Alternatively, we can determine the average dot product $\langle \hat{p} \cdot \hat{p} \rangle$, by discretely sampling two sets
of angles, $\{\phi_{p_1}, \theta_{p_1}\}$ and $\{\phi_{p_2}, \theta_{p_2}\}$, based on the distributions (4-69) and (4-70), utilizing a von Mises distribution function in (4-65), and evaluating at $[E, b_{\theta_p}]$ pairs. The sets of angles yields two sets of dipole moment directions, $\{\hat{p}_1\}$ and $\{\hat{p}_2\}$, such that the average dot product $\langle \hat{p} \cdot \hat{p} \rangle$ can be evaluated by

$$\langle \hat{\boldsymbol{p}}_1 \cdot \hat{\boldsymbol{p}}_2 \rangle = \frac{1}{N_p} \sum_{k=1}^{N_p} \{ \hat{\boldsymbol{p}}_1 \}_k \cdot \{ \hat{\boldsymbol{p}}_2 \}_k.$$
(4-71)

Similarly, the average dot products $\langle \hat{p}_1 \cdot \hat{r} \rangle$ and $\langle \hat{p}_2 \cdot \hat{r} \rangle$ can be calculated by

$$\langle \hat{\boldsymbol{p}}_{1} \cdot \hat{\boldsymbol{r}} \rangle = \frac{1}{N_{p}} \sum_{k=1}^{N_{p}} \{ \hat{\boldsymbol{p}}_{1} \}_{k} \cdot \hat{\boldsymbol{r}},$$
(4-72)

$$\langle \hat{\boldsymbol{p}}_2 \cdot \hat{\boldsymbol{r}} \rangle = \frac{1}{N_p} \sum_{k=1}^{N_p} \{ \hat{\boldsymbol{p}}_2 \}_k \cdot \hat{\boldsymbol{r}}.$$
(4-73)

The sample size N_p is studied more in the next section. It is noted that the expressions (4-71)-(4-73) provide a means of estimating the required dot products and carry the assumption that the magnitudes of crystalline domains, p, are uniform across all crystallites, which places variance of (4-63) into \hat{p} , where $p = \langle p \hat{p} \rangle$. As a result,

$$W_{C} = \frac{p^{2}}{4\pi X_{c} V_{tot} \epsilon r^{3}} [\langle \hat{\boldsymbol{p}}_{1} \cdot \hat{\boldsymbol{p}}_{2} \rangle - 3 \langle \hat{\boldsymbol{p}}_{1} \cdot \hat{\boldsymbol{r}} \rangle \langle \hat{\boldsymbol{p}}_{2} \cdot \hat{\boldsymbol{r}} \rangle].$$

$$(4-74)$$

3.2.5 Formulation of electrostrictive model for different network models

3.2.5.1 Derivation of the discrete network model response under a principle deformation mode

In this subsection, we consider the interaction of the RVE with neighboring crystalline domains to study the coupling of the dipole-dipole forces between crystalline domains with the hyperelasticity of the amorphous phase within the RVE. Consider the chosen single-crystallite RVE (see Figure 4-3), which represents any chosen semicrystalline region in the material. The crystalline domain in this RVE, which we assume behaves as a rigid polarizable region (whose dipole moment is treated as a point dipole located at its center), experiences interactions with neighboring dipoles. To represent the dipole-dipole interaction effect of the nearest neighbor crystalline domains on the RVE, we draw a second body containing another crystalline domain at an arbitrary orientation Φ_r from the RVE's center point, as shown in Figure 4-3. The second body is shown more transparent because we are only considering its interaction effects on the main RVE. It should be noted that the ellipsoidal volumes are meant to give geometric definition to the regions of influence of any given crystallite dipole. Consequently, the deformation of these volumes is imposed with respect to the macroscopic deformation gradient, and not, as the figure might suggest, spherical contact mechanics. The interaction energy of the dipoles induces deformation over the affected volumes that produces strain energy within their amorphous phases in balance with the dipole energy. Additionally, it should be noted that the bodies are cut to better show the angles associated with the dipole moments of each crystalline domain. We further assume that all crystalline domains are separated by the same distance $R = 2R_{RVE}$, which allows us to conveniently adopt (4-57) without any changes in notation. The initial case we study is a fixed Φ_r ($\Phi_r = const.$), referred to as the discrete network model, which will allow us to determine the effects of relative spatial locations of the nearest crystalline domains on the RVE. Later adaptations of this framework will provide means of more complex network averaging schemes over a range of Φ_r .

The representative volume element (RVE) is assumed spherical, initially, and incompressible, such that the nominally constant volume can be written as

$$V_{RVE} = \frac{4}{3J} \pi (R_{RVE})^3, \tag{4-75}$$

Where the Jacobian *J* is defined in (4-78).

Given the analysis of this paper focuses on electrostriction, the semicrystalline body is assumed to undergo affine deformation in the principle space, for which the deformation gradient holds the form

$$\mathbf{F} = \begin{pmatrix} \lambda_1 & 0 & 0\\ 0 & \lambda_2 & 0\\ 0 & 0 & \lambda_3 \end{pmatrix}, \tag{4-76}$$

As shown in Figure 4-8, the network model is defined within a Cartesian coordinate system with basis vectors $\{\hat{e}_1, \hat{e}_2, \hat{e}_3\}$ and two spherical coordinate systems with coordinates $\{R, \Theta, \Phi\}$ and $\{r, \theta, \phi\}$ in the undeformed and deformed configurations, respectively. These coordinates allow for more convenient derivations of the energy of the network model.

Maintaining the assumption of uniaxial deformation, the magnitude of the vector \mathbf{r} between the two dipoles in Figure 4-8 can be written in terms of the spherical coordinate system as $\mathbf{r} = R(\lambda_1 \cos \Theta_r \sin \Phi_r \, \hat{\mathbf{e}}_1 + \lambda_2 \sin \Theta_r \sin \Phi_r \, \hat{\mathbf{e}}_2 + \lambda_3 \cos \Phi_r \, \hat{\mathbf{e}}_3)$ (4-77)

where the principle stretch terms λ_i correspond to each principle direction. The common incompressibility assumption is made, so that J = 1, where J is the Jacobian defined as

$$J = det \mathbf{F}.$$
(4-78)

Consequently,

$$J = \lambda_1 \lambda_2 \lambda_3 = 1. \tag{4-79}$$

Typically, electrostriction is measured in terms of the applied field versus strain in the direction of the field, which is conventionally the 3-direction. To comply with existing measures, we set $\mathbf{E} = E\hat{\mathbf{e}}_3$, and put focus on λ_3 . We assume that the directions transverse to the direction of polarization will experience equivalent electromechanical strain, i.e., the principal electrostriction in-plane is isotropic. The resulting stretch dependences can be written as

$$\lambda_1 = \lambda_2 = \lambda_3^{-\frac{1}{2}}.\tag{4-80}$$

Substitution of (4-80) into (4-77) yields a stretch dependent expression for the deformed radius magnitude, r, namely

$$r = R \sqrt{\lambda_3^{-1} \sin^2 \Phi_r + \lambda_3^2 \cos^2 \Phi_r}.$$
 (4-81)

We can also consider rewriting λ_A for the electrostriction case, utilizing (4-80), which reduces (8) to

$$\lambda_A = \frac{1}{(1-v_c)} \sqrt{(\sin^2 \Phi) \lambda_3^{-1} + (\cos^2 \Phi) \lambda_3^{-2} - \frac{v_c}{(1-v_c)}}.$$
(4-82)

Additionally, the constitutive relation in (4-46) can be written in terms of λ_3 by substitution of (4-80). This form is amenable to the determination of electrostrictive response, *E* vs. λ_3 and given by

$$\frac{\partial W}{\partial \lambda_3} = 0. \tag{4-83}$$





Figure 4-8. The RVE is represented by a sphere annotated with quantities with a subscript of 1, while a neighboring RVE is indicated by the second sphere annotated with quantities with subscript 2. Depiction of spheres in (a) represent the undeformed shapes of the RVE and neighboring body with no field present, at an initial relative orientation of crystallites defined, and (b) illustrates the deformed shapes of the RVE and neighboring body under an applied field along \hat{e}_3 , creating nominal \hat{e}_3 -alignment and dipole-dipole forces, and subsequent uniaxial compression of the RVE. Please note that the neighboring RVE display does not represent a spherical contact problem.

The substitution of $R = 2R_{RVE}$, dipole moment magnitude (4-61), N = 1 (for one crystalline domain in RVE), and the radius (4-81) into the energy in (4-74), yields

$$W_{C} = \frac{P_{sat}^{2}}{3\nu_{c}\epsilon(\lambda_{3}^{-1}\sin^{2}\Phi_{r}+\lambda_{3}^{2}\cos^{2}\Phi_{r})^{3/2}} [\langle \hat{\boldsymbol{p}}_{1} \cdot \hat{\boldsymbol{p}}_{2} \rangle - 3\langle \hat{\boldsymbol{p}}_{1} \cdot \hat{\boldsymbol{r}} \rangle \langle \hat{\boldsymbol{p}}_{2} \cdot \hat{\boldsymbol{r}} \rangle].$$
(4-84)

For simplicity, we will drop the subscript on λ_3 such that $\lambda_3 = \lambda$. The dot products in (4-84) are averages determined by the sampling from the von Mises distribution ($N_{\omega} = 10^5$) in (4-65) – (4-67), and the orientation Φ_r . Upon substitutions made into (4-84), and the resulting simplifications, W_C becomes independent of R and R_{RVE} , allowing us to isolate the influences of only orientation components of the dipole positions when calculating electrostriction. Consequently, this RVE structure will suffice for the purpose of this study.

The elastic energy can also be simplified for the electrostriction case. The local invariant, in its general form $I_1^A = (\lambda_1^A)^2 + (\lambda_2^A)^2 + (\lambda_3^A)^2$, can be written as $I_1^A = (2\lambda_3^A)^{-1} + (\lambda_3^A)^2$, in which λ_i^A are the principal stretches of just the amorphous region, as defined in (4-43). With the substitution for I_1^A , the total energy can be written,

$$\widetilde{W} = \frac{(1-v_c)}{A_{el}} \sum_{i}^{N_{\Theta}} \sum_{j}^{N_{\Phi}} f_{sc} (\Phi_j, \Theta_i) W_{8ch} (I_1^A (\lambda, \Phi_j, \Theta_i), \lambda_m, C_1) sin \Phi \Delta \Theta \Delta \Phi +$$

$$(1-v_c) \frac{\lambda^{-2}}{2} \epsilon_r \epsilon_0 (\boldsymbol{E} \cdot \boldsymbol{E}) + \frac{P_{sat}^2}{3v_c \epsilon (\lambda^{-1} \sin^2 \Phi_r + \lambda^2 \cos^2 \Phi_r)^{3/2}} [\langle \boldsymbol{\hat{p}_1} \cdot \boldsymbol{\hat{p}_2} \rangle -$$

$$3 \langle \boldsymbol{\hat{p}_1} \cdot \boldsymbol{\hat{r}} \rangle \langle \boldsymbol{\hat{p}_2} \cdot \boldsymbol{\hat{r}} \rangle].$$

$$(4-85)$$

3.2.5.2 Analysis of the response of two cases of the discrete network model

Two limiting cases emerge from the arbitrarily oriented discrete network model shown in Figure 4-8: the parallel, \parallel , and perpendicular, \perp , discrete neighboring crystallite locations. These cases derive their names from the neighboring RVE's orientation with respect to the applied field. Hence, the parallel discrete network model consists of crystalline domains oriented parallel to *E*, and the perpendicular case assumes neighboring crystalline domains to exist perpendicular with respect to *E*. These two specific cases of the discrete network model are illustrated in Figure 4-9.



Figure 4-9. The discrete network model is shown for two specific cases. The parallel case is shown in (a), where the vector to nearest neighboring crystalline domain, \boldsymbol{r} , is parallel to the external field, while in the perpendicular case in (b) \boldsymbol{r} is perpendicular to the external field.

The significance of the two specific discrete network cases is apparent from the behavior of W_c , which is based on dipole-dipole interactions. The energy (4-84) is a function of the direction of r, and the orientations of p_1 and p_2 . The energy can be reduced in each special case as $E \to \infty$, yielding

$$\lim_{E \to \infty} \overline{W}_{C,\parallel} = -\left(\frac{2P_s^2}{3\nu_c \epsilon}\right) \lambda^{-3},\tag{4-86}$$

And

$$\lim_{E \to \infty} \overline{W}_{C,\perp} = \left(\frac{P_s^2}{3v_c \epsilon}\right) \lambda^{3/2} \,. \tag{4-87}$$

To gain a better understanding of the geometry-dependent qualitative differences between the two limiting cases of the discrete network model, we non-dimensionalize each subcase's energy function by the constant $P_s^2/(3X_c\epsilon)$ leaving

$$W_C^* = W_C \left(\frac{P_s^2}{3\nu_c \epsilon}\right)^{-1} \tag{4-88}$$

In (4-88), the asterisk on W_C denotes its non-dimensionalized property. The limit of the parallel and perpendicular single chain network models as $E \rightarrow \infty$ are expressed below in (4-89) and (4-90), respectively.

$$\lim_{E \to \infty} \overline{W}_{\mathcal{C}, \parallel}^* = -2\lambda^{-3} \tag{4-89}$$

$$\lim_{E \to \infty} \overline{W}_{C,\perp}^* = \lambda^{3/2} \tag{4-90}$$

These energies are compared in Figure 4-10.a as functions of λ , both exhibiting an increase in energy as λ increases. Due to the coefficients in (4-89) and (4-90), the parallel case is greater in magnitude by a factor of 2 than the energy of the perpendicular case at $\lambda = 1$ (as shown in Figure 4-10.a), and the rate of change of the parallel case is higher below $\lambda = 1$ due to the higher order exponential on λ in (4-89)compared to (4-90). Furthermore, the signs of (4-89) and (4-90) are opposite, which implies that in the transition between the two extreme cases, there is a state of orientation that generates zero interaction energy.

The stress generated by the interaction energy of the dipoles can be determined by

$$T_C^* = \frac{\partial W^*_C}{\partial \lambda}.$$
(4-91)

With (4-91), the forces corresponding to the two cases of the single chain network model are shown in (4-92) and (4-93),

$$\lim_{E \to \infty} T_{C,\parallel}^* = 6\lambda^{-4} \tag{4-92}$$

$$\lim_{E \to \infty} T_{C,\perp}^* = \frac{3}{2} \lambda^{1/2}.$$
(4-93)

These dipolar stresses are plotted in Figure 4-10.b. There are significant differences between (4-92) and (4-93): firstly, the parallel case, (4-92), has a coefficient of 6, while the perpendicular case, (4-93), has a coefficient of 3/2. Thus, when $\lambda = 1$ and $E \to \infty$, the ratio of $T_{C,\parallel}^*: T_{C,\perp}^*$, is 4:1. In addition, the fourth-order exponent on λ in (4-92) results in a quartic increase as $\lambda \to 0$, while the exponent in (4-93) is 1/2. Consequently, as $\lambda \to 0$, the ratio of the forces, $T_{C,\parallel}^*: T_{C,\perp}^*$ diverges towards ∞ .



Figure 4-10. The non-dimensionalized energies (a) and the corresponding forces (b) of the two cases of the single chain network model, parallel and perpendicular, are plotted versus the stretch in the direction of the applied electric field. The comparisons highlight key differences in the strengths of the interactive energies/forces between the parallel and perpendicular cases.

In comparison, the dielectric energy of the amorphous phase, under the incompressibility

assumption and $\boldsymbol{E} = E \hat{\boldsymbol{e}}_3$, can be simplified to

$$W_{A,DE} = (1 - v_c) \frac{{\lambda_3}^{-2}}{2} \epsilon_r \epsilon_0 {E_3}^2$$
(4-94)

The stress generated by the amorphous phase's dielectric response can also be defined,

$$T_{A,DE} = \frac{\partial W_{A,DE}}{\partial \lambda_3} = \lambda_3^{-3} \epsilon_r \epsilon_0 E_3^2$$
(4-95)

Recall that the dielectric constant of the amorphous phase ϵ_r is relatively low: approximately between 1 and 5 (Capsal et al. 2012). At low fields, when $E_3 \sim 10^6 V/m$, the order of magnitude of the amorphous phase's dielectric contribution is $\sigma_{A,DE} \sim 10$. At high fields, when $E_3 \sim 10^8 V/m$, $\sigma_{A,DE} \sim 10^4$.

In contrast, the crystalline phase's dielectric response is driven by dipole-dipole interactions. The parallel and perpendicular spatial alignment cases are considered for the dipole-based energy, which yield the stresses,

$$T_{C,\parallel} = \frac{P_s^2 \lambda^{-4}}{v_c \epsilon_r \epsilon_0} F(E),$$

$$T_{C,\perp} = \frac{P_s^2 \lambda^{3/2}}{2v_c \epsilon_r \epsilon_0} F(E).$$
(4-96)

The function F(E) contains the Langevin orientation model (found in second term of equation 33) and determines the dot products of the dipole moment, and separation vectors. The output of F(E) varies between 0 and 1 for the perpendicular case, and 0 and 2 for the parallel case; furthermore, due to a relatively low field saturation of the crystalline phase, the contributions of the crystalline regions will quickly reach their maximum values within the typical range of applied field in experiments, which is between 0 and 150 MV/m. Consequently, the magnitudes of the crystalline phase's forces (at non-extreme λ values; i.e, $\lambda \sim 1$) are largely determined by the order of magnitude of $P_s^2/(v_c \epsilon_r \epsilon_0)$, which is 10^9 if $P_s^2 \sim 10^{-2} C/m^2$, $\epsilon_r \epsilon_0 \sim 10^{-12} F/m$, and $v_c \sim 10^{-1}$. Thus, dipolar stresses generated by the crystalline phase will achieve stress on the order of 10^9 Pa near saturation (i.e., 150 MV/m).

Due to its relatively small stress generation with respect to the crystalline phases, the dielectric contribution of the amorphous phase can be ignored within the ranges that will be analyzed in this study.

Consequently, the non-dimensionalization of the coupled response can be calculated by the traction-free equilibrium of only the crystalline phase's dipolar response and the amorphous

phase's elastic response. Enforcing equilibrium $\frac{dW_C^*}{\partial \lambda} + \frac{dW_{A,El}^*}{\partial \lambda} = 0$, results in

$$\frac{\partial W_{A,El}^*}{\partial \lambda_3} = -\mathrm{H}\left(\frac{\partial W_C^*}{\partial \lambda_3}\right). \tag{4-97}$$

where the parameter H is a combination of model parameters defined as $H = P_{sat}^2/(3C_1v_c\epsilon)$, and the energy $W_{A,El}^*$ is a result of non-dimensionalization, $W_{A,El}^* = W_{A,El}/C_1$. Accordingly, the forms of both sides of (4-97) may be determined by substituting the dipolar energy, (4-84), and the elastic energy in (4-53), into (4-97), yielding

$$\frac{(1-\nu_{c})}{A_{el}}\sum_{i}^{N_{\Theta}}\sum_{j}^{N_{\Phi}}f_{sc}(\Phi_{j},\Theta_{i})W^{*}_{8ch}(I_{1}^{A}(\lambda,\Phi_{j},\Theta_{i}),\lambda_{m})sin\Phi\Delta\Theta\Delta\Phi$$

$$=-H\frac{\partial}{\partial\lambda_{3}}\Big[(\lambda^{-1}\sin^{2}\Phi+\lambda^{2}\cos^{2}\Phi)^{-\frac{3}{2}}\Big](\langle \hat{p}_{1}\cdot \hat{p}_{2}\rangle$$

$$-3\langle \hat{p}_{1}\cdot \hat{r}\rangle\langle \hat{p}_{2}\cdot \hat{r}\rangle)$$
(4-98)

The right hand side of (4-98) is the product of H and the dimensionless stress corresponding to the dipole energy. This dipolar stress will vary for the two extreme cases because the dot products $\langle \hat{p}_1 \cdot \hat{r} \rangle$ and $\langle \hat{p}_2 \cdot \hat{r} \rangle$ depend on placement of neighboring crystalline domains, which affects \hat{r} . In the parallel case ($\mathbf{R} = R\hat{e}_3$), an increase of field will yield $\langle \hat{p}_1 \cdot \hat{r} \rangle \rightarrow 1$, and $\langle \hat{p}_2 \cdot \hat{r} \rangle \rightarrow 1$. In contrast, as the field increases in the perpendicular case ($\mathbf{R} = R\hat{e}_1$), the dot products will approach towards $\langle \hat{p}_1 \cdot \hat{r} \rangle \rightarrow 0$, and $\langle \hat{p}_2 \cdot \hat{r} \rangle \rightarrow 0$. The assessments of $\langle \hat{p}_1 \cdot \hat{r} \rangle$ and $\langle \hat{p}_2 \cdot \hat{r} \rangle$ are performed by sampling a set of dipole moments from the von Mises distribution. The electric field dependence of these dot products causes the dipolar stress to be driven by the electric field, for which we choose a dimensionless parameter, $E^* = E/E_s$, implemented into (4-67) to obtain the orientation distribution parameter b_{Φ_p} used to calculate dipole moment angles.

To visually illustrate equilibrium, the two sides of (4-98), representing the elastic and dipolar stresses, are plotted versus λ in Figure 4-2. The parameter H is set to 1 (one). The intersection

between the elastic and the dipolar stress differences indicates the equilibrium stretch-state for the traction-free body. Figure 4-2.a plots the stress differences for the parallel case, and Figure 4-2.b for the perpendicular case. Initially in each figure, at $E^* = 0$ the dipole energy is identically zero and the elastic energy crosses the *y*-axis at $\lambda = 1$ signaling the undeformed state as expected. As E^* increases, the dipolar stress difference curve amplifies, shifting the equilibrium towards the left, meaning a contraction in the \hat{e}_3 direction (i.e., electrostriction response). However, while the perpendicular case in Figure 4-2.b depicts intersection over the full range of E^* shown, the parallel case in Figure 4-2.a becomes tangent to the dipole stress near $E^* = 7$. Consequently, beyond $\lambda = 7$ there is no solution to (4-98). This phenomenon can be physically interpreted as the field strength at which the dipolar stresses overcome the elastic stresses of the material, resulting in a phenomenon similar to the pull-in instability, as defined in Zhao and Wang (2014).





Figure 4-11. The left- and right-hand sides of (4-98) are plotted versus λ for the extreme cases, (a) parallel single chain, and (b) perpendicular single chain. The blue line is the stress difference associated with the dipolar energy. The intersections of the curves indicate the equilibrium locations.

This type of mechanical failure has been studied for dielectric materials (i.e., pull-in instability), but it has not been studied for RFE polymers in the same manner (Plante and Dubowsky, 2007; Zhao and Suo, 2008; Zhao and Wang, 2014; Dorfmann and Ogden, 2017). Pull-in instability in dielectric elastomers is typically studied via the energy for an ideal linear dielectric material with a Neo-Hookean elastic model, as developed in Zhao and Wang (2014), in the form of

$$W_{LD} = C_{10}(\lambda^2 - 2\lambda^{-2} - 3) + \frac{\lambda^2 D^2}{2\varepsilon},$$
(4-99)

where $C_{10} = \frac{1}{2}NkT$, ε is the dielectric constant, and *D* is the electric displacement in the \hat{e}_3 direction. The nominal stress calculated by Zhao and Wang (2014) is

$$s_3 = C_{10}(\lambda - \lambda^{-2}) + \frac{\lambda D^2}{\varepsilon},\tag{4-100}$$

and D is related to E by the relation,

$$E = \frac{\Upsilon}{h} = \frac{\lambda^2 D^2}{\varepsilon},\tag{4-101}$$

in which Υ is the voltage and *h* is the distance between two compliant electrodes attached to the dielectric elastomer.

Substituting (4-101) into (4-100) and setting $s_3 = 0$ for traction free equilibrium yields

$$\sqrt{\frac{\varepsilon}{C_{10}}\frac{\Upsilon}{h}} = \sqrt{(\lambda - \lambda^4)}.$$
(4-102)

Multiple similarities can be drawn between (4-102) and (4-98). Namely, the left- and righthand sides of both equations represent stresses related to either the elastic or the dipolar energies, with a coefficient $H = \sqrt{\varepsilon/C_{10}}$ in (4-102) and $H = P_{sat}^2/(3C_1v_c\epsilon)$ in (4-98). In our model, ϵ represents the electric permittivity of the medium between the dipoles, whereas in the linear dielectric model, ϵ represents the average permittivity (or dielectric constant) of the entire material. Meanwhile, the differences between the two approaches are evident when considering the physical representations of the models. While the energies explored in our proposed model can be attributed to the interactions between domains and the amorphous phase's elastic response, the dielectric and elastic energy components in (4-99) are not based on any microstructure characteristics, and instead rely on phenomenological modeling.

The electrostrictive response of a material can be calculated by solving the nonlinear equation (4-98) for pairs of $[E, \lambda]$. The percent strain S_{33} (%) commonly reported in experimental results can be related to λ by

$$S_{33} = 100 * \ln \lambda,$$
 (4-103)

yielding pairs of $[E, S_{33}]$. These pairs, when plotted over a range of *E*, define electrostriction; hence, the relationship between *E* and S_{33} will be referred to as electrostriction in subsequent discussions.

3.2.5.3 Derivation of the PDF-based network model

In this section, we consider averaging techniques for the *spatial* orientation aspect of the dipole-dipole interaction energy function, e.g. r. This is not to be confused with the von Mises

probability density function applied to the distribution of *dipole moment vector* orientations. A probability distribution function will enable exploring the effect of intermediate orientations between the parallel and perpendicular cases on the electrostrictive response.

Consider a single (central) dipole residing in the RVE surrounded by a probability density of neighboring dipoles at some distance r from the central dipole and some azimuthal angle Φ_r . The total interaction energy of the system is the volume average scaled by the corresponding probability density, given by

$$\langle W_C \rangle = \frac{1}{A_f} \int_{-\pi}^{+\pi} f_{\Phi_r}(\Phi_r) W_d(\Phi_r) \sin \Phi_r d\Phi_r, \qquad (4-104)$$

where $f_{\Phi_r}(\Phi_r)$ is a distribution function for the spatial orientations of dipoles. However, W_C must be calculated discretely to assess the dipole-dipole interactions under realist conditions; thus, an analytical solution is not obtainable to the continuous integral in (4-104). As a result, the PDFbased average of W_C is calculated *discretely* below, in (4-105).

$$\langle\langle W_C \rangle\rangle = \frac{1}{A_f} \sum_{i}^{N_{\Phi_r}} f_{\Phi_r}(\Phi_{r_i}) W_d(\Phi_{r_i}) \Delta \Phi_r.$$
(4-105)

where the double bracket $\langle \langle \cdot \rangle \rangle$ denotes the discrete average. Given a small arc length, $\Delta \Phi_r$, discretization offers a viable option to solve the integral in (4-104). Furthermore, $\Delta \Phi_r$ is directly related to N_{Φ_r} by

$$\Delta \Phi_r = \frac{2\pi}{N_{\Phi_r}}.$$
(4-106)

A convergence study on the averaged energy in (4-105) evaluated at $W_C(\Phi_{r_i}) = 1$ is plotted in Figure 4-12, revealing that $N_{\Phi_r} = 500$ is sufficiently large to yield an approximation with less than 0.1% error compared to the integral in (4-104) with $W_C(\Phi_{r_i}) = 1$.

To determine the spatial distribution function $f_{\Phi_r}(\Phi_r)$, we consider the location of any dipole, earlier defined as r_{ij} , which lies on the surface ∂V of a sphere. Assuming axisymmetry, the spatial distribution can be defined as a 1D von Mises distribution on $\Phi_r \in [0, \pi]$,

$$f_{\Phi_r}(\Phi \mid \mu_{\Phi_r}, b_{\Phi_r}) = \frac{e^{b_{\Phi_r} cos(\Phi_r - \mu_{\Phi_r})}}{2\pi I_0(b_{\Phi_r})},$$
(4-107)

where μ_{Φ_r} is the mean, and b_{Φ_r} is the concentration of the distribution of azimuthal angles Φ_r . As depicted in (4-98), the dipolar energy is not dependent on Θ , and consequently, the probability density is not affected by Θ , again a consequence of assuming isotropy in-plane. Moreover, the only adjustable parameters in this model is b_{Φ_r} because the mean is assumed collinear with the applied field direction \hat{e}_3 ; hence $\mu_{\Phi_r} = 0$.



Figure 4-12. The discretized distribution function in (4-105) is evaluated at $W_C(\Phi_{r_i}) = 1$ for varying values of N_{Φ_r} .

The averaged dipolar energy can be written by substituting (4-84) and (4-107) into (4-105), yielding

$$\langle\langle \overline{W}_{C}\rangle\rangle = \sum_{i=1}^{N_{\Phi_{r}}} \left[\frac{e^{b_{\Phi_{r}} cos(\Phi_{r} - \mu_{\Phi_{r}})}}{2\pi I_{0}(b_{\Phi_{r}})} \right] \left[\frac{P_{sat}{}^{2} [\langle \widehat{\boldsymbol{p}}_{1} \cdot \widehat{\boldsymbol{p}}_{2} \rangle - 3\langle \widehat{\boldsymbol{p}}_{1} \cdot \widehat{\boldsymbol{r}} \rangle \langle \widehat{\boldsymbol{p}}_{2} \cdot \widehat{\boldsymbol{r}} \rangle]}{3N_{\Phi_{r}} v_{c} \epsilon (\lambda^{-1} \sin^{2} \Phi_{r} + \lambda^{2} \cos^{2} \Phi_{r})^{3/2}} \right].$$

$$(4-108)$$

The distribution parameter is defined as $b_{\Phi_r} \in [0, \infty)$. Due to its undefined maximum, b_{Φ_r} is difficult to relate to a physical structure. For performing a microstructure-based analysis, the model will be evaluated with respect to $\kappa \in [0, 1/3]$, which, given the von Mises distribution, has been used to discuss various physical structures in literature (Gasser et al, 2006). It is evaluated as

$$\kappa = \frac{1}{4} \int_0^{\pi} f_{\Phi_r} (\Phi \mid \mu_{\Phi_r}, b_{\Phi_r}) \sin^3 \Phi_r d\Phi_r,$$
(4-109)

and as a result, (4-107) can be plotted as a function of κ . The significance of κ is that it is a more physically relatable term; $\kappa = 0$ implies a singular orientation with unit probability, while $\kappa = 1/3$ implies a uniform distribution. Since $\kappa \in [0,1/3]$ is a clearly defined parameter, it will be used in the remaining calculations and analyses.

Various configurations of the distribution in (4-107) can be conceived by modifying μ_{Φ_r} and κ , as an alternative to the single chain network models. The distributions can be visualized by scatter plot of randomly generated points based on the distribution, as in Figure 4-13. The examples shown in Figure 4-13 are in the reference configuration, representing the spatial distribution of dipoles over an initially spherical, undeformed state of the RVE. Each point indicates a dipole that interacts with the central dipole. The spatial distribution is plotted for two cases, $\mu_{\Phi_r} = 0$ and $\mu_{\Phi_r} = \pi/2$ (representing nominally parallel and perpendicular single chain RVEs), at $\kappa = 1/3$, 1/4, 1/10, 1/20, 1/80. The distribution at $\kappa = 0.333$ is a uniform distribution, and thus it is the same for any μ_{Φ_r} . As κ is gradually decreased, the points begin coalescing towards their respective centers; eventually, as $\kappa \to 0$, all points will reside at a single location.



Figure 4-13. The von Mises PDF is graphically represented by a scatter plot of randomly generated points based on the distribution evaluated at varying values of κ . The PDF is centered either at $\mu_{\Phi_r} = 0$ and $\mu_{\Phi_r} = \pi/2$ as shown, where these distributions move toward approximating the parallel and perpendicular discrete single chain models .

3.2.5.4 Analysis of the response of the PDF-based single chain network model

This subsection evaluates the total energy density substituted into (4-98) at varying electric field strengths to simulate the electromechanical response of the material. To study the effects of the spatial distribution function, the averaged dipolar energy in (4-108) replaces the third term in (4-58), yielding

$$\langle \overline{W} \rangle$$
 (4-110)

$$= \frac{(1-v_c)}{A_{el}} \sum_{i}^{N_{\Theta}} \sum_{j}^{N_{\Phi}} f_{sc}(\Phi_j, \Theta_i) W_{8ch}(I_1^A(\lambda, \Phi_j, \Theta_i), \lambda_m, C_1) sin\Phi \Delta \Theta \Delta \Phi$$
$$+ \left(\frac{P_{sat}{}^2 F(E)}{3N_{\Phi_r} v_c \epsilon}\right) \sum_{i=1}^{N_{\Phi_r}} \left[\frac{e^{b_{\Phi_r} cos(\Phi_r - \mu_{\Phi_r})}}{2\pi I_0(b_{\Phi_r})}\right] \left[\frac{1}{\left(\lambda^{-1} \sin^2 \Phi_{r_i} + \lambda^2 \cos^2 \Phi_{r_i}\right)^{3/2}}\right] .$$

The stress differences based on this energy can be calculated in the same manner as in (4-97) and (4-98) by non-dimensionalizing via $H = P_{sat}^2/(C_1\epsilon)$.

To highlight the dependence of $\langle \overline{W} \rangle$ on *E*, we use $F(E) = \langle \widehat{p}_1 \cdot \widehat{p}_2 \rangle - 3 \langle \widehat{p}_1 \cdot \widehat{r} \rangle \langle \widehat{p}_2 \cdot \widehat{r} \rangle$, which combines the terms indirectly related to *E* via the orientation distribution function in Section 4.2. The value of N_{Φ} is set to 50, determined from the convergence study in Section 5.3. The variable λ is solved for by substituting (4-110) into (4-83) and solving for pairs of $[E, \lambda_3]$, which are transformed via (4-103) into $[E, S_{33}]$. Recall that this relationship between *E* and S_{33} was defined earlier as electrostriction. Based on fitting of the eight-chain hyperelastic model in (4-48) to experimental data performed later in this paper, we chose $\lambda_m = 2.5$ for the following model calculations. Consequently, the model will be capable of plotting electrostriction with only a few adjustable parameters, μ_{Φ_r} , and κ^5 .

The electrostriction model can be analyzed by varying the two adjustable parameters in the context of the earlier defined parallel and perpendicular cases. We consider two cases for μ_{Φ_r}

⁵ Although the von Mises distribution is a function of b_{Φ_r} , we redefined the distribution in terms of κ , which is indirectly related to the distribution function via (4-109). Consequently, we consider κ as an adjustable parameter instead of b_{Φ_r} .

that approximate the parallel and perpendicular models: $\mu_{\Phi_r} = 0$ and $\mu_{\Phi_r} = \pi/2$, respectively. For these two cases, the model calculates the electrostrictive response of (4-110) at varying dispersion factors, κ , as plotted in Figure 4-14. Note that the electrostriction is plotted in terms of a normalized strain, $S_{33}^* = S_{33}/S_{max}$, where S_{max} is the maximum obtained strain in all of the electrostriction calculations, and a normalized electric field $E^* = E/E_{sat}$. As $\kappa \to 1/3$, the distribution functions for both cases, $\mu_{\Phi_r} = 0$ and $\mu_{\Phi_r} = \pi/2$, are identically uniform, and thus, the electrostrictive responses of both cases converge at a value as shown in Figure 4-14a, indicating internal consistency of the model. In contrast, as $\kappa \to 0$, the distributions rapidly diverge, and their electrostrictive responses diverge toward separate asymptotes at Figure 4-14 point c and Figure 4-14 point e. Specifically, as $\kappa \to 0$ for $\mu_{\Phi_r} = 0$, the electrostriction significantly increases as the distribution function approaches a singular point, as shown in Figure 4-14 point c. However, as $\kappa \to 0$ for $\mu_{\Phi_r} = \pi/2$, the electrostriction observes a small decrease, shown in Figure 4-14 point e. These results imply that the forces generated by dipoles whose relative positions are parallel to the field-direction dominate the electrostriction response of a material, and to maximize the electrostrictive response of a material, the target microstructure should contain crystalline domains that are aligned parallel to the direction of the field (often the direction of the thickness). However, it should be noted that the model failed to compute pairs of $[E, S_{33}]$ for $\mu_{\Phi_r} = 0$ below $\kappa \approx 0.0005$, shown in Figure 4-14 point c, due to the material failure phenomenon discussed in Section 5.2. Thus, spatially aligning domains to a very high order of $\kappa < 0.001$ may be counterproductive as it may trigger material failure at high field strengths. As a result, perfectly (spatially) aligned domains may not be desired, especially at field strengths above $E^* = 15.$

An anomaly in the energy functions is revealed near Figure 4-14 point d, where the electrostriction for $\mu_{\Phi_r} = \pi/2$ observes a dip near $\kappa \approx 0.05$. This behavior reflects the phenomenon introduced earlier in Section 5.2, where the possibility of a state of orientation

between $0 < \Phi_r < \pi/2$ yielding zero interaction energy was mentioned. Furthermore, due to the relatively weak energy at $\Phi_r = \pi/2$, the zero-energy orientation affects the averaging of the energy more significantly for $\mu_{\Phi_r} = \pi/2$, thus causing the drop in electrostriction at Figure 4-14 point d. Considering this phenomenon, alignments in the diagonals may not be ideal for achieving a high electrostrictive response.

114



Figure 4-14. Comparison of the two extreme cases with PDFs in prediction of electrostriction. As $\kappa \to 1/3$, both PDF-based responses converge toward the discrete network responses, which is why the discrete network models are placed at the low end of the κ range. Due to theoretical material failure, electrostriction could not be determined at field strengths above $E^* = 15$ for $\mu_{\Phi_r} = 0$ as $\kappa \to 0$; as a result, saturation is not observed at (c).

3.2.6 Deriving The Permittivity of the Amorphous Phase

The dipole-dipole interaction energy requires the relative permittivity ε_r of the medium that holds the dipoles. This medium corresponds to the amorphous phase of the material. However, there are no direct methods of measuring the permittivity of the amorphous phase of the EAPs to the author's knowledge. In this section, some indirect approaches to determining the permittivity of the amorphous phase are explored.

A common approach to determining the relative permittivity of the amorphous phase of a semicrystalline polymer is by effective medium theory. However, only one study does this to the author's knowledge, in which the relative permittivity of the amorphous phase of the P(VDF-TrFE) copolymer is determined to be 8 [53]. Since the model presented in this paper is mainly focused on the mechanism behind the P(VDF)-based *terpolymer*, we turn to a different approach presented in Ref [29], in which the authors consider a biphasic polarization model for the relaxor ferroelectric terpolymer. Most importantly, Ref [29] proves the relationship between permittivity and polarization shown in (4-111).

$$\varepsilon_r = \frac{1}{\varepsilon_0} \frac{dP}{dE} + 1 \tag{4-111}$$

By substituting their description of polarization, Ref [29] is able to obtain a relationship between electric field and electric permittivity. Furthermore, the biphasic model in Ref [29] allows the model to differentiate between the contributions of the crystalline and the amorphous regions. Beyond the basic capability of their model, one other observation is made in the paper: the crystalline regions exhibit high nonlinearity by saturation, as more and more dipoles align with the field. On the other hand, the amorphous phase acts as a linear dielectric, by possessing a very high saturation field, E_{sat} , compared to that of the crystalline phase. This brings a key point: as the electric field is further increased, the amorphous region takes over the polarization response of the material and completely dominates once the crystalline regions saturate in alignment. More importantly, this is evident from permittivity plotted versus electric field, where the permittivity levels off at a certain value at very high fields (~400 MV/m). The experimental values for permittivity are obtained from the calculations found in Ref [2], in which the energy density of the material is calculated using the integral in (4-112).

$$W_e = \int EdP \tag{4-112}$$

This integral is calculated for a polarization curve, and the 'effective' dielectric constant, based on a linear dielectric assumption, at each point is determined by the (4-113).

$$W_e = \frac{1}{2}\varepsilon_r \varepsilon_0 E^2 \tag{4-113}$$

However, given a set of polarization data, the following question is raised: how accurate are the permittivity calculations based on the linear dielectric assumption? Furthermore, would a more direct calculation of the slope of the polarization curve based on discretized data points generate different results for the permittivity? To answer these questions, we discretize the polarization data, and evaluate the slope by a simple approximation by interpolation. If the step size in the discretized system is small enough, this generates an accurate approximation of the slope. The derivative approximation is calculated by (4-114).

$$\frac{dP}{dE}\Big|_{i} \approx \frac{P(i+1) - P(i+1)}{E(i+1) - E(i-1)}$$
(4-114)

Each point, i, is the average distance between two discrete data points. Discrete data from literature is extracted by the Web Plot Digitizer App. For plots that present smooth curves, the step size is reduced to a small enough value that can extract a trend based on the statistics of the data. Lastly, the biphasic permittivity model derived in Ref [29] is implemented into a least squares method, where MATLAB's fminsearch command is utilized. The permittivity equation is split into the two components that are associated to the different phases of the material. Since the amorphous phase almost acts as a linear dielectric, it is assumed to be constant for the sake of reducing the number of parameters for optimization. The permittivity model is shown in (4-115).

$$\varepsilon_{r,total} = \frac{1}{\varepsilon_0} 3E_{sat}(\varepsilon_c - \varepsilon_0) \left[-\frac{1}{\sinh^2\left(\frac{E}{E_{sat}}\right)} + \frac{E_{sat}^2}{E^2} \right] + \varepsilon_a$$
(4-115)

In equation (4-115), ε_c is the low field linear permittivity constant for the crystalline region, E_{sat} is the electric field at which the crystalline region begins exhibiting nonlinearity, and

 ε_a is the constant permittivity attributed to the amorphous phase. The parameters ε_c , ε_a , E_{sat} , will be fit to experimental data, and the amorphous phase's permittivity ε_a will be substituted into the dipole-dipole interaction energy as ε_r .

3.3 Results and Discussions of Microstructure-Based Model

This chapter addresses Hypothesis 2 by studying the effectiveness of the microstructurebased model in terms of predicting the behavior of EAPs, as well as its ability to address variability of relative spatial locations of crystalline domains. In this section, the electrostriction model is compared to multiple data sets by first determining material constants from experiments, then fitting the PDF-based network model via adjustment of κ .

3.3.1 Comparisons of network model responses to experimental data

Three materials have been chosen in this study. One of the materials was fabricated and tested by us, while the remaining two material data sets were borrowed from literature. All three materials are P(VDF)-based terpolymers; their processing methods are listed along with their chemical compositions in Table 4-3.

All three terpolymers were synthesized by bulk polymerization. The material fabricated for this study is polyvinylidene-trifluoroethylene-chlorotrifluoroethylene, also known as P(VDF-

TrFE-CTFE) 61.8/30.4/7.8%.

Table 4-3. Terpolymers are listed with chemical composition and processing details. The chemical compositions may differ from the literature because they have been normalized here to add up to 100%.

Polymer	Percent composition	Additional processing
P(VDF-TrFE-CTFE) (Saad et al., 2018)	61.8/30.4/7.8%	Annealed at 120°C for 9 hours
P(VDF-TrFE-CTFE) (Xu et al, 2001)	59.1/31.8/9.1%	Annealed at 100-120°C for 6 hours
P(VDF-TrFE-CFE) (Cheng et al. 2003)	59.6/36.5/3.8%	Annealed at 110°C for an unspecified period



Figure 4-15. The single-phase polarization model is fitted to the polarization responses of three data sets. The fitted parameters P_s and E_s (in legend) are also listed in Table 4-3.

The polarization model in Capsal, et al. (2012), which is based on (4-66), was used to determine the saturation polarization of the material by least squares fitting the polarization model to experimental data from our laboratory and borrowed from literature. Figure 6-6 shows the fitted models to each set of measured data. Results are tabulated in Table 3 and used to determine electrostriction in equations (4-83) and (4-110).

The elastic properties of P(VDF-TrFE-CTFE) 7.8% are determined by fitting the modified eight chain hyperelastic model $W_{8ch}(I_1^A)$ to experimental tensile data conducted in our laboratory (not shown). The energy is fitted for $v_c = 0.36$ (actual crystallinity) and also $v_c = 0$ to study the effects of crystallinity. The energy with zero crystallinity will provide us a measure of an average C_1 , while the corrected energy apportioned to only the amorphous phase will provide us a C_1 value for only the amorphous phase. As shown in Figure 4-16, the best fit for the average and amorphous C_1 constants are 170 MPa and 14.1 MPa, respectively. The amorphous phase's C_1 is more than an order of magnitude lower than the average, which is expected, as the rigidity of the crystallites will significantly increase the average elastic response of the material. Note that the models are fitted to experiments within a small range of λ , because electrostriction often does not exceed 10% strain.

For the other terpolymers, the reported Young's modulus is used and as a result, we employ a simple Neo-Hookean model to model the hyperelastic response of the material. Results are tabulated in Table 4-4 and used to predict electrostriction in equations (4-83) and (4-110).



Figure 4-16. Tensile test data for the CTFE 7.8% terpolymer, and two best-fit models: modified 8-chain hyperelastic model $W_{8ch}(l_1^A)$ with crystallinity $v_c = 0$ and $v_c = 0.36$.

In addition to the saturation polarization, the permittivity of the amorphous phase ε_a is also required for calculating the dipole-dipole interaction energy. Thus, we utilize the biphasic model derived by Capsal et al. (2012), defined in (4-115). This model is fitted to each experimental data (both from our laboratory and borrowed from other studies) to obtain average values of the bulk permittivity $\varepsilon_{r,total}$. The biphasic model assumes that the crystalline phase dominates the material's polarization response at low field strengths of about 0 to 150 MV/m, and eventually the amorphous phase begins to dominate the material's response, behaving similar to a linear dielectric. Thus, we make the assumption that the amorphous phase has a constant permittivity of ε_a , to reduce the number of unknown variables.

$$\varepsilon_{r,total} = \frac{1}{\varepsilon_0} 3E_s(\varepsilon_c - \varepsilon_0) \left[-\frac{1}{\sinh^2\left(\frac{E}{E_s}\right)} + \frac{{E_s}^2}{E^2} \right] + \varepsilon_a$$
(4-116)

All literature data is analyzed by Web Plot Digitizer, which is an online tool that aids in extracting data points on an image. The images are analyzed by a step size of 1 pixel. The relative electric permittivity of a dielectric material is defined as

$$\varepsilon_r = \frac{1}{\varepsilon_0} \frac{dP}{dE} + 1, \tag{4-117}$$

in which ε_r is the relative electric permittivity, *P* is the polarization, and *E* is the electric field. We must determine the permittivity by approximating the derivative of the polarization with respect to the electric field, which can be determine from a set of data as

$$\frac{dP}{dE}\Big|_{i} \approx \frac{P(i+1) - P(i+1)}{E(i+1) - E(i-1)} \text{ (for } i = 1, 2, \dots n_{P}\text{)}.$$
(4-118)

Once we obtain a set of permittivity data, we fit the model to the data by a least squares method to determine ϵ_r . Given known variance in the polarization data for P(VDF-TrFE-CTFE) 7.8%, we fit the model to one standard deviation below, one standard deviation above, and exactly at the average measured values. As a result, we obtain a range of permittivity for the amorphous phase for this material, from 4.0 to 14, with an average of 9.4. Variance in the other materials are studied by exploring the effects of adding weights to the least squares fitting method to prioritize the polarization responses at higher field strengths, which will better reflect the amorphous phase's permittivity. Due to the complexity of extracting the exact value of ϵ_r from experiments, we focused on using approximate (averaged) values of ϵ_r instead of analyzing a spectrum of ϵ_r per material (such an analysis is beyond the scope of this paper). The approximate results are listed in Table 4-4. The relatively low permittivities for the amorphous phase are consistent with the theory that the amorphous phase should have a low dielectric response compared to the crystalline phase.

Table 4-4. Parameters used for the simulations in Figure 15.

	v_c (%)	$C_1(MPa)$	λ_m	Y(GPa)	ϵ_r	$P_s\left(\frac{C}{m^2}\right)$	E_s
						<u>(m²)</u>	(W V/m)
P(VDF-TrFE-CTFE)	36	14.1	2.5	-	9.4	0.0722	38.7
61.8/31.4/7.8%							
P(VDF-TrFE-CFE)	43	-	-	1.1	3.0	0.113	59.1
59.6/36.5/3.8%							
P(VDF-TrFE-CTFE)	56	-	-	0.4	4.6	0.0961	33.2
59.1/31.8/9.1%							

The parameters listed in Table 4-4 are used in the electrostriction model for each material at varying values of κ at $\mu_{\Phi_r} = 0$ and $\mu_{\Phi_r} = \pi/2$. Figure 4-17 shows the model evaluated at $0.025 \le \kappa \le 0.333$ for $\mu_{\Phi_r} = 0$ and at $0.05 \le \kappa \le 0.333$ for $\mu_{\Phi_r} = \pi/2$ compared to data for P(VDF-TrFE-CTFE) 7.8%. The best-fit curve to experiments (by least square errors) is signaled by a dashed line, with the fitted values $\mu_{\Phi_r} = 0$ and $\kappa = 0.111$. A distribution of $f_{\Phi_r}(\Phi_r; \mu_{\Phi_r} = 0, \kappa = 0.111)$ leans towards a narrow distribution of neighboring RVE locations around the $\Phi_r = 0$ (parallel) position. Furthermore, the fitted distribution may be interpreted as

spatial distributions of crystalline domains, and consequently, the distribution represents an implied microstructure characteristic. The implied microstructure the fitted model for CTFE 7.8% RFE polymer contains crystalline domains with relative locations that are to varying degrees parallel with respect to the field, as depicted in Figure 4-17.



Figure 4-17. Comparison of the electrostriction model evaluated at varying distribution parameters with experiments for P(VDF-TrFE-CTFE) 7.8%. Data is from (Erol et al. 2019).



Figure 4-18. Comparison of the electrostriction model evaluated at $0 < \kappa < 1/3$ for $\mu_{\Phi} = 0$ and $0.05 < \kappa < 0.\overline{3}$ for $\mu_{\Phi} = \pi/2$ with experiments for P(VDF-TrFE-CTFE) 9.1%.

A similar fitting procedure is performed for the CTFE 9.1% RFE polymer, and the model is plotted over the same range of distributions in Figure 4-19. For this material, the best-fit distribution is $f_{\Phi_r}(\Phi_r; \mu_{\Phi_r} = 0, \kappa = 0.145)$, indicating a very similar distribution to the best-fit model for the CTFE 7.8% polymer. Similarly, the best-fit model implies that neighboring crystalline domains would be at varying degrees of alignment to the applied-field direction, with most concentrated at the parallel orientation.

It is important to note that the comparisons in Figure 4-17 and Figure 4-19 reveal a best-fit model with a distribution center of $\mu_{\Phi_r} = 0$ and a moderate degree of spatial alignment. Despite their moderately narrow distributions, the best-fit models for the CTFE RFE polymers still show considerable probabilities of spatial arrangements of neighboring crystallites far beyond $\Phi_r = 0$. Conversely, the model comparisons for P(VDF-TrFE-CTFE) 9.1% result in a very narrow distribution of $f_{\Phi_r}(\Phi_r; \mu_{\Phi_r} = 0, \kappa = 0.034)$, which is between the visual depictions of $\kappa = 1/80$ and $\kappa = 1/20$ in Figure 4-18. Such a distribution implies very little variation beyond the parallel arrangement.

Beyond fitting the electrostriction model to experiments, the model may be used predictively to determine microstructures required for desired properties. It is apparent from the model-experiment comparisons (as well as preliminary comparisons of nondimensionalized energies) that spatial distributions centered parallel to the field with very narrow widths generate the largest electrostrictive responses. Consequently, we would prefer to fabricate materials with similar microstructural arrangements. For example, Figure 4-17 implies that, if the predicted microstructure for the tested P(VDF-TrFE-CTFE) 7.8% terpolymer is correct, it is possible to increase the material's electrostrictive response by more than a factor of 2 if we reduce the distribution width to $\kappa = 0.025$ through material processing techniques during fabrication, which would theoretically yield greater than 10% electrostrictive strain at 150 MV/m. Using this method, the model may assist materials scientists to assess the performance envelope of a given set of fabrication methods or achieve target materials properties through tailored processing. Namely, processing methods such as the simultaneous application of multiple fields (e.g. magnetic and electric fields) could be used to manipulate the microstructure to achieve a distribution with a precise μ_{Φ_r} and κ . However, it is important to note that due to a lack of availability of experimental data on spatial arrangements of crystallites within RFE polymer microstructures, it is currently not feasible to validate the model's predictive capabilities. Furthermore, a validation procedure is necessary before the model can be used for targeting desired material properties. Nonetheless, the model's significance remains from its ability to address the microstructural characteristics that would be necessary to predict structure-property relationships.

Although the model fits the experimental data well, especially at high field strengths (above 100 MV/m), some characteristics of the curves are not captured entirely at lower field strengths. For example, there is a relatively high error in fitting the model to CTFE 7.8% at approximately 40 MV/m in Figure 4-17. This is due to a spike in electrostriction observed in experiments; it is hypothesized that these spikes may result from two separate crystalline phases. Based on in situ X-ray diffraction (XRD) data on CTFE 7.8% terpolymer, β -phase has a coercive

field around 40-50 MV/m, which may explain the electrostrictive spike. Additionally, a similar spike is observed for CTFE 9.1% at 50 MV/m in Figure 4-18. The relatively low initial electrostrictive response in the experimental data for CTFE 7.8% (Figure 4-17) may also be due to the resolution of the experimental measurements, which could amplify the spike at 40 MV/m. These spikes are not as clearly observed in the other data sets. Additionally, predicting low-field responses may be improved by adding the effects of dipole-induced fields. However, including dipole-induced fields in a local field calculation would create new computational challenges beyond the scope of this study.



Figure 4-19. Comparison of the electrostriction model evaluated at varying distribution parameters with experiments for P(VDF-TrFE-CFE) 3.8%.

3.4 Conclusions

The microstructure-based model is developed and analyzed such that it could answer Hypothesis 2. The model utilizes an approach which captures the electromechanical coupling within the relaxor ferroelectric terpolymer based on its dipole-dominant microstructure, with only a few adjustable parameters, all of which possess physical significance. A free energy function composed of both an electrostatic and elastic term is implemented, where the electrostatic component is the interaction energy of dipoles (Coulomb), which are prevalent in the microstructure of the terpolymer and a direct mechanism behind the nonlinear electromechanical coupling behavior. A probability distribution function for both the spatial orientation of polar regions and the orientation of their dipole moments provides a more realistic picture of the microstructure—as it is beyond just two points of crystal regions connected via an elastic chain. Fitting the model to experimental results by varying these PDF parameters provides a better grasp of the orientations of crystal regions with respect to each other, and this yields a framework for more detailed studies on the actual shape and sizes of these domains.

The modeling results showed that the anisotropy of the dipole-dipole interactions influences the electromechanical coupling of the system. Namely, arrangements of crystalline regions parallel to the field can generate the largest electromechanical strain, while perpendicular arrangements generate about an order of magnitude less.

Additionally, the model can capture a phenomenon known as pull-in instability, which can either cause material failure or be harvested to yield larger electromechanical coupling depending on boundary conditions. Parallel arrangements were found to be prone to induce pullin instability, and the exact strength of the field required for the instability can be predicted by the model. With this information, dipole-based EAPs can be fabricated and processed to either promote or inhibit pull-in instability by manipulating the arrangements of the dipolar regions.

To improve the modeling methods, multiple spatial means (averages) may be implemented into the PDF for spatial orientations, resembling network models such as the threechain or eight-chain model. In addition, crystal regions may be modeled as aggregates of single dipolar molecules or structures, with a PDF dependent on a third variable, r, to be implemented in a multi-scale framework. In such a model, exact domain sizes, shapes, and other structural properties will be directly employed for a more accurate picture of the microstructure of EAPs. In addition to these expansions of the model, the model can be implemented for materials outside the scope of EAPs, for other dipole-based active materials such as magneto-active [80].

CHAPTER 4

Link Between Electrostrictive Material Response and Structural Deformation: Optimization of a Multi-Field Actuated Multi-Layered and Segmented Beam

This Chapter focuses on the third hypothesis, which states that an analytical model for multilayer, segmented composite beams incorporating the EAP constitutive model and magnetic torques can accurately predict multi-field device response, and can be used for the design optimization for a target shape containing ideal folds. To answer the hypothesis, two objectives are completed.

Objective 3.1: Derive an analytical model for the bending of an arbitrarily segmented, multilayered, multi-field actuating composite beam that accounts for large displacements, dipole-based electrostrictive constitutive relations of EAPs, and magnetic dipoles of MAEs.

Objective 3.2: Define and execute a multi-objective design optimization problem on a case study with preset material types and fixed number of segments for work, shape, and cost. The shape function must contain ideal folds, and the parameters must include segment lengths and the Young's modulus of at least one material. The results of the multi-objective optimization will determine how well folding can be achieved by segment length variation.

Note: This section is the author's 2016 SMASIS conference proceeding (Erol et al. 2016) and article paper submitted to a special issue of Smart Materials and Structures.

4.1 Introduction to Self-Actuated Origami-Inspired Devices

This section provides an introduction to the topics related to addressing Hypothesis 3, including Origami-inspired engineering, self-folding structures, compliant mechanisms, magnetoand electro-active structures, multi-field actuated structures, and specifically multi-field actuated bimorphs. Each of these areas provides useful insight into how the new methodologies can incorporate state-of-the-art materials into a new framework for the optimization of a self-folding structure utilizing two or more types of active materials. While the introductions in Chapters 2 and 3 provide the groundwork for modeling and engineering materials, this introduction focuses on a background for the implementation of those materials in structures for specific purposes, such as self-folding Origami-inspired structures.

4.1.1 Self-Folding Structures

Designs of foldable structures in engineering have drawn inspiration from origami, the Japanese art of folding paper, which offers fold patterns that have been employed for centuries (Schenk and Guest 2011; Peraza-Hernandez et al. 2014; Onal et al. 2015). Some examples include deformable wheels (Lee et al. 2013; Yu She, Hurd, and Su 2015), locomotive worm devices (Onal, Wood, and Rus 2013), reconfigurable metamaterials (Filipov, Tachi, and Paulino 2015), pneumatic actuators (Martinez et al. 2012), and robotic tentacles with 3D mobility (Martinez et al. 2013).

Recent advances in smart materials and their applications have focused attention toward developing self-folding structures (Stoychev, Puretskiy, and Ionov 2011; S. M. Felton et al. 2013; S. Felton et al. 2014). Several types of origami-inspired self-folding structures have been made in the past, including those utilizing the Miura-ori (Cowan 2015) and Waterbomb (Bowen et al. 2015) patterns, origami cranes (S. M. Felton et al. 2013), barking dog (Bowen et al. 2017) and an origami inspired forceps (Edmondson et al. 2013).

4.1.2 Compliant Mechanisms

Some smart devices utilize materials that are relatively compliant, i.e,. they have a relatively small Young's modulus, such as that of rubbers and elastomers (Kota and Ananthasuresh 1995; Howell 2001; Bruns and Tortorelli 2001). A compliant material has both advantages and disadvantages in the context of self-folding structures. Folding structures typically possess stiff facets that experience little to no deformations, and thus any compliance would be undesirable in those regions/parts. Meanwhile, the creases of a foldable structure exhibit very large deformations, and can only be achieved by a relatively compliant material. Hence, folds are limited to finite

curvatures, which has led to studying smooth folds in compliant structures (Peraza Hernandez 2016).

Compliant mechanisms have been combined with active self-folding concepts for achieving a wide range of applications (S. M. Felton et al. 2013; Saggere and Kota 1999; Frecker 2003; Crivaro et al. 2016; Calogero et al. 2017). Some potential applications may seek a complex deformed shape upon actuation, i.e. target shape, that may not consist entirely of creases and flat surfaces, and instead possess regions of both flat and curved surfaces. Thus, materials like elastomers and polymers whose mechanical properties can be finely tuned are ideal candidates for foldable structures.

4.1.3 Magneto-Active Structures

Hard magnets have been used to actuate self-folding because the magnetic torque is strong enough to generate large displacements, especially in compliant mechanisms(S. M. Felton et al. 2013; Edmondson et al. 2013; Calogero et al. 2017). Magnetic torque is possible due to the anisotropic magnetic properties of hard magnets, which have a preferred magnetic direction determined by their magnetization. The net magnetization M of a material generates a net torque τ under an external magnetic field H following:

$$\boldsymbol{\tau} = \boldsymbol{M} \times \boldsymbol{H} \,. \tag{5-1}$$

The magnetic torque is shown schematically in Figure 5-1, where a magnetically susceptible body (red rectangle) is subjected to an external magnetic field (wide blue arrows) applied perpendicular to the direction of the body's net magnetization (red arrows). Consequently, an induced torque tends to rotate a body with volume V to minimize the Zeeman energy, E_{Zeeman} , by aligning **M** with **H**.

$$E_{Zeeman} = -\mu_0 \int_V \boldsymbol{M} \cdot \boldsymbol{H} dV \tag{5-2}$$

Magneto-active elastomers (MAEs) are a suitable class of magnetically actuated smart materials for self-folding structures due to their controllable magnetic properties and ease of fabrication into desired shapes. In recent work, MAEs have been fabricated using barium hexaferrite (BAM) particles embedded inside an elastomeric substrate while prescribing magnetic volume fraction and magnetic alignment direction during curing to control magnetic properties (Von Lockette, Kadlowec, and Koo 2006; Breznak and Lockette 2016).



Figure 5-1. A magneto-active body is represented by a rectangle in (a), with its magnetization M indicated by the red arrows. An external field H induces a torque T in the body, as shown in (b).

The body's Zeeman energy is minimized by aligning M with H as shown in (c).

4.1.4 Electro-Active Structures

Electro-active polymers (EAPs) have also been deployed in active self-folding structures due to their ability to generate large displacements upon actuation (Ahmed et al. 2015; Ahmed, Ounaies, and Arrojado 2017; Ahmed, Arrojado, and Ounaies 2016). Electrostrictive EAPs yield relatively high strains as they contract in the direction of the applied electric field and expand in the transverse directions. The actuation of EAPs in self-folding structures relies on at least one secondary layer of passive material attached to the EAP, which will constrain the deformation on one surface, inducing bending in the overall composite structure.



Figure 5-2. Bimorph composed of an EAP and MAE layer. Magnetic actuation yields bending towards the left, while electric actuation yields bending towards the right.

4.1.5 Multi-Field Actuated Structures

In this work, we seek to combine magneto- and electro-active materials as possible layers and /or segments of a multifunctional structure. The combination of magneto- and electro-active materials in an active self-folding structure has been studied recently, and has several advantages (Frecker 2003). Two different actuation mechanisms enable more possible deformed configurations, e.g. multiple target shapes, as shown in Figure 5-2. The example in Figure 5-2 shows a bimorph actuator, which consists of an EAP and an MAE. The initial shape of the beam is straight, as shown in Figure 5-2.a. With magnetic actuation the beam bends in one direction (Figure 5-2.b), while with electric actuation the beam bends in the opposite direction (Figure 5-2.c). When both fields are present simultaneously, however, the beam deforms into a more complex shape, highlighting the potential of multi-field actuation.

The purpose of this study is to understand how the choice of active material, configuration of that active material, device compliance, and layer geometries of a multi-field actuated device may influence its self-folding behavior. This goal is achieved by developing a model of the device's response, and then exploring the performance space associated with design variables within the model. The next section develops the geometry for our self-folding compliant beam structure consisting of two possible active materials, device geometry in terms of layer thickness and length,
and active material placement in the multi-layered, segmented structure. Next, the constraints of the design are determined based on manufacturing considerations, and the parameters are determined based on influence on self-folding actuation. Objective functions are next defined followed by implementation of a previously validated modeling approach to predict the multi-field actuation of the structure. Results of simulations across the design space are explored and analyzed in terms of the objective functions, allowing us to assess the efficacy of the objective functions and the relative significance of our chosen parameters.

4.1.6 Multi-field Bimorph Design

Multi-field bimorph actuators have been studied in active self-folding applications due to their ability to generate complex shapes (Erol et al. 2016; Sung et al. 2016; W. Zhang, Ahmed, et al. 2018). Consequently, they will be utilized as model segments in determining the structure for this study. In its most basic form, the multi-field bimorph consists of two layers, an MAE and EAP layer, as shown in Figure 5-2. In this study, we will examine a more complex version of this basic bimorph by (1) dividing the MAE into separate, individual patches with alternating orientations, and (2) adding a passive layer. The segmentation of the MAE layer allows us to add a level of complexity to the deformed shape, and the ability to control the amount of magnetic material by varying patch sizes. The separation of the MAE layer into individual patches introduces the concept of a segmented beam. The passive layer is added to provide support to the segments lacking MAEs, while also constraining the EAP layer to promote bending in those segments.

4.2 Modeling Multi-Layer Multi-Segment Composites Methodology

To perform a formal design optimization of a multi-field, segmented self-folding structure, a modeling approach with appropriate functions must be chosen. The chosen model should be able to accommodate large displacements, electro- and magneto-mechanical coupling, multi-layering, and segmentation. This section covers a model that can address each function and be used for an arbitrary optimization problem.

4.2.1 Model Framework

An existing modeling technique previously derived and validated by the authors is selected as a framework for predicting the deformed shape of the configurations allowing computation of the objective functions (Erol et al. 2016). The model is based on a nonlinear, large displacement beam method for a composite beam of an arbitrary number of segments N_S , each composed of an arbitrary number of layers N_{iL} , as shown in Figure 3.a. The indices *i* and *j* represent the segment and layer locations, respectively. Thus, any segment-layer coordinate along the length of the beam may contain a set of material and geometric properties, S_{ij} .

Let the beam of length *L* be discretized into *p* nodes. Consider a differential element between two points along the beam, n - 1 and n + 1, such that the element contains three nodes. This type of three-node element is chosen for the convenience it provides when describing each element curvature by the curvature at node *n*, which falls at the center of the element. The governing equations can be written with two equilibrium equations for the differential element,

$$\sum_{i=1}^{N} F_{x,ij} = 0, (5-3)$$

$$\sum_{i=1}^{N} M = 0, (5-4)$$

where $F_{x,ij}$ is the force in the direction of the beam's deformed neutral axis at segment *i* and layer *j*. The forces can be substituted with the integrals of the stresses over the surface of each cut, such that the sum of forces and moments, respectively, can be expressed as

$$\sum_{j=1}^{N_{iL}} \left[\int_{A_{ij}} \sigma_{n+1,ij} dA_{ij} + F_{N,n+1} \right] - \sum_{j=1}^{N_{iL}} \left[\int_{A_{ij}} \sigma_{n-1,ij} dA_{ij} \right] = 0,$$
(5-5)

$$\sum_{j=1}^{N_{iL}} \left[\int_{A_j} \sigma_{n+1,ij} z dA_{ij} + 2V_{n+1} \Delta x \right] + \sum_{j=1}^{N_{iL}} \left[-\int_{A_j} \sigma_{n-1,ij} z dA_{ij} \right] + \sum_{j=1}^{N_{iL}} \left[2\tau_{n,ij}(H) t_{MAE} w \Delta x \right] = 0,$$
(5-6)

where
$$\sigma_{n,ij}$$
 are the stresses in the *x* direction at node *n*; A_{ij} is the cross-sectional area; Δx is the
length between two nodes; $\tau_{n,ij}(H)$ is a magnetic torque dependent on magnetic field strength; *w*
is the width in the *y* direction; V_{n+1} is a shear force; $F_{N,n+1}$ is the normal force; and *z* is the distance

in the direction of the thickness, as defined in Figure 5-3.b. The indices *i* and *j* refer to segment and layer locations, respectively. Each summation from j = 1 (layer 1) to $j = N_{iL}$ is shown separately to emphasize that the number of layers N_{iL} may be different for each node, n, n - 1 and n + 1.





can have an arbitrary material type, denoted by the color code (the colors indicate different material types, which can be any type). In (b), a differential element of the composite beam at an arbitrary location along its length is provided, with cut faces subject to normal stresses and shear forces.

The following constitutive equation is used for stress,

$$\sigma_{n,ij} = Y_{ij}(\varepsilon_n + \varepsilon_{ij}^e(E)), \tag{5-7}$$

in which ε_n is the elastic strain at node *n*, defined by beam kinematics,

$$\varepsilon_j = -K(z+\bar{z}). \tag{5-8}$$

The variable *K* is the curvature, and \bar{z} is the distance to the neutral axis. Meanwhile, $\varepsilon_{ij}^e(E)$ is the electrostrictive strain as a function of an applied electric field *E*, determined from a microstructure-based electrostriction model for a nonlinear EAPs (Erol et al. 2019). The model is based on averaging the strain-energy density of a semicrystalline microstructure, consisting of amorphous regions that behave like a hyperelastic material, and crystalline regions that behave like

dipoles interacting with each other. The generalized form of the strain energy density for a semicrystalline EAP is

$$\widetilde{W} = \frac{(1 - v_c)}{A_{el}} \sum_{m}^{N_{\Theta}} \sum_{n}^{N_{\Phi}} f_{sc}(\Phi_n, \Theta_m) W_{8ch}(\Phi_n, \Theta_m, \lambda_m, C_1) \sin \Phi \Delta \Theta \Delta \Phi$$

$$+ \frac{P_{sat}^2}{3v_c \epsilon (\lambda^{-1} \sin^2 \Phi_r + \lambda^2 \cos^2 \Phi_r)^{\frac{3}{2}}} [\langle \widehat{\mathbf{p}}_1 \cdot \widehat{\mathbf{p}}_2 \rangle - 3 \langle \widehat{\mathbf{p}}_1 \cdot \widehat{\mathbf{r}} \rangle \langle \widehat{\mathbf{p}}_2 \cdot \widehat{\mathbf{r}} \rangle]$$
(5-9)

where Φ_n and Θ_m are microstructural parameters describing spatial orientations of crystalline regions, λ_m and C_1 are elastic constants for the 8-chain hypelastic model W_{8ch} (Arruda and Boyce 1993), f_{sc} is a probability density function, \hat{p}_1 and \hat{p}_2 are the unit vectors of two neighboring dipole moments separated by a vector whose unit vector is \hat{r} , v_c is the volume fraction of the crystalline regions, λ is the macroscopic stretch, A_{el} is a constant of integration, P_{sat} is the saturation polarization, and ϵ is the permittivity of the amorphous phase. All constants are grouped into a set Γ_{ij} , which will serve as a parameter set for electrostrictive properties of materials at each layer j of each segment i. The constitutive relation between the strain-energy density \tilde{W} and the Cauchy stress tensor T is given by

$$\boldsymbol{T} = \frac{2}{J} \frac{\partial W}{\partial \boldsymbol{B}} \boldsymbol{B} + q \boldsymbol{I}, \tag{5-10}$$

where q is a Lagrange multiplier to enforce an incompressibility constraint, I is the identity tensor, and $B = F^T F$, in which F is the deformation gradient tensor. The interactions between the dipoles changes as a function of the external field via the alignments of \hat{p}_1 and \hat{p}_2 , which shifts the equilibrium between the stresses generated from each phase. Finding the strain ε^e at which equilibrium is satisfied for each field strength is how the electromechanical coupling $\varepsilon_{ij}^e(E)$ is determined. The magnetic torque at each location, $\tau_{n,ij}$, is determined by the amount of magnetic material is present in the volume of magneto-active layer at each location *i*, *j*. The torque is a function of the relative angle of the point *n*, such that

$$\tau_{n,ij} = V_{\Delta} \boldsymbol{M}_{n,ij} \times \boldsymbol{H}, \tag{5-11}$$

where $M_{n,ij}$ is the magnetization of the material at n, $V_{\Delta} = \Delta x t_{ij} w$ is the volume of the magnetic layer in the differential element, and H is the externally applied magnetic field.

If a force F_{tip} is applied at one end of the beam, then the shear term V will be non-zero. Since the model is assuming large displacements, V cannot be assumed constant at each node. While this does not strictly satisfy equilibrium for each element, the difference between V_{n-1} and V_{n+1} can be assumed to reach 0 as $\Delta x \rightarrow 0$. Thus, if $\Delta x \ll L$, then the equilibrium can be approximated by (5-5) and (5-6).

The value of *V* at any node is a function of the deformation, as it will change based on the orientation of the element. For instance, if the beam is fixed on one end and F_{tip} is applied on the other end, which is free, then upon actuation, the beam will experience large deformations, and the shear force will change at each location depending on the orientation of the element. The orientations of the elements are dependent on the curvatures of the element and all those prior to it (starting from the fixed end, since it is more convenient to apply the boundary condition this way). Similar arguments apply for the normal force, F_N . The normal and shear forces can be defined by

$$\boldsymbol{F}_{N,n} = F_{tip,x} sin\theta_n + F_{tip,y} cos\theta_n, \tag{5-12}$$

$$\boldsymbol{V}_{\boldsymbol{n}} = F_{tip,x} cos\theta_{\boldsymbol{n}} + F_{tip,y} sin\theta_{\boldsymbol{n}},$$
(5-13)

where θ_n is the angle of the element relative to the *x*-coordinate (counter-clockwise is positive). The angle and the shear and normal forces are illustrated in Figure 5-4.



Figure 5-4. A tip force is applied to one end of the beam. The beam is cut at a location to show the angle of the location and the normal and shear forces acting at the cut surface.

The governing equations are written for every possible three-node element across the beam, such that a system of equations can be written,

$$\boldsymbol{C}_{mn}\boldsymbol{K}_{n} = \boldsymbol{f}_{n}, \tag{5-14}$$

where K_n are the unknown curvatures at each node, $C_{mn} = C_{mn}(t_{ij}, \epsilon_{ij}, Y_{ij}, L, w)$ is a stiffness matrix, $f_n = f_n(K, H, F_{tip})$ are the forcing terms. After solving for the curvatures, the curvatures can be transformed into coordinates in the deformed configuration, x_n , by a rotation matrix R_{mn} in the relation $x_n = R_{mn}K_n$. Details of the model can be found in (Erol et al. 2016).

Due to the nonlinearity of the problem, the model is solved by an iterative technique that utilizes the previous solution to determine the next solution at every step. At each iteration, the deformed shape solutions for given parameter sets S are calculated in MATLAB via the vpasolve numerical solver, and solutions of previous field increments are used as an initial guess for the next increment's solution. Between each iteration, the field strengths are incrementally increased to allow the solver to find the next solution. Thus, different sequences of electric or magnetic actuation can be simulated by changing the field strengths at each iteration in the order of our choosing. The order in which the fields are applied in multi-field actuation is significant because it is a path-dependent problem. A structure with the same configuration may yield different deformed shapes if the order of actuation is reversed. Multi-field actuation in experiments is typically conducted with the magnetic field applied first and held constant, while the electric field is increased afterwards (to

avoid prolonged electric actuation that may result in dielectric breakdown). As such, in the simulation the magnetic field is ramped first until it reaches its maximum, then while holding the magnetic field constant, the electric field is ramped until it also reaches its maximum value.

4.2.2 Model Validation for Basic Configurations

To validate the model prior to optimization, the model was compared to a few simplified configurations. The first configuration is a unimorph (a beam with one active layer, and one passive layer) composed of a P(VDF)-based EAP and Scotch tape as the constraining layer. This setup was tested experimentally under just an electric field, and its resultant tip displacement was measured as shown in Figure 5-5.

Each sample was fixed on one end as a cantilever beam. For electric actuation, the strips of electrodes were placed on both sides of the terpolymer, and copper wires were attached on each side. The copper wires were connected to a voltage generator and amplifier. The function generator was operated at a DC voltage varying the amplitudes from 0 to 3 kV to produce the through-thickness electric field, which was calculated as E = V/t, where t is the thickness of the terpolymer and V is the applied voltage. For magnetic actuation, the setup was placed between two poles of an electromagnet. A power supply was connected to the electromagnet to produce a current-induced magnetic field. The unimorph was actuated up to 62.5 MV/m.



Figure 5-5. Resultant, R, is measured as the tip displacement from before and after actuation.

The model for the unimorph case is assumed to be a single segment containing two layers: one layer EAP and one layer passive material. The beam is discretized into 200 nodes for the numerical solution. The results are presented in Figure 5-6, which shows good agreement between the model and experimental data, especially at higher field strengths where the larger displacements could be more accurately measured. The model used the same dimensions of the unimorph and the material properties of the EAP and Scotch tape, listed in Table 5-1.



Figure 5-6. Experimental and modeling results for the EAP-based (terpolymer) unimorph. The figures in (a) and (b) present a deformed shape comparison at 60 MV/m, while (c) is the layer configuration of the unimorph, and (d) shows the tip displacement results. R is the resultant tip displacement and L is the original length when the beam is flat.

Table 5-1. Material parameters.

	MAE	Terpolymer	Scotch tape
E (MPa)	1.5	200	1600
M(A/m)	50,000	0	0
$\alpha\left(\frac{C}{V^2m}\right)$	0	3×10^{-18}	0

Layer height (µm)	500	40	60
Width (cm)	2	2	2

The second case used to validate the model is the bimorph, which contains two active material layers. The bimorph contains an MAE layer with 0.5 mm thickness attached to the EAP, and the field strength was tested up to 80 MV/m. Since the EAP exhibits a nonlinear dielectric response, a nonlinear method must be implemented. The constitutive model from Part II is employed for the relationship between electrostrictive strain and the electric field.



Figure 5-7. Model and experiments compared for the bimorph with two separate fields turned on. (a) compares the tip displacements for the electrical actuation of the bimorph while (b) is for the magnetic actuation.

The bimorph was tested under two separate fields. The top layer was the terpolymer and the bottom layer was the MAE. An MAE magnetization of M = 50000 A/m (approximately 0.05 Tesla), determined using vibrating sample magnetometry experiments, was used. Figure 5-7.a shows the

electrical response of the bimorph sample and the model. Based on the results, the nonlinear electrostriction is better at predicting the tip displacement of the bimorph. Therefore, the linear electrostrictive coefficient is not suitable for the model of this structure. It is likely that this deviation from experimental behaviors is emphasized more so in the bimorph than in the unimorph because of the different constraining layer, and it should also be noted again that the comparisons of the unimorph were only performed up to 62.5 MV/m. On the other hand, Figure 5-7.b shows the magnetic response of the bimorph and the model, which shows good agreement as well.

After the validation of the model for separate actuation cases, the combined effects of the external fields are investigated via multi-field simulations. These effects, as shown in Figure 5-8, produce more interesting results that could further provide information on how to construct the bimorph and which combinations of field strengths are required for a target shapes to be achieved.

Figure 5-8.a shows that for a fixed electric field, varying the magnetic field can change the structure's orientation significantly. For zero magnetic field, the structure's tip ends in the first quadrant, for H=20,000 A/m it ends in the second quadrant, and for H=-20000 A/m it ends in the fourth quadrant. The transition of shape from H=0 to H=-10,000 A/m also suggests that a *neutrally enforced* flat state, balancing magnetic and electric actuation is also possible. Figure 5-8.b also shows how the structure can change shape for a fixed magnetic field while varying the electric field.



Figure 5-8. Simulations of the bimorph under (a) constant electric field with varying magnetic fields, and (b) constant magnetic field with varying electric fields.

4.2.3 Objective Functions

When considering the performance of a self-folding structure, a key metric is its ability to match the actuated shape it is designed to achieve. The target shape is defined in terms of a set of four panels and three folds, as shown in Figure 5-9. To examine how closely the structure can reach ideal folded shape within the chosen design space, the fold angles are used. For example, all fold angles in the target shape, as defined in Figure 5-9, are set to 60 degrees.

The shape error objective function is computed in terms of the difference between the target shape's coordinates and the calculated coordinates under multi-field actuation at discrete points along the beam. Shape matching was originally proposed for rigid link mechanisms, and later applied to compliant mechanisms (Jovanova et al. 2016). Thus, it is an adequate measure of the self-folding structure's actuation capabilities.

$$f_{shape} = \sum_{i=1}^{N} \frac{\left(x_{model,i} - x_{target,i}\right)^2}{L^2}$$
(15)

The target shapes coordinates are defined as $x_{target,i}$, and the simulated coordinates are $x_{model,i}$ for i = 1, 2, ..., N, where N is the number of discrete points along the beam's neutral axis (note the simulation methods yields the deformation along the beams' neutral axis). The differences between the locations of corresponding points on the simulated and target shape are divided by L^2 (*L* is the total length of the beam) to normalize the error.



Figure 5-9. Target shape drawn as a line through the structure's neutral axis. Segments are identified between points along the shape's path.

In terms of design, the cost of a self-folding structure is another important measure when evaluating the feasibility of the design. Two of our structural parameters, the length ratio and magnetization of the MAEs, directly influence how much magnetic material is present in the structure, and the magnetic material, BAM, is by far the most expensive material at approximately \$8,000 per kg for nanoparticles (Sigma Aldrich). The magnetic cost of a design is defined as in Equation 8 which depends on the total volume of BHF across all MAE patches, V_{MAE} , a volume percentage, v, and the cost per volume, c.

$$f_{cost} = c * v * V_{MAE} \tag{5-15}$$

The total volume of the MAEs is dependent on the length parameter,

$$V_{MAE} = 4 * w * l_{MAE} * t_{MAE}.$$
(5-16)

where $l_{MAE} = l_{even}$ is the length of the MAE patch, and t_{MAE} is the thickness of the MAE patch.

In addition, a third objective function is considered for the design optimization problem: the work performed by the structure on a force applied at the tip. This objective function is a significant addition to the optimization because (i) it is a metric of performance that can be directly applied to a gripper-like application (Shian, Bertoldi, and Clarke 2015; W. Zhang, Hong, et al. 2018), and (ii) it has not been previously included in an optimization for a multi-field actuated device.

The objective function for work is defined as

$$f_{work} = \int_0^s \boldsymbol{F_{tip}} \cdot \boldsymbol{dr}, \tag{5-17}$$

where F_{tip} is the applied tip force, dr is the incremental displacement of the tip, and *s* is the path traveled by the tip.



Figure 5-10. The location of the tip force is shown on the structure schematic.

4.2.4 Formal Optimization Setup

The results of the parameter study helped determine that each parameter influences the deformed shapes significantly. However, while the best designs contained a wide range of magnetizations and length ratios, the Young's modulus was consistently low. Consequently, the Young's modulus will be kept constant for the formal optimization. Additionally, a gap between the MAE patches was another common feature among the best designs, which means the optimization algorithm would benefit from a gap constraint.

The formal optimization problem is defined as:

Minimize: $f_{shape}, f_{cost}, f_{work}$

Subject to: $C_{mn}K_n = f_n$

$$x_n = R_{mn}K_n$$
$$0 \le M_k \le M_{max}$$
$$\sum_{i=1}^8 l_i = 1$$
$$l_i > 0$$

where the variables are

$$\mathbf{S} = \{N_S, N_{iL}, M_{ij}, t_{ij}, \epsilon_{ij}, Y_{ij}, L, w, H, E\}.$$
(5-18)

Two constraints are added to ensure that segment length fractions, l_i , remain positive ($l_i > 0$) and sum to unity ($\sum_{i=1}^{8} l_i = 1$).

For the optimization, a genetic algorithm developed by MATLAB, called *gamultiobj*, is chosen. This algorithm is a variant of the multiobjective genetic algorithm NSGA-II, and it well suited for problems with a relatively small number of variables. More details can be found on the base algorithm in the original publication (Deb et al. 2002). The genetic algorithm options are held at default except for the population size set to 100.

4.2.5 Case Study for Parameter Study and Optimization

As a case study, consider a beam composed of a layer of EAP and a layer of passive material stretching along the entire length of the beam, with four MAE patches that are placed as shown in Figure 5-11. Due to the MAE placements, and their magnetization orientations, the beam is divided into eight segments. The four-MAE-patch configuration allows the EAP layers in the two-layer segments to generate bending that can cooperate with the MAEs in folding the structure forming the so-called 'accordion' bending configuration (a double humped 'M' shape).



Figure 5-11. Schematic of a simple bimorph composed of eight segments with *j* layers, fixed on the left end. An external magnetic field *H* is applied upward. Layer 1 is an EAP (green); layer 2 is a passive material (blue); even numbered segments contain a layer 3, which is an MAE (magnetization directions signaled by yellow arrows). Dimensions are not to scale.

The direction of the force, F_{tip} , is assumed constant, and always in the x-direction.

Thus, f_{work} can be simplified to

$$f_{work} = F_{tip}\partial_x,\tag{5-19}$$

where ∂_x is the tip-displacement in the x-direction.

4.2.6 Chosen Parameters

In this subsection, the system parameters are explored such that the optimization problem can be narrowed by utilizing only a few key parameters, which will save computational costs.

Prior to any reductions to the parameters, consider the complete set of model parameters that describe the geometry, materials, and external stimuli, which are:

$$\boldsymbol{S} = \{M_{ij}, t_{ij}, \epsilon_{ij}, Y_{ij}, L, w, H, E\}.$$
(5-20)

The parameters with subscripts are associated with layer j at ith segment: M_{ij} are the magnetizations; t_{ij} are the layer thicknesses; ϵ_{ij} are the electrostrictive strains; and Y_{ij} are the Young's moduli. The parameters L and w are the length and width of the structure, respectively. The magnetic and electric fields are H and E, respectively.

In the generalized form, the structure must contain geometric and material information for every segment and layer, such that there are N_S segments with lengths l_i , yielding a total device length $L = \sum_{i=1}^{N_S} l_i$. Total length L is kept constant, allowing focus on the relative lengths, l_i . All potential geometric and material parameters are listed in Table 5-2.

Since EAPs are difficult to manufacture, and their properties are difficult to fine-tune, the EAP layers' properties will be held constant for all segments present, resulting in $\epsilon_{ij} = \epsilon$ when j = 1 and $\epsilon_{ij} = 0$ otherwise.

We assume the width transverse to all layers and segments is constant, w, and all applied fields, E and H, are held constant globally (i.e., no spatial variation in either field).

The subset of parameters for the case study are reduced to

$$\mathbf{S} = \{M_2, M_4, M_6, M_6, l_1, l_2, l_3, l_4, l_5, l_6, l_7, l_8, Y_P\}.$$
(5-21)

where Y_p is the Young's modulus of the passive layer for all segments (i.e., $Y_{i2} = Y_p$ for all *i*), M_i are the magnetizations of each MAE patch (*i* representing segment number), and l_i are the segment lengths. The layer indices, *j*, are dropped for the case study due to the predefined layer locations of each material (i.e., the passive layer is always at *j* = 1, EAPs at *j* = 2, and MAEs at *j* = 3).

These parameters are chosen due to their potentially large influences on the actuation of the structure, and consequently the objective functions.

For instance, one of the purposes of this study is to determine how compliance can affect the deformation and folding of a smart structure, making the Young's modulus of the materials an important variable. The Young's modulus of at least one of the materials is thus considered, and since varying the Young's modulus of an active material may result in nonlinear influences on its actuation properties, the passive layer's Young's modulus is selected as a parameter.

Another method of varying the bending stiffness along the length of the structure is by changing the lengths of the segments, which have different stiffnesses depending on the number of layers.

Beyond the stiffness of the structure, magnetization was considered as the third parameter, since the magnetization of an MAE is controllable during fabrication by adjusting the volume content of magnetic material of known magnetization response (Breznak and Lockette 2019). Magnetization also influences both the actuation capability (see eq. 1), and the cost of the structure.

4.2.7 Parameter Study Formulation and Results

A preliminary parametric study was performed prior to the optimization to understand the viability and usefulness of the chosen objective functions and a few key parameters (Erol, Lockette, and Frecker 2018). The parameter study focused on two objective functions, f_{shape} and f_{cost} , and considered the effects of three parameters, the magnitude of the magnetization of the MAE patches, M, the Young's modulus of the passive layer, Y_p , and the ratio of the lengths of segments with the MAE patches to those without MAE patches, $\Upsilon = l_{odd}/l_{even}$. The reason for reducing the length parameters from all l_i to a ratio of the lengths of the odd segments to even segments is because all eight l_i was unwieldy for the parametric study, and creating a length ratio for varying the lengths produces a single parameter that affects all segment lengths. The segment lengths were thus divided into two categories: odd and even numbered segments. A length ratio Υ

was defined as $\Upsilon = l_{odd}/l_{even}$, where l_i are the fraction of each segment length with respect to the total length of the structure.

The tip force was F = 0, since f_{work} was not included in the study. The set of parameters for the parametric study can be shown as,

$$S^* = \{M, \Upsilon, Y_{i2} = Y_P\},\tag{5-22}$$

where Y_P is the Young's modulus of the passive layer for all segments (i.e., $Y_{i2} = Y_P$ for all *i*), Y is the length ratio of each segment with respect to each other. To generate symmetric torques, M_{ij} are held constant as well, hence $M_{ij} = M$ for all even segments (i = 2,4,6,8), at j = 3, and $M_{ij} =$ 0 for all others.

Table 5-2. All possible geometric quantities and material properties for all layers in each segment for the chosen four MAE patch bimorph configuration.

	Geometric Quantities			Material Properties			
	Segment	L	Layer Thickness		Lavor i –	Laver i –	Lavor
	Lengui	Layer $j = 1$	Layer $j = 2$	Layer $j = 3$	1	2	j = 3
Segment $i = 1$	lı	t_{EAP}	t_P	-	Γ, Y_{EAP}	Y _P	_
Segment $i = 2$	l ₂	t _{EAP}	t_P	t _{MAE}	Γ, Y_{EAP}	Y _P	M, Y_{MAE}
Segment $i = 3$	l ₃	t _{EAP}	t_P	-	Γ, Y_{EAP}	Y _P	_
Segment $i = 4$	l_4	t _{EAP}	t_P	t _{MAE}	Γ, Y_{EAP}	Y _P	M, Y_{MAE}
Segment $i = 5$	l_5	t _{EAP}	t_P	-	Γ, Y_{EAP}	Y _P	_
Segment $i = 6$	l ₆	t _{EAP}	t_P	t _{MAE}	Γ, Y_{EAP}	Y _P	M, Y_{MAE}
Segment $i = 7$	l ₇	t _{EAP}	t_P	-	Γ, Y_{EAP}	Y _P	-
Segment $i = 8$	l ₈	t _{EAP}	t_P	t _{MAE}	Γ, Y_{EAP}	Y _P	M, Y_{MAE}

The same large displacement composite beam model discussed in the Methodology section was used to simulate the structures for the combinatorial set of parameters listed below, yielding 125 cases.

$$M = \{0.1, 0.2, 0.3, 0.45, 0.6\} T, \tag{5-23}$$

$$Y_P = \{0.1, 0.5, 1, 5, 10\} Y_{EAP}, \tag{5-24}$$

$$\Upsilon = \{0, 1/3, 1, 3, 7\}.$$
(5-25)

The magnetization M was varied from 0.1 Tesla to 0.6 Tesla; the increments of the Young's Modulus of the passive layer, Y_P , were chosen relative to the Young's modulus of the EAP, Y_{EAP} ; and the ratio of the lengths of two-layer to three-layer segments were varied from 0, which means the entire beam is covered with MAEs, to 7, which means the MAE patches were very short. A visual representation of the length ratios, Υ , are shown in Figure 5-12.



Figure 5-12: Chosen length parameter is visualized on a sample configuration. Red layers are MAEs, blue layers are passive, and green layers are EAPs. The yellow arrows on the MAE patches indicate direction of magnetization. From Υ_1 to Υ_5 , segments containing MAEs increase in length, while the remaining segments reduce in length.

The remaining material properties and structure dimensions are borrowed from literature (Erol, Lockette, and Frecker 2018), and listed in Table 5-3. The width was w = 1 cm, and the length is L = 3 cm. The maximum fields used were $H_{max} = 30 kA/m$ and $E_{max} = 50 MV/m$. The fields were incremented at $\Delta H = H_{max}/15$, followed by $\Delta E = E_{max}/15$, totaling 30 steps to reach the final deformed shape.

Table 5-5. Waterial and layer properties for WAE, EAT, and the passive layer.				
Parameter	MAE	EAP	Passive Layer	
t (μm)	520	30	62	
Y (MPa)	3.5	200	Y_P (varied)	

Table 5-3. Material and layer properties for MAE, EAP, and the passive layer

Quantitative comparisons were made to determine how much the objective functions varied based on parametric changes. The simulations were assessed in a performance space to view the tradeoffs between the objectives, as shown in Figure 5-11, where each point represents a feasible design. The points are also color coded to distinguish their length ratio. The best designs are expected to be in the lower left corner of Figure 5-13.

Samples of the deformed shapes of the best designs, plotted with a custom graphics code in MATLAB that schematically represents the number of layers, segments, and material properties, are shown in Figure 5-13. Each layer's color intensity signifies a specific property. The MAEs become more red as their magnetization M increases; the passive layer becomes more blue as Y_p increases; and the EAP is a static green to indicate constant electrostrictive properties. Recall that the MAEs are oriented in the directions shown in Figure 5-11, with an external field upward. The layer thicknesses are not shown to scale, and thus any overlapping on the images does not imply physical contact between MAE patches from different segments. In reality, the thicknesses are small enough to allow very large curvatures.

Configurations within length ratio 1 were relatively low cost due to a low volume of MAEs, but they performed approximately in the middle of the total group of simulations. By contrast, length ratio 5 performed poorly across all parameter sweeps, and was the most costly due to the high volume of MAEs. Length ratios 2, 3, and 4 were more diagonally distributed on the f_{shape} versus f_{cost} map.

The three best performing configurations for each length ratio, as indicated by arrows in Figure 5-13, were identified. Conversely, the worst designs are entirely of length ratio 5 (top right schematic in Figure 5-13), showing little change as other parameters are varied. This may be a result of the high stiffness of the MAEs; when the MAEs are continuous across the length of the beam, no region of the structure exhibits relatively high bending, as in the case of the other length ratios. Thus, any attempt at achieving folding or even moderate displacements may require spacing between the MAEs, which agrees with findings reported in the literature (Cowan 2015).



Figure 5-13. The performance space is shown. Each dot represents a simulation of a different combination of M, Y_P , and Υ . The length ratio, Υ , is indicated by color. The colored arrows on the bottom left indicate the best of the feasible designs.

The effectiveness of the shape function's ability to gauge how well a design approximates the target shape was also studied by visually comparing the deformed shapes of designs along with their f_{shape} metrics. For example, the deformed shapes of the best designs in terms of shape are displayed in Figure 5-14. The target shape is also displayed as an overlay on each simulated shape. It is evident from Figure 5-14 that as the objective function f_{shape} decreases among the best designs, the simulated shape better matches the target shape. In addition, the objective function also decreases as folding becomes more prominent at the creases between the MAE patches. This is further support for utilizing the objective function in seeking target shapes with sharp folds. However, this finding also implies that the length ratios 2, 3, and 4 are likely ideal starting locations for a formal optimization problem, and could potentially save significant time in searching for the best possible active self-folding design.



Figure 5-14. Deformed shapes of the three best designs with overlays of the target shape (red lines). From left to right: L2-M5-Y1, L3-M4-Y1, L4-M3-Y1.

4.2.8 Formal Optimization Problem Results and Discussions

The results of the parametric study informed choice of parameters such that the optimization problem would yield even better designs. Namely, the Young's modulus of the passive layer is removed as a parameter, and kept constant at $Y_P = 0.1Y_{EAP}$, since the lowest Y_P produced the best designs in the parametric study. Furthermore, the magnetizations and segment lengths are not assumed equal for each MAE patch or segment, respectively. As a result, the set of parameters for the optimization problem is

$$\mathbf{S} = \{M_2, M_4, M_6, M_6, l_1, l_2, l_3, l_4, l_5, l_6, l_7, l_8\}.$$
(5-26)

The same constants from the parameter study are used for the optimization (see Table 5-3). Additionally, a tip force is present in the optimization problem to calculate f_{work} . The magnitude of the force is manually chosen to be 10 mN, and the magnetic field is increased to 70 kA/m, which allows the structure to fold beyond the target shape at the configuration with the highest BHF content.

Since a genetic algorithm is employed for optimization, an initial search population must be selected. MATLAB's genetic algorithm, gamultiobj, can generate a default initial population, but a few preliminary optimization iterations utilizing the default initial population did not yield a diverse set of optimal designs.

To address this issue, the initial population is modified by (1) adding the best designs from the parametric study, which can aid the algorithm in finding more optimal spaces, and (2) designs with more extreme segment lengths, particularly those with $\Upsilon > 7$, are introduced, which helps promote diversity in the initial population. The custom designs selected for the initial population do not add to the total population size, which is 100. Consequently, the remaining individuals in the initial population are generated by the default algorithm.

The optimization is performed with the genetic algorithm, and allowed to converge toward a Pareto front for minimizing the three objective functions, shown in Figure **5-15**. The axes represent the three objective functions, and each circle in the Pareto front represents an optimal individual whose performances in terms of the objective functions cannot be improved by changing their parameters. The path of the Pareto front is 3-dimensional and contains individuals with a range of performances in terms of the objective functions. For example, there are individuals with high shape error, low cost and low work on one end, and those with low shape error, high cost, and high work on the other end. Individuals at around the mid-point of the Pareto front have a more balanced performance in terms of the three objectives.



Figure 5-15. The Pareto front for the three objective functions is shown in a 3D plot, as obtained from the genetic algorithm. Each point represents an optimal individual based on the objective functions.

Samples of the deformed shapes of feasible designs on the Pareto front are provided in Figure **5-15**. Design 1 has the highest shape error (16), lowest cost (\$1.6), and least work done (0.12). Design 2 has moderate shape error (5.1), cost (\$7.1), and work (0.84). Design 3 has the least shape error (0.12), and high cost (\$25.4), and work (1.49). Design 4 has the low shape error (0.71), highest cost (\$25.4), and the best work (1.81). A summary of these results are listed in Table 5-4.

	D1	D2	D3	D4
f_{shape}	16	5.1	0.12	0.71
f _{cost} (\$)	1.6	7.1	17.3	25.4
$-f_{work}\left(10^{-4}\right)$	0.12	0.84	1.49	1.81
<i>M</i> ₂ (T)	0.093	0.19	0.36	0.45
$M_4(T)$	0.037	0.20	0.29	0.57
<i>M</i> ₆ (T)	0.15	0.31	0.24	0.45
<i>M</i> ₈ (T)	0.068	0.19	0.31	0.32
l_1	0.200	0.137	0.051	0.051
<i>l</i> ₂	0.050	0.090	0.183	0.199
l_3	0.200	0.154	0.052	0.051
l_4	0.051	0.084	0.216	0.198
<i>l</i> ₅	0.199	0.152	0.050	0.051
l ₆	0.052	0.120	0.216	0.201
l_7	0.198	0.147	0.050	0.050
l ₈	0.051	0.117	0.183	0.199

Table 5-4. Objective function evaluations, magnetizations, and segment length ratios for the highlighted in designs in Figure 11.

The design with the highest shape error (or least matching shape), Design 1, has very short MaE patches, while those with the best matching shapes near the other end of the Pareto front, such as Designs 3 and 4, have the largest MAE patches. In between designs 1 and 4, there is a gradual change in MAE patch size. Interestingly, the lengths of the segments containing MAEs (i.e., even

numbered segments) are relatively even within each Pareto individual. Since there was no constraint on the relative lengths (as there were in the parametric study), these results imply that symmetry may promote shape matching.

Similar to the MAE patch lengths, the magnetizations of MAE patches also gradually change along the Pareto front. Design 1's MAE patches have the least magnetization among all designs, while Design 4's patches have the highest magnetizations. The values of the magnetizations for each patch on all four selected designs are listed in Table 5-4.

Another notable trend is that the magnetization of the patch on the fourth segment, M_4 , is lowest among the patches in Design 1, but highest in Design 4. In fact, M_4 in Design 4 is greater than M_4 in Design 1 by more than a factor of 15, the largest factor across any two magnetizations. Thus, this means that the system is highly sensitive to M_4 (combined with a steady change in MAE patch lengths), perhaps more than any other parameter that was used in the optimization. Similarly, the system is also sensitive to M_2 , which changes by almost a factor of 5 between Design 1 and Design 4.

Another trend in the Pareto space is that M_8 remains the lowest in almost every design. It is likely that M_8 is lowest in most designs since it is located on the 8th segment, which is the free end of the beam. Regions near the free end of the beam may not require as much torque, which can be useful information when designing a magnetically-actuated cantilever device.

The only exception to this rule is found in Design 3. Unlike the other selected designs, Design 3 has an M_8 that is not the weakest of its MAE patches. In fact, the magnetizations of the MAE patches in Design 3 are relatively uniform. The smallest magnetization ($M_6 = 0.24 T$) in Design 3 is only 66% of the largest magnetization ($M_2 = 0.36 T$). Compared to the designs (25% for Design 1, 61% for Design 2, and 57% for Design 3), this is the closest relative magnetizations between the weakest and strongest MAE patches in a design. There is also better symmetry in Design 3 than in any other design. The outer MAE patches (M_2 and M_8) have the highest magnetizations while the inner MAE patches (M_4 and M_6) have the lowest. Conversely, Designs 1 and 2 are asymmetric with M_6 being greater than the next strongest MAE patch in each design, and without a balance on the other end of the beam (i.e., M_2 or M_4). Similarly, Design 4 is skewed toward the fixed end of the beam. Thus, the results imply that symmetry and relatively uniform magnetizations are best for matching the chosen target shape. This is not a coincidence, since the chosen target shape is also symmetric about the midpoint. However, it is also important to point out that the magnetizations and lengths of Design 3 are not *exactly* uniform or symmetric, which would be expected for the chosen target shape. This is likely due to (i) a bias with the initial population, which led to a convergence toward a non-symmetric or uniform design in terms of magnetizations, and (ii) a relatively low population size of 100. It may also be due to the boundary conditions (fixed end, and free end with applied load) combined with numerical approximation errors.

Nonetheless, despite the optimization results not being exactly symmetric as expected for some of the designs, the methodologies presented in this work have yielded quick results that perform significantly better than any design found via the parameter study. For instance, the lowest shape error acquired from the parameter study was 0.34, which is nearly three times greater than the lowest shape error from the optimization (0.12, Design 3). Similarly, the lowest cost from the parameter study, \$3.5 (L3-M4-Y1), is more than twice as much as the lowest cost of a Pareto design from the optimization, \$1.6 (Design 1). Thus, the optimization method improved upon the parameter study in terms of the objective functions, and it took about 40 hours to run on a 100-core cluster, which is a feasible computational speed for results several times better than any results that could be obtained from trial and error, or a parametric study.

Based on this analysis, if an application requires ideal shape matching, then the best design should have relatively high, uniform, and symmetric magnetizations and long MAE patches throughout the length of the device as in Design 3. If an application prioritizes costs, then shortening the lengths of the MAE patches and reducing their magnetizations, while maintaining patch placement symmetry, as in Design 1, is ideal. For maximizing work, the magnetizations should be increased near the limits, with M_4 the highest among the MAE patches, as in Design 4. If an application requires more complex priorities, then one can select another design based on the tradeoffs in the Pareto front that meet their needs.

4.3 Conclusions on Self-Folding Composite Optimization

The purpose of this study was to answer hypothesis 3 by developing a multi-objective design optimization problem for a multi-field actuated device employing magneto- and electroactive materials. An existing model accommodating large displacements and magneto- and electromechanical coupling was used and extended for the application of a force on one end of the beam. The design optimization problem was written for a multi-field actuated device consisting of several parameters, including number of segments and layers; thicknesses, lengths, and all material properties. Three objective functions were chosen: (1) shape error, i.e., the error between calculated and target shapes, (2) cost, and (3) work performed on the tip load.

To assist the formal optimization, a case study consisting of eight segments was considered, reducing the number of parameters and possible combinations of materials at each layer and segment. Furthermore, a preliminary parameter study was performed on the eight-segmented bimorph structure design to predict how a set of parameters affect the self-folding of the structure. Only the shape and cost objective functions were considered. The parameters chosen for the study were the magnetization of the MAEs, the Young's modulus of the passive layer, and the length ratio of the segments. The results of the parameter study found that each parameter influenced shape approximation by almost an order of magnitude. Furthermore, the designs that yielded the lowest shape error performed significantly better in both matching the target shape and achieving self-folding, which implies that the chosen shape objective function was adequate in assessing the degree of folding.

The results of this parameter study were used in determining the constraints and initial search population for the optimization algorithm, and in reducing the number of design variables. For example, relatively long MAE patches were used as a good starting point, since long MAE patches performed best among all length ratios in terms of folding. Configurations with MAE

patches covering the entire length of the structure were omitted from the design space, and very small MAE patches were also eliminated. The parameter study also highlighted the significance of the Young's modulus of the passive layer and suggested that self-folding requires very low Young's modulus of the passive layer with respect to the EAP layer. Thus, the Young's modulus was preset to the lowest value from the parametric study. Using the best designs from the parameter study in the initial population of the genetic algorithm yielded better optimization results in terms of the three objective functions.

The multi-objective genetic algorithm produced a Pareto front with a range of optimal individuals to select from based on the priority of the design. The Pareto front showed optimal designs in terms of cost, shape, and work. Nearly all designs were relatively symmetric in length segments and contained a range of magnetizations. However, some MAE locations, such as M_4 , played a more important role in actuation than others, especially in terms of shape approximation. Furthermore, the results yielded much better designs in terms of the chosen objective functions than any design obtained from the parameter study. It can be concluded from the genetic algorithm results that the optimization method is an improvement over trial and error or parametric studies, and performs well in terms of computational speed (two days with 100 nodes).

Future improvements on the optimization can be made through adjustments to the settings. For example, different custom initial populations can yield different Pareto fronts, which can potentially contain more optimal individuals. Particularly, the diversity of the initial population plays a significant role, which can be studied further. In addition, the number of elites was not modified, which could also influence which traits are passed down in the genetic algorithm.

Additionally, several variables were not considered in this study, such as the width and length of the structure, Young's moduli of the MAE and EAP, and the electrostrictive properties of the EAP. Both the electric and magnetic fields were kept constant for the simulations, and we chose a specific order of actuation (magnetic field first, and then electric field). Varying any of these parameters may alter influence the self-folding behavior of the structure, which means that the scope of this study is limited to structures with similar configurations and constants. As a result, a more thorough investigation utilizing a larger parameter space may generate more optimal Pareto fronts. However, our results show this methodology may be useful for the multi-objective optimization of arbitrary multi-field-active, compliant, self-folding structures.

CHAPTER 5

Thesis Conclusions, and Future Works

As a doctoral thesis, this document presents works on modeling and simulating dipolebased materials on multiple scales. This chapter aims to summarize the findings, discuss conclusions in a wider context, and project future outlooks for the work.

5.1 Conclusions and Future Works

Each chapter in this document completed the objectives related to their respective hypothesis outlined in Chapter 1.

In Chapter 2, the first hypothesis was addressed, which was on how particle simulations utilizing ferrohydrodynamics could provide evidence for whether applying magnetic and electric fields to particles suspended in a fluidic polymer matrix yields a microstructure with multiple levels of hierarchy. To answer this hypothesis, the objective to develop simulation methods for electro-magnetically susceptible particles in a fluid matrix was accomplished, and simulation results were used to analyze five processing cases. The differences between the cases revealed that four levels of structural hierarchy could be obtained: particle, stack, micro-chain, and macrochain levels. Each level was quantitatively assessed via a dipole-based spanning tree algorithm, which was found to be an effective way to group particles into structures commonly identified. Furthermore, metrics such as percolation and the amount of mass in a spanning tree were effective in distinguishing the structures at each length-scale. In fact, the differences in these metrics for structures across all five cases showed microstructural differences that could lead to changes in material properties. For example, the higher percolation in Case 5 versus Case 3 is the primary difference between the microstructures of these cases, and percolation alone can influence elastic, dielectric, and conductive properties. Additional analysis of the distributions of the orientations of the structures across all cases also showed how well alignment can be

manipulated by processing conditions, and in turn, properties such as the net magnetic torque. For example, high particle alignment directly correlates with the magnetic torque of a bulk composite containing magnetically dipolar materials, and the simulations show how each processing condition influences their alignments.

In Chapter 3, the second hypothesis was addressed, which was on how network models of semicrystalline hyperelasticity accounting for microstructure by representing different relative locations of crystalline regions can form an effective framework for modeling electrostriction. The chapter developed a framework incorporating various aspects of the microstructure of a semicrystalline class of EAPs, relaxor ferroelectrics, and in doing so, related the relative spatial alignments of crystalline domains with neighboring crystalline domains. The analyses revealed that parallel arrangements of crystalline regions yielded much greater electromechanical coupling than other configurations, due to the anisotropic nature of dipole-dipole interactions. Additionally, due to this anisotropy, parallel configurations were also found to cause material failure through instabilities. The model's ability to relate these arrangements to predict material response and even failure can provide engineers a means of avoiding instabilities, or possibly promoting them, since researchers have found ways to harness them to generate larger deformations. Finally, a probability density function (PDF) for the relative spatial arrangements of dipolar regions proved useful in fitting the model to data. The PDF-based network model offered a higher fidelity approach to modeling EAPs and could allow for a means of validating the model to more realistic microstructures through the dispersion parameter.

In Chapter 4, the third hypothesis was addressed, which was on how an analytical model for multi-layer, segmented composite beams incorporating the EAP constitutive model and magnetic torques can accurately predict multi-field device response, and can be used for the design optimization for a target shape containing ideal folds. To address this hypothesis, a new multi-layered, segmented composite beam model accommodating large displacements and constitutive relations for magneto- and electro-active models was developed. The model was used in an optimization problem for a specific configuration with the objectives of matching a shape with ideal folds, minimizing cost and maximizing work performed on a tip force. Optimization results revealed the importance of gaps between MAE patches and the uniformity and symmetry in their magnetization for matching symmetric shapes with ideal folds. For lowering costs, short MAE patches with high magnetization were ideal. For greatest work performance, long MAE patches with near maximum magnetization were essential, offering a trade-off between work and shape approximation. While these results could be improved with a larger and more diverse initial population, the methodology demonstrated the ability to quickly achieve near-optimal designs with a wide selection of designs based on application priorities. Studies also highlight the need to begin the optimization with a population of relatively strong candidates, possibly found from a parameter study, as opposed to using a random initial population.

Designers can ultimately choose the appropriate design for a given application based off the trade-offs between the three objectives. The differences between the designs, such as increase in magnetization or change in the segment lengths, determine the trade-offs, and knowing how these parameters play a role in the performance (or Pareto front) can help one design the ideal structure for an application.

The work presented in this document still has a lot of room for advancement. For instance, the simulation methods for multi-field processing need more robust methods including more realistic interactions between particles, such as friction, ellipsoidal forms of drag terms, and nonlinear external fields. The electrostriction model can be expanded further to include different crystalline phases, sizes, shapes, and defects within the microstructure, also with a metric for the varying distances between crystalline regions. The optimization can be expanded to include aspects of the first two links, i.e. the links between (1) processing and microstructure and (2) microstructure and material response, such that the optimization problem can be defined to link processing directly to structural response, as shown in Figure 5-1. In this manner, an engineer

could find optimal processing parameters for a given material constituent set for a multi-objective trade-space.



Figure 6-1. A full-scale optimization problem may only require material set properties and structure configuration such that the only variables can be processing conditions, and the outputs structural response relating to multiple objectives. This way, a direct link between processing conditions and structural response objectives, f_{shape} , f_{cost} , and f_{work} , can be obtained.

5.3 Funding Acknowledgements

I gratefully acknowledge the support of the National Science Foundation grant numbers EFRI 1240459, MEP 1762188, and the Air Force Office of Scientific Research. Any opinions, findings, and conclusions or recommendations expressed in this material are those of the author and do not necessarily reflect the views of the National Science Foundation.

REFERENCES

- [1] Xu, Yuhuan. Ferroelectric Materials and their Applications, North-Holland, Amsterdam;New York;, 1991;2013;.
- [2] Chu, Baojin, et al. "Relaxor Ferroelectric Poly(Vinylidene Fluoride-TrifluoroethyleneChlorofluoroethylene) Terpolymer for High Energy Density Storage Capacitors." IEEE Transactions on Dielectrics and Electrical Insulation 13.5 (2006): 1162-9. Web.
- [3] Wang, Yong, et al. "Recent development of high energy density polymers for dielectric capacitors." Dielectrics and Electrical Insulation, IEEE Transactions on 17.4 (2010): 10361042.
- [4] Neese, Bret, et al. "Large electrocaloric effect in ferroelectric polymers near room temperature." Science 321.5890 (2008): 821-823.
- [5] Wu, Shan, et al. "A high-K ferroelectric relaxor terpolymer as a gate dielectric for organic thin film transistors." Applied Physics Letters 102.1 (2013): 013301.
- [6] Carpi, Federico, and ScienceDirect (Online service). Dielectric Elastomers as Electromechanical Transducers: Fundamentals, Materials, Devices, Models and Applications of an Emerging Electroactive Polymer Technology. Amsterdam, Netherlands;Oxford, UK;Boston [Mass.];: Elsevier, 2008;2011;. Web.
- [7] Ohigashi, H., et al. "Piezoelectric and ferroelectric properties of P (VDF-TrFE) copolymers and their application to ultrasonic transducers." Ferroelectrics 60.1 (1984): 263-276.
- [8] Bar-Cohen, Yoseph. Electroactive polymer (EAP) actuators as artificial muscles: reality, potential, and challenges. Vol. 136. SPIE press, 2004.
- [9] Ting, Robert Y., and Qi M. Zhang. New Electrostrictive PVDF Copolymers for Large-Strain Actuator Application, vol. 4073, 2000.doi:10.1117/12.396414.
- [10] Erol, Anil, et al. "Analysis of Microstructure-Based Network Models for the Nonlinear Electrostriction Modeling of Electro-Active Polymers" ASME 2017 Conference on Smart Materials, Adaptive Structures and Intelligent Systems. American Society of Mechanical Engineers. *Submitted May 2017*.
- [11] Bauer, F., et al. "Ferroelectric copolymers and terpolymers for electrostrictors: synthesis and properties." Electrets, 2002. ISE 11. Proceedings. 11th International Symposium on. IEEE, 2002.
- [12] Legrand, J. F. "Structure and ferroelectric properties of P (VDF-TrFE) copolymers." Ferroelectrics 91.1 (1989): 303-317.
- [13] Zhang, Q. M., Vivek Bharti, and X. Zhao. "Giant electrostriction and relaxor ferroelectric behavior in electron-irradiated poly (vinylidene fluoride-trifluoroethylene) copolymer." Science 280.5372 (1998): 2101-2104.
- [14] Bharti, Vivek, et al. "Polarization and structural properties of high-energy electron irradiated poly (vinylidene fluoride-trifluoroethylene) copolymer films." Journal of Applied Physics 87.1 (2000): 452-461.

- [15] Bobnar V, Vodopivec B, Levstik A. 2003. Dielectric Properties of Relaxor-like Vinylidene Fluoride-Trifluoroethylene-Based Electroactive Polymers
- [16] Xu, Haisheng, et al. "Ferroelectric and electromechanical properties of poly (vinylidenefluoride-trifluoroethylene-chlorotrifluoroethylene) terpolymer."Applied Physics Letters 78.16 (2001): 2360-2362.
- [17] Klein, Rob J., et al. "Influence of composition on relaxor ferroelectric and electromechanical properties of poly (vinylidene fluoride-trifluoroethylene-chlorofluoroethylene)." Journal of applied physics 97.9 (2005): 094105.
- [18] Lu, Yingying, et al. "Structural Dependence of Phase Transition and Dielectric Relaxation in Ferroelectric Poly (vinylidene fluoride- chlorotrifluoroethylene- trifluoroethylene) s." The Journal of Physical Chemistry B 112.34 (2008): 10411-10416.
- [19] Ang, C., and Z. Yu. "Ferroelectric, Electroactive, and Dielectric-Relaxation Behavior of Fluoropolymers." ADVANCED MATERIALS 16.12 (2004): 979-. Web.
- [20] Cheng, Zhong-Yang, et al. Electromechanical Properties and Molecular Conformation in P(VDF-TrFE)-Based Terpolymer, vol. 4695, 2002.doi:10.1117/12.475162.
- [21] Lu, Yingying, et al. "Structural Dependence of Phase Transition and Dielectric Relaxation in Ferroelectric Poly(Vinylidene Fluoride-Chlorotrifluoroethylene-Trifluoroethylene)s." The journal of physical chemistry. B 112.34 (2008): 10411. Web.
- [22] Bao, Hui-Min, et al. "Phase transitions and ferroelectric relaxor behavior in P (VDF-TrFE-CFE) terpolymers." Macromolecules 40.7 (2007): 2371-2379.
- [23] Duan, CG, et al. "Simulations of Ferroelectric Polymer Film Polarization: The Role of Dipole Interactions." PHYSICAL REVIEW B 69.23 (2004)Web.
- [24] Kühn, M., and H. Kliem. "Monte Carlo simulations of ferroelectric properties based on a microscopic model for PVDF." physica status solidi (b) 245.1 (2008): 213-223.
- [25] Nakhmanson, S. M., M. Buongiorno Nardelli, and J. Bernholc. "Collective polarization effects in β-polyvinylidene fluoride and its copolymers with tri-and tetrafluoroethylene." Physical Review B 72.11 (2005): 115210.
- [26] Su, Haibin, Alejandro Strachan, and William A. Goddard III. "Density functional theory and molecular dynamics studies of the energetics and kinetics of electroactive polymers: PVDF and P (VDF-TrFE)." Physical Review B 70.6 (2004): 064101.
- [27] Abe, Y., and K. Tashiro. "Computer simulation of structure and ferroelectric phase transition of vinylidene fluoride copolymers. IV. The factors governing the ferroelectric phase transition of VDF–TrFE copolymers." Journal of Polymer Science Part B: Polymer Physics 39.6 (2001): 689-702.
- [28] Prosandeev, S. A. "Orientational and Fluctuation Polarizations of Langevin Dipoles in a Random Electric Field." Physics of the Solid State 45.9 (2003): 1774-9. Web.
- [29] Capsal, Jean-Fabien, et al. "Evaluation of macroscopic polarization and actuation abilities of electrostrictive dipolar polymers using the microscopic Debye/Langevin formalism." Journal of Physics D: Applied Physics 45.20 (2012): 205401.

- [30] Yang, Lianyun, et al. "Novel Polymer Ferroelectric Behavior Via Crystal Isomorphism and the Nanoconfinement Effect." Polymer 54.7 (2013): 1709-28. Web.
- [31] Zäh, Dominic, and Christian Miehe. "Multiplicative Electro-Elasticity of Electroactive Polymers Accounting for Micromechanically-Based Network Models." Computer Methods in Applied Mechanics and Engineering 286 (2015): 394-421. Web.
- [32] Treloar, Leslie Ronald George. The physics of rubber elasticity. Oxford University Press, USA, 1975.
- [33] Mooney M. A theory of large elastic deformation. J Appl Phys 1940;11(9):582–92.
- [34] Rivlin R. Large elastic deformations of isotropic materials. IV. Further developments of the general theory. Philos Trans Roy Soc Lond 1948;A 241:379–97.
- [35] Tu, Jiyuan, Guan Heng Yeoh, and Chaoqun Liu. Computational fluid dynamics: a practical approach. Butterworth-Heinemann, 2012.
- [36] Gent, A. N. "A new constitutive relation for rubber." Rubber chemistry and technology 69.1 (1996): 59-61.
- [37] Wu, P. D., and Erik Van Der Giessen. "On improved network models for rubber elasticity and their applications to orientation hardening in glassy polymers." Journal of the Mechanics and Physics of Solids 41.3 (1993): 427-456.
- [38] Ogden, Raymond W. Non-linear elastic deformations. Courier Corporation, 1997.
- [39] Arruda, E. M. and Boyce, M. C., 1993, A three-dimensional model for the large stretch behavior of rubber elastic materials, J. Mech. Phys. Solids, 41(2), pp. 389–412.
- [40] Sencadas, V.; Gregorio, R.; Lanceros-Mendez, S. Alpha to Beta Phase Transformation and Microstructural Changes of PVDF Films Induced by Uniaxial Stretch. J. Macromol. Sci. B 2009, 48, 514–525.
- [41] Branciforti, M. C.; Sencadas, V.; Lanceros-Mendez, S.; Gregorio, R. New Technique of Processing Highly Oriented Poly(Vinylidene Fluoride) Films Exclusively in the Beta Phase. J. Polm. Sci. Polym. Phys. 2007, 45, 2793–2801.
- [42] Sencadas, V.; Gregorio, R.; Lanceros-Mendez, S. Processing and Characterization of a Novel Nonporous Poly(Vinilidene Fluoride) Films in the Beta Phase. J. Non-Cryst. Solids 2006, 352, 2226–2229.
- [43] Ranjan, V.; Nardelli, M. B.; Bernholc, J. Electric Field Induced Phase Transitions in Polymers: A Novel Mechanism for High Speed Energy Storage. Phys. Rev. Lett. 2012, 108, 087802-1–087802-5.
- [44] Ranjan, V.; Yu, L.; Nardelli, M. B.; Bernholc, J. Phase Equilibria in High Energy Density PVDF-Based Polymers. Phys. Rev. Lett. 2007, 99, 047801-1–047801-4.
- [45] Zhang, Shihai, et al. "Direct spectroscopic evidence of field-induced solid-state chain conformation transformation in a ferroelectric relaxor polymer." Journal of applied physics 99.4 (2006): 044107.

- [46] Chu, Baojin. PVDF-based copolymers, terpolymers and their multi-component material systems for capacitor applications. Diss. The Pennsylvania State University, 2008.
- [47] Ullah, Amir, et al. "Enhancement of dielectric and energy density properties in the PVDFbased copolymer/terpolymer blends." Polymer Engineering & Science 55.6 (2015): 1396-1402.
- [48] Smith, O'Neil L., et al. "Enhanced permittivity and energy density in neat poly (vinylidene fluoride-trifluoroethylene-chlorotrifluoroethylene) terpolymer films through control of morphology." ACS applied materials & interfaces 6.12 (2014): 9584-9589.
- [49]"Investigators from Ben-Gurion University of the Negev Target Mechanics (Multiscale Analysis of the Electromechanical Coupling in Dielectric Elastomers)." Electronics Newsweekly (2014): 228. Web.
- [50] Bystrov, Vladimir S., et al. "Molecular modeling of the piezoelectric effect in the ferroelectric polymer poly (vinylidene fluoride)(PVDF)." Journal of molecular modeling 19.9 (2013): 3591-3602.
- [51] Richards, A. W., and G. M. Odegard. "Constitutive Modeling of Electrostrictive Polymers using a Hyperelasticity-Based Approach." Journal of Applied Mechanics 77.1 (2010): 14502. Web.
- [52] Ahmed, Saad, and Zoubeida Ounaies. "Self-Clearing of Metalized Electrodes and its Impact on Electroactive Polymer (EAP) Based Actuators." ASME 2016 Conference on Smart Materials, Adaptive Structures and Intelligent Systems. American Society of Mechanical Engineers, 2016.
- [53] Ploss, Bernd. "Dielectric nonlinearity of PVDF–TrFE copolymer." Polymer 41.16 (2000): 6087-6093.
- [54] Erol, Anil, et al. "On the modeling and experimental validation of multi-field polymer-based bimorphs." ASME 2016 Conference on Smart Materials, Adaptive Structures and Intelligent Systems. American Society of Mechanical Engineers, 2016.
- [55] Lang, R. 1989. "Origami: Complexity Increasing." Engineering & Science, 52(2), 16-23.
- [56] Nishiyama, Yutaka. 2012. "Miura Folding: Applying Origami to Space Exploration." *International Journal of Pure and Applied Mathematics* 79 (2): 269-279. ISSN: 1311-8080. Osaka, Japan.
- [57] Miura, Koryo. 1980. "Method of Packaging and Deployment of Large Membranes in Space." 31st Congress of the International Astronautical Federation. IAF-80 A31. Tokyo, Japan.
- [58] Schenk, Mark and Simon D. Guest. 2013. "Geometry of Miura-Folded Metamaterials."
 Proceedings of the National Academy of Sciences 110 (9): 3276-3281.
 doi:10.1073/pnas.1217998110.
- [59] Zhao, Xilu, Yabo HU, and Ichiro Hagiwara. 2011. Study on crash characteristics of half cut type vehicle side member structure of energy absorption ability by using origami engineering. Journal of Computational Science and Technology 5 (1): 13-25.
- [60] Hill, David. 2015. Researchers unfold the structural benefits of origami. Civil Engineering 85 (11): 36.
- [61] Dae-Young Lee, Gwang-Pil Jung, Min-Ki Sin, Sung-Hoon Ahn, and Kyu-Jin Cho. 2013.
 "Deformable Wheel Robot Based on Origami Structure." 2013 IEEE International Conference on Robotics and Automation (ICRA): 5612-5617. doi:10.1109/ICRA.2013.6631383. Karlsruhe, Germany.
- [62] Dae-Young Lee, Ji-Suk Kim, Sa-Reum Kim, Je-Sung Koh, and Kyu-Jin Cho. 2013. "The Deformable Wheel Robot Using Magic-Ball Origami Structure." *International Design Engineering Technical Conferences & Computers and Information in Engineering Conference*. DETC2013-13016. Portland, OR.
- [63] Onal, C. D., R. J. Wood, and D. Rus. 2013. "An Origami-Inspired Approach to Worm Robots." *IEEE/ASME Transactions on Mechatronics* 18 (2): 430-438. doi:10.1109/TMECH.2012.2210239.
- [64] Martinez, Ramses V., Carina R. Fish, Xin Chen, and George M. Whitesides. 2012.
 "Elastomeric Origami: Programmable Paper-Elastomer Composites as Pneumatic Actuators." *Advanced Functional Materials* 22 (7): 1376-1384. doi:10.1002/adfm.201102978. Weinheim, Germany.
- [65] Sacca, B., and CM Niemeyer. 2012. DNA origami: The art of folding DNA. Angewandte Chemie-International Edition 51 (1): 58-66. Link
- [66] Wauer, Tobias, Hendrik Dietz, Mark Bathe, Carlos Ernesto Castro, Philipp Wortmann, Fabian Kilchherr, Do-Nyun Kim, and Enrique Lin Shiao. 2011. A primer to scaffolded DNA origami. Nature Methods 8 (3): 221-9.
- [67] Nangreave, Jeanette, Dongran Han, Yan Liu, and Hao Yan. 2010. DNA origami: A history and current perspective. Current Opinion in Chemical Biology 14 (5): 608-15.
- [68] Chandrasekaran, AR, M. Pushpanathan, and K. Halvorsen. 2016. Evolution of DNA origami scaffolds. Materials Letters 170: 221-4
- [69] Kuribayashi, Kaori, et al. "Self-Deployable Origami Stent Grafts as a Biomedical Application of Ni-Rich TiNi Shape Memory Alloy Foil." Materials Science & Engineering A 419.1 (2006): 131-7. Web.
- [70] Peraza-Hernandez, Edwin, et al. "Design and Optimization of a Shape Memory Alloy-Based Self-Folding Sheet." Journal of Mechanical Design 135.11 (2013)Web.
- [71] D'Amato, Matteo, et al. "Photo-Origami-Bending and Folding Polymers with Light." Applied Physics Letters 100.16 (2012): 5. Web.
- [72] Na, Jun-Hee, et al. "Programming Reversibly Self-Folding Origami with Micropatterned Photo-Crosslinkable Polymer Trilayers." Advanced Materials 27.1 (2015): 79-85. Web.
- [73] Bowen, Landen, Kara Springsteen, Hannah Feldstein, Mary Frecker, Timothy W. Simpson, and Paris von Lockette. 2015. "Development and Validation of a Dynamic Model of Magneto-Active Elastomer Actuation of the Origami Waterbomb Base." Journal of Mechanisms and Robotics 7 (1). doi:10.1115/1.4029290.

- [74] Cowan, B. (2015) "Magnetically Induced Actuation and Optimization of the Miura-ori Structure," Master's Thesis(advisor, P. vonLockette), Penn State University, University Park, Pa.
- [75] Crivaro,A; Sheridan, R; Frecker,M; Simpson, T; Von Lockette, P. "Bistable compliant mechanism using magneto active elastomer actuation." Journal of Intelligent Material Systems and Structures (2015) doi:10.1177/1045389X15620037
- [76] R Sheridan, J Roche, S E Lofland, and P R vonLockette, "Numerical simulation and experimental validation of the large deformation bending and folding behavior of magnetoactive elastomer composites", Smart Materials and Structures: Special Issue on Origami Engineering, vol 23:9 (2014), pg. 094004.
- [77] von Lockette, Paris R. and Robert Sheridan. 2013. "Folding Actuation and Locomotion of Novel Magneto-Active Elastomer (MAE) Composites." ASME 2013 Conference on Smart Materials, Adaptive Structures and Intelligent Systems. SMASIS2013-3222. Snowbird, UT.
- [78] Xu, Haisheng, et al. "Ferroelectric and electromechanical properties of poly (vinylidenefluoride-trifluoroethylene-chlorotrifluoroethylene) terpolymer."Applied Physics Letters 78.16 (2001): 2360-2362.
- [79] Bauer, F., Qiming Zhang, and E. Fousson. "Ferroelectric Relaxor Terpolymers: Properties and Potential Applications". Web.
- [80] von Lockette, P.; Lofland, S. E.; Biggs, J.; Roche, J.; Mineroff, J.; Babcock, M., "Investigating new symmetry classes in magnetorheological elastomers: cantilever bending behavior". Smart Mater. Struct. 2011, 20 (10), 105022.von Lockette, P.R., Lofland, S., Mineroff, J., Roche, J., Babock, M. (2011) "Investigating new symmetry classes in magneto-rheological elastomers," *Smart Materials and Structures*. 20, 105022.
- [81] Capsal, Jean-Fabien, et al. "Evaluation of macroscopic polarization and actuation abilities of electrostrictive dipolar polymers using the microscopic Debye/Langevin formalism." Journal of Physics D: Applied Physics 45.20 (2012): 205401.
- [82] Leo, Donald J. Engineering Analysis of Smart Material Systems. Hoboken, N.J: Wiley, 2007. Web.
- [83] Borcea, L. and O. Bruno. *On the magneto-elastic properties of elastomer–ferromagnet composites* in Journal of the Mechanics and Physics of Solids. 49 (2001): 2877-2919.
- [84] Dorfmann, A. and R. Ogden. *Magnetoelastic modelling of elastomers* in European Journal of Mechanics A/Solids. 22 (2003): 497-507.
- [85] Kankanala, S. and N. Triantafyllidis. *On finitely strained magnetorheological elastomers* in J. Mech. Phys. Solids. 52. (2004): 2869-2908.
- [86] Yin, H., L. Sun, and J. Chen. Micromechanics-based hyperelastic constitutive modeling of magnetostrictive particle-filled elastomers in Mechanics of Materials. 34 (2002): 505-516.
- [87] Coquelle, E. *Micromechanical analysis of an elastomer filled with particles organized in chain-like structure* in Journal of Material Sci. 41. (2006): 5941-5953.

- [88] Ponte Castañeda, P.; Galipeau, E., Homogenization-based constitutive models for magnetorheological elastomers at finite strain in J. Mech. Phys. Solids 2011, 59 (2), 194-215.
- [89] Tuan, H. and B. Marvalova. Magnetoelastic anisotropic elastomers in a static magnetic field: Constitutive equations and FEM solutions. Taylor & Francis Group, London, UK. (2010)
- [90] Galipeau, E.; Ponte Castañeda, P., The effect of particle shape and distribution on the macroscopic behavior of magnetoelastic composites in Int. J. Solids Struct. 2012, 49 (1), 1-17.
- [91] Galipeau, E.; Ponte Castañeda, P., A finite-strain constitutive model for magnetorheological elastomers: Magnetic torques and fiber rotations in Journal of Mech. Phys. Solids 2013, DOI. http://dx.doi.org/10.1016/j.jmps.2012.11.007.
- [92] J. Ginder and e. al., "Magnetostrictive phenomena in magnetorheological elastomers," International Journal of Modern Physics, vol. 16, no. B, pp. 472-478, 2002.
- [93] Barham, M., White, D., and Steigmann, D., Finite element modeling of the deformation of magnetoelastic film, in Journal of Computational Physics 2010, 229, pp 6193-6207.
- [94] Frecker, Mary I., and William M. Aguilera. "Analytical Modeling of a Segmented Unimorph Actuator using Electrostrictive P(VDF-TrFE) Copolymer." Smart Materials and Structures 13.1 (2004): 82-91. Web.
- [95] Sung, Elaine, et al. "Characterization of Self-Folding Origami Structures Using Magneto-Active Elastomers." ASME 2016 International Design Engineering Technical Conferences and Computers and Information in Engineering Conference. American Society of Mechanical Engineers, 2016.

APPENDIX A

Intermediary Calculations

Recall that the coordinates of the deformed configuration, (x, y, z), are related to the coordinates of the reference configuration, (X, Y, Z), by the relations in (6-1).

$$x = \lambda_1 X$$
, $y = \lambda_2 Y$, $z = \lambda_3 Z$ (6-1)

Since the elastic energy is a function of principal invariants I_i , and the electrostatic energy is a function of the stretches λ_i , the chain rule must be applied, as shown in (6-2).

$$\frac{\partial \widetilde{W}}{\partial \boldsymbol{B}} = \sum_{i=1}^{3} \frac{\partial \widetilde{W}}{\partial I_i} \frac{\partial I_i}{\partial \boldsymbol{B}} = \sum_{i=1}^{3} \frac{\partial \widetilde{W}}{\partial \lambda_i} \frac{\partial \lambda_i}{\partial \boldsymbol{B}}$$
(6-2)

The unknowns now are the derivatives of the invariants I_i and the stretches λ_i with respect to the left Cauchy-Green deformation tensor **B**. By the definitions of I_i , **B**, and the derivative, the relations in (6-3) are true.

$$\frac{\partial I_1}{\partial \boldsymbol{B}} = \boldsymbol{I} \quad , \qquad \frac{\partial I_1}{\partial \boldsymbol{B}} = I_1 \boldsymbol{I} - \boldsymbol{B} \quad , \qquad \frac{\partial I_1}{\partial \boldsymbol{B}} = I_3 \boldsymbol{B}^{-1} \quad , \tag{6-3}$$

The deformation tensor **B** can also be written in terms of its basis vectors, \hat{b}_i .

$$\boldsymbol{B} = \sum_{i=1}^{3} B_i \, \widehat{\boldsymbol{b}}_i \otimes \widehat{\boldsymbol{b}}_i \,, \tag{6-4}$$

where B_i are the components of **B**, and \otimes is the tensor product. Recall that $B = FF^T$ and the polar decomposition of F produces the relation

$$F = RU, \qquad (6-5)$$

in which **R** is the orthogonal tensor relating the deformation to rotations, and **U** is the stretch tensor. Thus, the relations between \hat{b}_i and any arbitrary orthonormal basis vectors \hat{E}_i in the reference configuration are

$$\widehat{\boldsymbol{b}}_i = R\widehat{\boldsymbol{E}}_i \,. \tag{6-6}$$

Since in there is no rotation in the case of the RVE developed in this section ($\mathbf{R} = \mathbf{I}$), the bases $\hat{\mathbf{b}}_i = \hat{\mathbf{E}}_i$, and within a Euclidian point space and orthonormal coordinate system, $\hat{\mathbf{E}}_i = \hat{\mathbf{e}}_i$, where $\hat{\mathbf{e}}_i$ are the orthonormal bases of the deformed configuration. As a result, **B** is rewritten as

$$\boldsymbol{B} = B_1 \hat{\boldsymbol{\iota}} \otimes \hat{\boldsymbol{\iota}} + B_2 \hat{\boldsymbol{j}} \otimes \hat{\boldsymbol{j}} + B_3 \hat{\boldsymbol{k}} \otimes \hat{\boldsymbol{k}} , \qquad (6-7)$$

where $B_1 = B_2 = \lambda_3^{-1}$ and $B_2 = \lambda_3$. Thus, to obtain the second unknown, the derivative of stretches λ_i with respect to **B**, the chain rule in (6-8) is employed.

$$\frac{\partial \lambda_i}{\partial \boldsymbol{B}} = \frac{\partial \lambda_i}{\partial B_i} \frac{\partial B_i}{\partial \boldsymbol{B}}$$
(6-8)

The relation between the stretches λ_i and B_i is known: $B_i = \lambda_i^2$, and therefore,

$$\frac{\partial \lambda_i}{\partial B_i} = \frac{1}{2\lambda_i} \frac{\partial B_i}{\partial \boldsymbol{B}},\tag{6-9}$$

and by the definition of the derivative,

$$\frac{\partial B_i}{\partial \boldsymbol{B}} = \widehat{\boldsymbol{b}}_i \otimes \widehat{\boldsymbol{b}}_i \,. \tag{6-10}$$

When (6-9) and (6-10) are substituted into (6-2), we obtain

~ -

$$\frac{\partial \widetilde{W}}{\partial \boldsymbol{B}} = \sum_{i=1}^{3} \frac{1}{2\lambda_i} \frac{\partial \widetilde{W}}{\partial \lambda_i} \widehat{\boldsymbol{b}}_i \otimes \widehat{\boldsymbol{b}}_i .$$
(6-11)

When (6-11) is substituted into the formula for Cauchy stress T,

$$T_i = \lambda_i \frac{\partial \widetilde{W}}{\partial \lambda_i}.$$
 (no sum on i) (6-12)

Lastly, this is substituted into the equilibrium equation. The divergence calculates the derivative of the diagonal components with respect to the principle directions, such that

$$(div\mathbf{T})_i = \frac{\partial}{\partial x_i} \left[\lambda_i \frac{\partial \widetilde{W}}{\partial \lambda_i} \right], \quad (\text{no sum on } i)$$
 (6-13)

where x_i are any arbitrary coordinates in the deformed configuration. In this case, they are x, y, and z. The relationship between these coordinates and the stretches are provided in (6-1). By the chain rule, (6-12) is simplified as

$$(div\mathbf{T})_{i} = \frac{\partial}{\partial\lambda_{i}} \left[\lambda_{i} \frac{\partial \widetilde{W}}{\partial\lambda_{i}} \right]. \quad (\text{no sum on } i)$$
(6-14)

APPENDIX B

Methodologies for Extracting Parameters from Experiments

B.1 Permittivity of the Amorphous Phase

The dipole-dipole interaction energy includes the relative permittivity of the medium in which the dipoles interact with each other. In the case of semicrystalline RFPs, this medium corresponds to the amorphous phase. As discussed in Chapter 2, dipoles within the crystalline phase of the material saturate at high fields, so their influence over the polarization response of the material diminishes. At high fields, the amorphous phase takes control over the polarization response of the material and it behaves as a linear dielectric. Thus, the permittivity of the amorphous phase can be extracted from available polarization data by determining the slope of polarization versus the electric field at very high field strengths.

Chapter 3 describes a method for determining the permittivity of an EAP, which is by calculating its energy density based on the polarization data, and evaluating each point based on the linear dielectric equation (4-113). Additionally, Chapter 3 derives an alternative method for the calculation of permittivity, which directly estimates the slope (or derivative) of the curve. To analyze the differences between the linear dielectric calculations and the derivative approximations, the data found in Ref [2] is used.

Figure 18 compares the linear permittivity calculations in Ref [2], and the results from the point-wise derivative approximation in (4-114) implemented on the polarization curve for P(VDF-TrFE-CFE) 63/37/7.5% found in Ref [2]. Additionally, the biphasic model discussed in section 3.3 is fitted to the derivative approximation points by a least-squares method, and the least-errored model is also displayed in Figure 15. The derivative approximation captures much greater nonlinearity at low fields compared to the constants derived by the linear dielectric approach in Ref [2]. As a result, the derivative approximation generates a sharper change in slope at the transition between lower and higher fields; this directly reflects the nonlinear behavior of the

polarization observed in RFPs. On the other hand, both the linear dielectric approximation and the derivative approximation follow a similar trend at high fields. In fact, the permittivity value found from least-errors for the derivative method is 12.9, which is very close to the reported value of 13 in Ref [2]. The amorphous regions dominate the high field response of the field and act like linear dielectrics, and thus the linear dielectric approximation is more accurate at higher field strengths. Similarly, Ref [29] fits the same biphasic model to the permittivity data calculated by the linear dielectric method in Ref [2], and their reported amorphous permittivity value is 12.



Figure 6-2. The relative permittivity (dielectric constant) is plotted versus electric field. The derivative approximation is obtained from the discretized points from the data set in Ref [2], and the linear dielectric approximation is the set of calculated constants based on the linear dielectric energy density equation. The derivative curve fit is the least-error biphasic model to predict the high field permittivity values based on the derivative approximation.

Since the very high field polarization responses of some of the terpolymers taken from literature are not available, the point-wise derivative approximation is more convenient to use, as it can directly predict high field permittivity based on low field data sets. Figure 6-3 shows the derivative approximation method performed on two sets of data from Ref.'s [20] and [16]. The biphasic model is then fitted to the data obtained from the derivative approximation method.



Figure 6-3. The dielectric constant versus electric field response is extracted from the polarization responses found in (a) Ref [20] and (b) Ref [16]. The high field dielectric constant is 7.7 for (a) and 11.7 for (b).

In addition to these two sets of data, experimental measurements for P(VDF-TrFE-CTFE) 61.8/30.4/7.8% are also available. The derivative approximation method is applied to the polarization data to obtain the dielectric constant at various fields, and the results are shown in Figure 6-4. However, since this material exhibits a strong double hysteresis loop (DHL), the biphasic model borrowed from Ref [29], which is suited for a single hysteresis loop (SHL), does not predict the permittivity of the material very well *at low fields*. As a result, the least-error high field relative permittivity is almost exactly 1. The purpose of this analysis is to obtain this high permittivity of the amorphous phase, which is the medium in which the dipoles exist. The biphasic model should thus predict the permittivity better at higher fields in order to obtain an accurate permittivity of the amorphous phase.



Figure 6-4. The dielectric constant for P(VDF-TrFE-CTFE) 61.8/30.4/7.8%, as calculated by applying the derivative method to experimentally measured values of polarization versus electric field. The amorphous phase's relative permittivity, ε_a , is 1.

The biphasic model in (4-115) is fitted into the data set without the low field response, as shown in Figure 6-5. The biphasic model is fitted for a range of responses based on the standard deviation calculated from multiple experiments. The model is fitted for one standard deviation above the averages, and one standard deviation below. The fitted parameters reveal that the relative permittivity ε_a must be between 4 and 14 for the data to lie within a standard deviation from the average permittivities. In addition, the model is fitted a third time based on the average values of permittivity calculated by the derivative approximation, which results in $\varepsilon_a = 9.4$.

Finally, all of the values for ε_a obtained from fitting the biphasic model in (4-115) to the permittivities calculated by the derivative approximation method are listed in Table 6-1. The P(VDF-TrFE)-based terpolymers exhibit, on average, an ε_a between 7.7 and 13. However, the statistical analysis in Figure 6-5 implies that it is possible for ε_a to reach values up to 14 and higher, as well as drop to 4 and below.



Figure 6-5.The dielectric constant for P(VDF-TrFE-CTFE) 61.8/30.4/7.8%, as calculated by applying the derivative method to experimentally measured values of polarization versus electric field for only the high field response. The biphasic model is fitted to both one standard deviation above and below the average experimental values and fitted again to the average measurements.

High field permittivity	
13	
12	
12.9	
7.7	
11.7	
9.4	

Table 6-1.Terpolymer high field permittivities are recorded based on least-squaring of the biphasic model to the derivative approximation data.

B.2. Obtaining the saturation polarization parameters for the dipoles

The remaining electrostatic parameters are the saturation electric field for the dipole orientations, E_{sat} and the saturation polarization, P_s . To obtain these values, the biphasic polarization model is no longer viable, because most EAPs are known to exhibit breakdown at high fields[45], so the low field response of the material is more relevant, which is dominated by dipole-dipole interactions. The application of the biphasic model was strictly to gain the high-field permittivity property of the material.

A simple approach to extracting E_{sat} is by constructing a polarization model based on Ref [29] with only a single term that entirely corresponds to the dipole-dipole interactions within the crystalline phase. First, recall the definition of polarization density, shown in (6-15).

$$P = \frac{\langle p \rangle}{\Delta V} \tag{6-15}$$

The average dipole moment $\langle p \rangle$ can be split into its magnitude *p* and its orientational component $\langle \cos \theta \rangle$. As a result, (6-15) can be rewritten as (6-16).

$$P = \frac{p(\cos\theta)}{\Delta V} \tag{6-16}$$

In (6-16), *p* is the magnitude of the net dipole moment over the small change in volume, ΔV . Thus, when the dipoles within the volume saturate at high fields, $\langle \cos \theta \rangle \rightarrow 1$. Assuming that any ΔV contains approximately the same number of dipoles (though not necessarily uniformly distributed), the relation in (6-17) must hold true.

$$\lim_{E \to \infty} \frac{p_{\Delta V} \langle \cos \theta \rangle}{\Delta V} = P_s \tag{6-17}$$

The parameter P_s is the same saturation polarization as the one found in the previous section. Thus, the final macroscopic polarization function is written as shown in (6-18).

$$P = P_{\rm s}(\cos\theta) \tag{6-18}$$

The expression for the average cosine $(\cos \theta)$ defined in Chapter 3 is substituted, which is a function of E_{sat} . The plots in Figure 6-6 display the polarization model in (6-18) fitted to experimental data by the least-squares method, and the optimized parameters are listed in Table 6-2.

Table 6-2. The optimized parameters from the single phase model for polarization

Material	$P_s\left(\frac{C}{m^2}\right)$	$E_{sat}\left(\frac{MV}{m}\right)$
P(VDF-TrFE-CFE) 62/38/4%	0.122	59.0
P(VDF-TrFE-CTFE) 65/35/10%	0.0733	31.1
P(VDF-TrFE-CTFE) 61.8/31.4/7.8%	0.0722	38.7



Figure 6-6. The polarization response and the fitted single phase polarization model are compared for (a) P(VDF-TrFE-CTFE) 61.8/30.4/7.8%, (b) P(VDF-TrFE-CFE) 62/38/4%, and (c) P(VDF-TrFE-CTFE) 65/35/10%. The optimal parameters are listed in Table 6-2.

APPENDIX C

Details of Modeling Methodology

C.1 Mathematical Formulation: Part I – Constitutive Response

The active response of electrostrictives is often modeled as an electric-polarization induced strain which is in turn integrated into a standard generalized small stress-strain response [81]. The small strain formulation is appropriate when the expected radii of curvature are moderate and the bimorph thickness is also small, resulting in small bending strains on outermost fibers which remain within the linear stress-strain response [76]. Such is the expected case for the bimorphs studied herein. The resulting response can be seen as the sum of elastic strains, ϵ_e , and electrostrictive strains, ϵ_E , that work to produce a combined stress, σ , given a compliance tensor, C.

$$\boldsymbol{\sigma} = \boldsymbol{C}: (\boldsymbol{\epsilon}_e + \boldsymbol{\epsilon}_E) \tag{6-19}$$

The elastic strain is the measurable, physical strain observed in the material while the electrostrictive strain is the strain induced by the electric field's development of polarized regions within the material. The latter strain is often represented in the uniaxial form of [82].

$$\epsilon_E = -\alpha E^2$$
 , (6-20)

where the negative sign ensures compression, α is the electrostrictive constant, and E is the electric field. In this form, and in the remainder of this work, it is assumed that $\alpha = \alpha_{31}$, $E = E_3$, ϵ_E acts in the uniaxial direction (which is \hat{e}_1), and $\epsilon_e = \epsilon_1$.

Development of equilibrium equations in the following section will require the onedimensional form for the constitutive equations which, assuming purely one-dimensional behavior, follow from equations (6-19) and (6-20) as

$$\sigma_{TP} = E_{TP}(\epsilon_e - \alpha E^2) \tag{6-21}$$

181

where σ_{TP} and E_{TP} are the stress and modulus in the terpolymer, respectively.

The constitutive response of MAE materials has been modeled by several researchers, however prior emphasis has been placed on soft-magnetic response [83[90] whereas the barium hexaferrite particles used here are hard-magnetic materials which have seen considerably less study [91]. The difference is critical when determining the effect of the particles on the matrix and the larger bimorph structure. Within a material, a magnetic dipole, M, subjected to an external magnetic field, H, will experience a magnetic torque $T = M \times H$. However, in soft magnetic materials M and H are collinear and hence T = 0. Therefore, magnetic torque will only be produced in materials with hard magnetic behavior. While it is true that strong geometric anisotropy may misalign M from H even in soft magnetic materials, experiments have shown that typical soft magnetic particles (e.g. carbonyl iron) are roughly spherical and hence produce no magnetic torque. Additionally, while magnetostriction is expected in MAE materials, the bulk deformation produced by the magnetic torque in MAE composites is orders of magnitude larger than typical magnetostrictive strains [80[92].

Given the prominence of magnetic torque based deformation over magnetostrictive deformation in MAE materials, and their use as active material "patches" in the geometries proposed below, MAEs will be modeled as loci of external body torques following

$$\boldsymbol{\tau} = \boldsymbol{m} \times \boldsymbol{H} \,, \tag{6-22}$$

where τ is magnetic torque density, and m is the magnetic dipole density. This torque density will be added to the formulation when developing equations for the equilibrium of moments in the bimorph. Incorporation of MAE behavior as an external load, specifically body forces in the case of soft-magnetic MAEs has precedence in the literature [93]. Hence, extension of this technique to magnetic torques is employed here.

Previous works predicated on the behavior of soft-magnetic responses are not capable of generating the mechanism of magnetic-torque necessary to model MAE with BaM particles.

Methods based on soft-magnetic behavior, while able to model magnetostrictive effects (eg. soft magnetic behavior) cannot accommodate magnetic torque based responses. These methods while able to model magnetostrictive effects (e.g. soft magnetic behavior) cannot accommodate magnetic torque based responses. The experimental stress-strain response ultimately seen in experiments remains in the linear stress-strain regime for the MAE material, hence Hooke's Law is used in this work to model MAE response as

$$\sigma_{MAE} = E_{MAE} \epsilon_e \tag{6-23}$$

where σ_{MAE} and E_{MAE} are the stress and modulus of the MAE material, respectively.

C.2 Mathematical Formulation: Part II – Mechanics and Equilibrium within the Bimorph Structure

The bimorph model consists of two layers of material, the terpolymer and the MAE, each having their own constitutive response bonded together at a continuous interface. In reality the two materials are bonded by a glue layer, however that layer is neglected in this work due to is relative thinness with respect to the overall bimorph geometry; formulation of a higher fidelity model incorporating the glue layer is in development. The geometry will be modeled as a one dimensional solid that conforms to prescribed bending kinematics transverse to the major axis and equilibrium of forces and moments throughout.

Given the previously mentioned assumptions, any arbitrary segment along the 1-D bimorph between two cut lines at x and $x + \Delta x$ must satisfy summation of forces

$$\frac{\partial N(x)}{\partial x} = 0 \tag{6-24}$$

and summation of moments

$$\frac{\partial M(x)}{\partial x} + \tau(x) * A_{MAE}(x) = 0$$
(6-25)

where $A_{MAE}(x)$ is the cross-sectional area of the MAE material at point *x*.

The stress-induced tractions along an arbitrary cut line at some point x yield the relationships to the normal force and moment, namely

$$N(x) = \Delta x \int_{\delta_1}^{\delta_2} \sigma(x, z) * t(x, z) * dz$$
(6-26)

And

$$M(x) = \Delta x \int_{\delta_1}^{\delta_2} z * \sigma(x, z) * t(x, z) * dz$$
(6-27)

where $z = \delta_1 \dots \delta_2$ signifies the range of integration through the height of the bimorph and z is measured from the base of the bimorph. Note that the integral will be evaluated over each layer in the bimorph, therefore the normal stress will be drawn from eqs. (6-21) or (6-23), respectively. Furthermore, the type of material may vary arbitrarily along any point x within a layer hence the x dependence in the stress function and bimorph thickness.

Finally, assuming kinematics of Bernoulli Euler bending, the elastic strain at some point x along the length of the bimorph can be expressed as

$$\epsilon_e(x) = -(z - \bar{z}(x)) * \kappa(x) \tag{6-28}$$

where $\bar{z}(x)$ is the height of the neutral axis from the base of the bimorph and $\kappa(x)$ is the radius of curvature.

Equations (6-21), (6-23) and (6-24)-(6-27) yield a set of differential equations which can be used to predict the local radius of curvature and height of the neutral axis given material parameters and the geometry of the bimorph.

C.3 Numerical Solution Techniques

The solution method to the first order differential system is a combination of analytical and numerical computations. The approach begins with the analytical solution to the neutral axis height from the bottom of the beam, i.e. \bar{z} . Since there are no external forces applied on the beam, this holds true across the entire length of the beam. As such, eqs. (6-24) and (6-26) yield

$$\bar{z} = A_1 + \frac{A_2}{\kappa} \alpha E^2 \tag{6-29}$$

where κ is the curvature, and $A_{1/2}$ are functions of the material properties and the geometry of each layer.

The substitution of eqs. (6-21), (6-23), and (6-27) into (6-25) yields a first order differential equation in $\kappa(x)$ that is solved with the forward finite difference method using the first order differential approximation

$$\frac{\partial \sigma_{\Omega}(x)}{\partial x} = \frac{\sigma_{\Omega}(x + \Delta x)}{\Delta x} - \frac{\sigma_{\Omega}(x)}{\Delta x}$$
(6-30)

In eq. (6-30), σ_{Ω} are the stresses of each layer of type Ω at an arbitrary point, and Δx represents an infinitesimal increment of the beam's length. Given eq. (6-30) the finite difference approximation of (6-25) can be represented as

$$0 = \int_{0}^{h_{A}} \left(\frac{\sigma^{A}_{x+\Delta x} - \sigma^{A}_{x}}{\Delta x} \right) z t_{A} dz$$

+
$$\int_{h_{A}}^{h_{B}} \left(\frac{\sigma^{B}_{x+\Delta x} - \sigma^{B}_{x}}{\Delta x} \right) z t_{B} dz + \tau h_{A} t_{A} \Delta x$$
(6-31)

where thicknesses t_A and t_B do not vary with x, and h_A and h_B are heights of the two layers. After integration, the resulting system of equations yields a finite difference scheme for the solution of $\kappa(x)$ discretized over the length of the bimorph. At each point *i*, (6-31) produces equations of the form

$$\psi_i(\kappa_{i+1},\kappa_i,C_i) = f(\tau_i) \tag{6-32}$$

where C_i is a function of parameters, A_i and factors from integration, and the desired curvatures κ_i , and the forcing term depends on the magnetic torque.

These equations can be rewritten in the matrix form below.

$$\begin{bmatrix} -\psi_i \ \psi_{i+1} & \cdots & 0\\ \vdots & \ddots & \vdots\\ 0 & \cdots & -\psi_n \end{bmatrix} \begin{bmatrix} \kappa_1\\ \cdots\\ \kappa_n \end{bmatrix} = \begin{bmatrix} f(\tau_i)\\ \cdots\\ f(\tau_n) \end{bmatrix}$$
(6-33)

the solution of which can be written as $\kappa = \psi^{-1} f(\tau)$.

The numerical component of this method arises when dealing with the torque strength as the MAE orients itself with the beam. If the magnetic torque is a constant ($\vec{\tau} = \vec{m} \times \vec{H}$), the linear relation with the external field results in a beam that bends past the alignment with the field. Accordingly, a numerical solution is required to adjust the torque as the beam aligns with the field. The cross product in 2 dimensions can also be presented as (6-34).

$$\tau = mHsin\theta \tag{6-34}$$

therefore, $\tau = \tau(\theta)$, where θ is the angle between the MAE and the field H. In addition to this dependence, θ is also a function of x, the distance along the beam, since the alignment of the MAE will vary as x changes. One solution to this problem is an iterative process that calculates θ at each point and adjusts the torque for the next point. A basic outline of this process is illustrated in Figure 6-7 below. Also, note that the angle in (6-34) is redefined as the angle θ' shown in Figure 6-8, and the form of (6-34) is changed to (6-35) for convenience in calculations.

Step 1: Initially solve for all K's with $\theta' = 0$ and
store K_1 (K at i=1)
Step 2: Find the angle θ' at i=1 and store θ'
Step 3: Calculate new τ as defined in (6-34)
Step 4: Solve for all K's again and store K_2
Step 5: Find the new angle θ' again and add it to
the stored θ'
Repeat steps 3-6 until i=n

Figure 6-7. The procedures of the iterative solution method for the curvatures with varying angles.

$$\tau = MHcos(\theta') \tag{6-35}$$

The resulting curvatures will always be adjusted according to their orientation with the field, since the cosine term will reduce the torque to zero as the angle approaches 90 degrees.

The next step is to convert the curvatures to coordinate locations, for which we will follow the approach used in [94]. The curvature is initially defined as below.

$$K = \frac{d\theta}{ds} \tag{6-36}$$

Using the separation of variables technique, this first order DE is set up to be solved as shown below.

$$ds = \frac{1}{K}d\theta \tag{6-37}$$

In (19), ds represents an infinitesimal arclength, which is a straight line (linear) under the assumption that ds is a very small distance. The following relations are derived from Figure 6-8.

$$\frac{dz}{ds} = \sin\theta \rightarrow ds = \frac{dz}{\sin\theta}$$

$$\frac{dx}{ds} = \cos\theta \rightarrow ds = \frac{dx}{\cos\theta}$$
(6-38)

Therefore, ds can be replaced with the above relations in (6-39).

$$\int_{0}^{\delta_{z}} dz = \int_{0}^{\theta_{0}} \frac{\sin\theta}{K} d\theta$$
 (6-39)

The integration generates the displacement in the z-direction, δ_z .

$$\delta_z = \frac{1 - \cos(Kl)}{K} \tag{6-40}$$

And similarly,

$$\delta_x = l - \frac{\sin(Kl)}{K} \tag{6-41}$$

The relation $\theta = Kl$, derived from (6-36), is used in the equations above, where l is an

incremental length between two points of interest.



Figure 6-8. An infinitesimal segment along the arc, which produces a linear relation between dx and dz.

The displacements are then used to determine the local coordinate components X_{t_i} and Z_{t_i} of each point in the deformed configuration. These coordinates are defined below in (6-42).

$$\begin{bmatrix} \cos\theta_{i-1} & -\sin\theta_{i-1} \\ \sin\theta_{i-1} & \cos\theta_{i-1} \end{bmatrix} \begin{bmatrix} X_{t_i} \\ Z_{t_i} \end{bmatrix} = \begin{bmatrix} l_i - \delta_{x,local_i} \\ \delta_{z,local_i} \end{bmatrix}$$
(6-42)

The above matrix must be calculated at each point, i, which will be used to determine the global coordinates below.

$$X_i = X_{t_i} + X_{t_{i-1}} \tag{6-43}$$

$$Z_i = Z_{t_i} + Z_{t_{i-1}} \tag{6-44}$$

APPENDIX D

Electrostriction Model's MATLAB Code

```
% Following program computes a microstructure based model
clc, clear
Emax = 100000000; % Must be in multiples of 10000000
Estep = 1000000;
Efieldvec = linspace(Estep,Emax,Emax/Estep); % electric field vector to
be used for plotting
stretchvals = zeros(Emax/Estep,1);
tic
% Known Parameters
Ps = 0.07;
Crystallinity = 1;
epsr = 12;
mu = 0;
C1 = 15000000;
ms = 2.5; % stretch at which the polymer chain network becomes locked
% Variable Parameters
Esat = 35000000;
muGauss1 = 0;
muGauss2 = 0;
Sigma1 = 0.06;
Sigma2 = 0.01;
% Code parameters
a = 16; % number of steps for phi (0 <= phi <= pi
b = 2*a; % number of steps for the (0 <= the <= 2*pi)
xx1 = linspace(-pi,pi,100);
Nsamples = 1000000; % actual sample size? double check if n is actually
used
muGauss = [muGauss1 muGauss2]; % mean vector
Sigma = [Sigma1 0; 0 Sigma2]; % deviance matrix
xxy1 = 0:pi/a:pi; xxy2 = 0:pi/a:2*pi; % first and second variables,
respectively
[X1,X2] = meshqrid(xxy1,xxy2);
for Efield = Estep:Estep:Emax
b1 = bfun(Efield,Esat,mu); % deviance parameter
b2 = 0;
pdf = double(vonMises(b1,mu,xx1));
pdf2 = double(vonMises(b2,mu,xx1));
% Find the cdf of the pdf. (NOTE: alternate method for this is using the
% cdf command within MATLAB
n = size(pdf);
vonMisescdf = (1/sum(pdf))*cumsum(pdf);
```

```
vonMisescdf2 = (1/sum(pdf2))*cumsum(pdf2);
% The following pseudorandom number generation method is based on an
% article written by John S. Denker
% for more information: http://matlabtricks.com/post-44/generate-random-
numbers-with-a-given-distribution
% and http://www.av8n.com/physics/arbitrary-probability.htm
% References
% John Denker, "Measurements and Uncertanties versus Significant Digits"
   " ", "Models and Pictures of Atomic Wavefunctions"
8
         " , "Introduction ot Probability"
9
% remove non-unique elements
[vonMisescdf, mask] = unique(vonMisescdf);
xx1 = xx1 (mask);
[vonMisescdf2, mask] = unique(vonMisescdf2);
xx2 = xx1(mask);
% create an array of 2500 random numbers
randomValues = rand(1, Nsamples);
randomValues2 = rand(1, Nsamples);
\% inverse interpolation to achieve P(x) -> x projection of the random
values
projection = interp1(vonMisescdf, xx1, randomValues);
projection2 = interp1(vonMisescdf2, xx1, randomValues2);
cosangle = zeros(1,Nsamples/2);
for i = 1:Nsamples/2
    cosangle(1,i) = cos(projection(i)-projection(2*i));
end
cosangle(isnan(cosangle)) = 0;
fprintf('average angle:\n')
avgangle = mean(cosangle)
% Next, compare to vectors
dipolemoments = zeros(Nsamples, 3);
dipolemoments(:,1) = cos(projection2).*sin(projection);
dipolemoments(:,2) = sin(projection).*sin(projection2);
dipolemoments(:,3) = cos(projection);
averages = sym(zeros(a,1));
% Dot product with vectors ranging from -pi to pi (due to axisymmetry,
we
% can ignore dot product with vectors that have varying theta
orientations
% Additional note: effective cosine mentioned before is supposed to be
```

```
% cos(phi) here, using the notation below. So the paper's theta = our
phi.
syms lambda q
assumeAlso(lambda, 'real')
assumeAlso(lambda > 0)
averages = averagesfun(Efield,Esat,mu,Nsamples,a,b);
averagesmat= zeros(b+1,a+1);
for i = 1:b+1
    averagesmat(i,:) = averages;
end
% Next, consider the energy equation:
% Udd = mu^2/(4*pi*eps)*(cos(thetal-theta2) - 3*(cos(phi-theta))^2/r^3)
syms the phi
assumeAlso(the, 'real')
assumeAlso(0 <= the <= 2*pi)</pre>
assumeAlso(phi, 'real')
assumeAlso(0 <= phi)</pre>
assumeAlso(phi <= pi)</pre>
eps0 = 8.854187*10^{-12};
r0 = 1; % original radius of RVE
r = r0*sqrt((lambda.^(-1/2).*sin(X1)).^2 + (lambda.*cos(X1)).^2); %
magnitude of the radius of RVE as function of stretch
Vd = Crystallinity*(4/3)*sym(pi)*r0^3; % volume of crystals - constant
because det J = 1
Vtot = (4/3) *sym(pi) *r0^3; % volume of total RVE - constant because detJ
= 1
dipmoment = Vd*Ps;
Udd = simplify(dipmoment^2./(Vtot.*4.*pi.*eps0.*epsr).*(avgangle -
3*averagesmat.^2)./r.^3);
% Apply multi-variate normal distribution for spatial distribution of
% particles (dipoles)
F = mvnpdf([X1(:) X2(:)],muGauss,Sigma);
F1 = reshape(F, length(xxy2), length(xxy1));
toc
sinvec = sin(0:pi/a:pi);
sinmat= zeros(b+1,a+1);
for i = 1:b+1
    sinmat(i,:) = sinvec;
end
Nf = sum(sum(F1.*sinmat*sym(pi)/a*sym(pi)/a))
EnergyMatrix = F1.*Udd.*(sym(pi)/a)*(sym(pi)/a).*sinmat;
TotalDipoleEnergy = (1/Nf) *sum(sum(EnergyMatrix));
% TotalDipoleEnergy = Udd(1,1);
```

```
% Next, we compute Cauchy stress
F = [lambda^{(-1/2)} 0 0; 0 lambda^{(-1/2)} 0; 0 0 lambda ]; % deformation
gradient
B = F*F; % did not do transpose because 1) [F] is a symmetric matrix,
and 2) MATLAB is being weird with transposes
        % the actual definition of B is: B = F^*F^T
% Cauchy stress for elastic energy
Tke = simplify(2*diff(Uelasticfun(C1,ms),lambda)*B);
Tkd = 2*diff(TotalDipoleEnergy,lambda)*B; % Cauchy stress related to
dipole-dipole energy
% Tkd = 2*diff(Udd(1,9),lambda)*B; % D-D energy for vertical chain
Tk = Tkd + Tke + q*eye(3); % eye(3) is the Identity matrix of 2nd order,
\dim(I) = 3
% next, solve for q
toc
qq = solve(Tk(3,3) == 0,q);
toc
Tk = Tkd + Tke + qq*eye(3); % Cauchy stress with q plugged in
syms x2
divT = diff(subs(Tk(2,2),lambda,(r0^2)/(x2^2)),x2);
divT = subs(divT, x2, r0*lambda^(-1/2));
toc
stretchvals(Efield/Estep) = vpasolve(divT == 0,lambda,1)
toc
end
% Convert stretch to strain
% pseudo-code: (stretch - 1)*100 = strain
strainvals = (stretchvals - 1).*100*(-1) % multiply by -1 so it looks
positive
strainvals2 = zeros(Emax/Estep + 1,1);
strainvals2(2:Emax/Estep+1) = strainvals;
Efieldvec2 = zeros(Emax/Estep+1,1);
Efieldvec2(2:Emax/Estep+1) = Efieldvec;
```

```
plot(Efieldvec2,strainvals2)
xlabel('Electric Field (MV/m)')
ylabel('-Strain (%)')
```

APPENDIX E

MATLAB Bimorph Code

```
% Program: 1D bimorph model with 3+ layer feature
% UPDATED ON 7/31/2016
% Log:
   Changes made to the following features
8
   (1) fmatrix fixed - appropriately subs in B.C. curvature and leaves
8
the
   rest at zero (unless there is an extra term in the sum of moments
8
   equation developed for the differential element
8
   (2) alpha*v^2 term written as negative, which fixes direction of
8
9
   curvatures (not 100% sure why)
9
   Other updates:
   (1) Verified all curvature calculations on Mathematica (for 2 layer
8
8
   segment)
9
    (2) Verified all boundary condition calculations on Mathematica (for
2
8
   layer segment)
00
   (3) Verified curvature calculations on Mathematica for 3 layer
segment
2
% UPDATES ON 8/16/2016
   (1) Found possible source of convergence issue: fmatrix1 was being
8
   multiplied by (q/Hsteps) at the beginning of each loop, but the loop
8
   itself solves for each element of that matrix, so it is not at
8
8
    equilibrium. Instead, the invidivual components will be multiplied
by
   the factor before the nested loop.
8
8
    (2) Coefficients are now properly being taken out of totalmoments.
CK1,
   CK2, Cf cells and their respective 2D arrays have been added.
8
8
   Appropriate changes have been made to the stiffness matrix.
8
% Updates on 8/31/2016
   (1) K is no longer solved consecutively for each point starting from
8
    the fixed end. Instead, the entire equation is calculated for K's,
8
and
   the angle at every point is calculated. These angles are then
8
8
   cumulatively added to represent the total angles, which are then
8
   plugged back into the fmatrix1.
8
8
   Updates on 9/1/2016
8
   (1) On initial fmatrix setup, changed Cfm(y) to Cfm(i), so the
correct
00
   Cfm element is assigned to each part of the fmatrix
8
8
   Updates on 9/2/2016
8
   (1) Changed the first calculation of Kmatrix to match the iteration
   steps when Hsteps is introduced for the 2nd part
8
8
   (2) Completely got rid of initial Kmatrix calculation. Fixed initial
8
   setup such that the iteration has something to start with (zeros for
9
   curvatures, angles, etc).
8
   (3) Found error in totalmoment equation: the negative was outside of
а
```

```
8
  paranthesis, instead of being inside (such that it is stress2 -
2
  stress1)
hold off
clc, clear
tic
S = 5; % Segment numbers
n = 200; % Number of FDM points
L = 0.03; % [m] Length of total beam
H = -5; % [A/m] External magnetic field strength
Efield = 0;
t = 0.01; \% [m]
hstiffener = 0.000001
% ADVANCED SETTINGS
maxiterations = 500
Hsteps = 2 % number of iterations for the loop below
% Enter properties in following order: [Pa] stiffness, # of layers,
% [m] thickness, [m] height, electrostrictive coefficient,
% [V/m] electric field, [T] magnetization
Lengths = [0 0.32 0.35 0.66 0.69 1]; % fraction of distance of starting
point of
2
                                                            each segment
from the fixed end of the beam
% Saad's parameters
props = cell(1, S);
props{1} = [5000000, 2, t, 0.00003,0,0,0; 2000000, 2, t, 0.00003, 3*10^-
18, Efield, 0];
props{2} = [5000000, 6, t, 0.00003, 0, 0, 0; 1000000, 0, t, 0.000005, 0, 0, 0]
0; 2000000, 0, t, 0.00003, 3*10^-18, Efield, 0; 1000000, 0, t, 0.000005,
0, 0, 0; 2000000, 0, t, 0.00003, 3*10^-18, Efield, 0; 1000000, 0, t,
0.000005, 0, 0, 0];
props{3} = [5000000, 2, t, 0.00003,0,0,0; 2000000, 0, t, 30*10^-6,
3*10^-18, Efield, 0];
props{4} = [5000000, 3, t, 0.00003, 0, 0, 0; 1000000, 0, t, 0.000005, 0, 0]
0; 2000000, 0, t, 30*10^-6, 3*10^-18, Efield, 0];
props{5} = [1600000000, 2, t, 0.00006,0,0,0; 200000000, 0, t, 30*10^-6,
3*10^-18, Efield, 0];
    % Call on layer number per "segment" as shown below
    props{1,3}(1,2);
```

C = cell(1, S);

```
CK1 = cell(1, S);
    CK2 = cell(1, S);
    Cf = cell(1, S);
    CK1m = zeros(1, S);
    CK2m = zeros(1, S);
    Cfm = zeros(1, S);
    KBCtot = cell(1, S);
for y = 1:S
        % Next, define the stress terms
        syms z zbar K K2 zbar2 theta qdup
        % determine heights for integration
        heights = zeros (props \{1, y\} (1, 2), 1);
            for i = 1:props{1,y}(1,2)
                if i < 2
                     heights(i) = props\{1, y\}(i, 4);
                else
                     heights(i) = props\{1, y\}(i, 4) + heights(i-1);
                end
            end
            heights;
        % Define stresses:
        stress = -(props{1,y}(:,1)).*(z - zbar)*K +
(props{1,y}(:,1)).*props{1,y}(:,5).*props{1,y}(:,6).^2
        stress2 = -(props{1,y}(:,1)).*(z - zbar2)*K2 +
(props{1,y}(:,1)).*props{1,y}(:,5).*props{1,y}(:,6).^2
        % Sum forces to find zbar
        totalforce = 0;
            for k = 1:props{1,y}(1,2)
                if k < 2
                     totalforce = totalforce +
props{1,y}(k,3).*int(stress(k),z,0,props{1,y}(k,4))
                else
                totalforce =totalforce +
props\{1, y\}(k, 3). *int(stress(k), z, heights(k-1), heights(k))
                end
            end
             % Solve for the neutral axis position, zbar
        zbar1 = solve(totalforce==0, zbar)
            % Sum forces to find zbar2
        totalforce = 0;
```

```
for k = 1: props\{1, y\}(1, 2)
                if k < 2
                    totalforce = totalforce +
props\{1, y\}(k, 3). *int(stress2(k), z, 0, props\{1, y\}(k, 4));
                else
                 totalforce =totalforce +
props\{1, y\}(k, 3). *int(stress2(k), z, heights(k-1), heights(k));
                end
            end
        % Solve for the neutral axis position, zbar2
        zbar3 = solve(totalforce==0, zbar2)
        % Next, solve for the boundary condition (i.e., electrostrictive
        % influence)
        totalmomentBC = 0;
            for p = 1:props{1,y}(1,2)
                if p < 2
                     totalmomentBC = totalmomentBC +
props{1,y}(k,3).*int(subs(stress(p),zbar,zbar1).*z,0,props{1,y}(p,4));
                else
                     totalmomentBC = totalmomentBC +
props{1,y}(k,3).*int(subs(stress(p),zbar,zbar1).*z,heights(p-
1), heights(p));
                end
            end
        KBC = solve(totalmomentBC==0,K)
        % Store BC into global BC curvatures, KBCtot
        KBCtot{y} = KBC;
        % Next, set up sum of moments equation for recurrence solver
        totalmoment = 0;
            for p = 1:props{1,y}(1,2);
                if p < 2
                    totalmoment = totalmoment + props{1,y}(k,3).*int((-
subs(stress(p),zbar,zbar1) +
subs(stress2(p),zbar2,zbar3)).*z,0,props{1,y}(p,4)) +
H*props{1,y} (p,3) *props{1,y} (p,4) * (L/n) *props{1,y} (p,7);
                 else
                     totalmoment = totalmoment + props{1,y}(k,3).*int((-
subs(stress(p),zbar,zbar1) + subs(stress2(p),zbar2,zbar3)).*z,heights(p-
1), heights (p)) +
H*props{1,y} (p,3) *props{1,y} (p,4) * (L/n) *props{1,y} (p,7);
                end
            end
totalmoment
```

```
C{y} = coeffs(totalmoment) % obtain the coefficients of the
moment equation to be used in the FDM below
        CK1{y} = coeffs(totalmoment, K);
        CK2{y} = coeffs(totalmoment, K2);
        Cf{y} = coeffs(totalmoment);
        CK1m(y) = CK1\{y\}(1,2);
        CK2m(y) = CK2\{y\}(1,2);
        if size(Cf{y}) < 3
            Cfm(y) = 0;
        else
            Cfm(y) = Cf\{y\}(1,1);
        end
end
% Forcing terms
fmatrix = [];
for i = 1:S
    fmatrix = vertcat(fmatrix, repmat(-
Cfm(i)*(qdup/Hsteps)*cos(theta),round(n*(Lengths(i+1)-Lengths(i))),1));
% qdup is a duplicate of q, which will be subbed in later on, while
Hsteps remains the same
    Cfm(i)
end
fmatrix(1) = 0;
if double(KBCtot{S}) == 0
    fmatrix(n) = 0.00001
else
fmatrix(n) = double(KBCtot{S});
end
forcingtermsdone = 1.000
% Code for the stiffness matrix
stiffness = zeros(n);
for q = 1:S
    if q < 2
        for i = 1:round(Lengths(q+1)*n);
            for j = 1:round (Lengths (q+1) *n);
                if i==j
                stiffness(i,j) = CK1m(q);
                else
                end
                if i+1 == j
```

```
stiffness(i,j) = CK2m(q);
                else
                end
            end
        end
    else
        for i = round(Lengths(q)*n):round(Lengths(q+1)*n)
            for j = round(Lengths(q)*n):round(Lengths(q+1)*n)
                if i==j
                stiffness(i,j) = CK1m(q);
                else
                end
                if i+1 == j
                stiffness(i,j) = CK2m(q);
                else
                end
            end
        end
    end
end
for i = 1:S
stiffness(round(Lengths(i+1)*n),round(Lengths(i+1)*n)) = CK1m(i);
end
stiffness(1,1) = 1;
stiffness(1,2) = 0;
stiffness(n,n) = 1;
% Addition for the BC's at the segment interfaces (do not need now)
%for i = 1:(S-1)
8
   stiffness(round(Lengths(i+1)*n), round(Lengths(i+1)*n)) = 1;
%fmatrix(round(Lengths(i+1)*n)) = double(KBCtot{i});
% end
% End of BC addition
stiffnessmatrixcomplete = 1.00
format shortEng
format compact
Kmatrix = zeros(1, n);
```

```
thetamatrix = zeros(n, 1)
fmatrix1 = zeros(n,1);
thetacell = cell(Hsteps, 1);
thetacell{1,1} = thetamatrix;
kcell = cell(Hsteps,1)
kcell{1,1} = Kmatrix;
fmatrixcell = cell(Hsteps, 1)
thetai = 0;
fcell=[] % create this to prevent failure of converting from double
for j = 1:n
        fcell{1} = subs(fmatrix(j), theta, thetacell{1,1}(j));
        fmatrix1(j) = subs(fmatrix1(j),qdup,1);
end
% Initial setup used as information for first calculation of the
iterations
% below
allkmatrices = [];
allfmatrices = [];
fmatrixcell{1,1} = fmatrix1;
toc
thetacollective = []
allsmatrices = []
for q = 1:Hsteps
    s matrix = zeros(maxiterations,1);
    s matrix(1,1) = 50;
    z = 1
    while s matrix(z,1) >= 1
    thetai = 0;
        Kmatrix = double(stiffness\fmatrix1); % fmatrix is defined from
before, and stiffness is constant
        kcell{z+1,1} = Kmatrix; % store Kmatrix into kcell for later use
        thetatemp = Kmatrix.*(L/n); % obtain temporary thetas at each
point
        thetacell{z+1,1} = cumsum(thetatemp); % total theta at each
point
```

 $\ensuremath{\$}$ loop below recalculates fmatrix1 based on angles found above

```
for k = 1:n
        fcell{k} = subs(fmatrix(k),theta,thetacell{z+1,1}(k)); % fmatrix
always in the most updated form, while thetacell is used in the error
function
        fmatrix1(k) = subs(fcell{k},qdup,q);
        end
        fmatrixcell{z+1,1} = fmatrix1;
        kcell{z+1,1} = Kmatrix;
        for i = 1:n
            s matrix(z+1,1) = s matrix(z+1,1) + (kcell{z+1,1}(i) -
kcell{z,1}(i)).^2;
        end
    s matrix(z+1,1) = sqrt(s matrix(z+1,1)/n);
       % if s matrix(z+1,1) - s matrix(z,1) > 0
        % restart same q from this last thetacell
        % s matrix(z+1,1) = 0.01;
         % for j = 1:n
         00
                 fmatrix1(j) = subs(fmatrix(j),theta,thetacell{z,1}(j));
         % end
         %else
        % end
    z = z + 1
    end
    a
    thetacollective{q,1} = thetacell;
    allsmatrices{q,1} = s matrix;
    allkmatrices{q,1} = kcell;
    allfmatrices{q,1} = fmatrixcell;
end
iterativesolutioncomplete = 1.00
l = L/n;
Kmatrix(1) = 0.000001; % replaces first curvature with a very small
number to
                       allow code to solve a linear system below (i.e.
8
to
0
                       avoid singular matrix)
angle = Kmatrix*1;
globalangle = cumsum(angle);
```

```
xdisp = 1 - (sin(Kmatrix.*1))./(Kmatrix);
ydisp = (1./Kmatrix).*(1 - cos(Kmatrix.*l));
xandydispcomplete = 1.00
syms Xglobal Yglobal
Xglobali = zeros(1,n);
Yglobali = zeros(1,n);
for i = 1:n
qq = Xglobal == (l - xdisp(i) -
sin(globalangle(i)).*Yglobal)/cos(globalangle(i));
zz = Yglobal == (ydisp(i) +
sin(globalangle(i)).*Xglobal)/cos(globalangle(i));
[A, B] = equationsToMatrix([qq,zz], [Xglobal, Yglobal]);
XY = double(linsolve(A,B));
Xglobali(i) = XY(1);
Yglobali(i) = XY(2);
end
globaldisplacementscomplete = 1.00
FinalX = cumsum(Xglobali);
FinalY = cumsum(Yglobali);
axis square
plot(FinalX,FinalY)
axis([-L L -L L])
toc
```

ANIL EROL

EDUCATION

The Pennsylvania State University PhD in Mechanical Engineering

The University of Kentucky

B.S. in Mechanical Engineering, Magna Cum Laude

709-3 W Cherry Ln, State College, PA 16803

Aug 2014 – Aug 2019 (Expected)

Aug 2010 - Aug 2014

Jun 2014 - Aug 2019

RESEARCH EXPERIENCE

Magneto-Active Composites and Structures (MACS) Lab

Campus: University Park, The Pennsylvania State University

Advisor: Dr. Paris von Lockette

- **Dissertation title**: Microstructure-Based Modeling of Electrostriction and Its Application In Analytical Modeling of Multi-Field Actuated, Origami-Inspired Multi-Layered Flexible Beams
- <u>Multi-Field Processing of Composites (Sponsored by NSF)</u>: I simulated particle dynamics (*via* ferrohydrodynamics) in polymer composites.
- <u>Design optimization of self-folding devices (Sponsored by NSF EFRI –</u> <u>Interdisciplinary ODISSEI Team):</u> I developed a composite beam model that accommodates magnetic torques, large displacements, and nonlinear material behaviors; I implemented design optimization via a NSGA-II genetic algorithm.
- <u>Modeling the mechanics of EAPs (Sponsored by NSF EFRI)</u>: I created a new modeling approach utilizing semicrystalline polymer micromechanics and statistical mechanics of polymer chains.

JOURNAL PUBLICATIONS

In Peraparation

3. Masud, A., **Erol**, **A**., Ounaies, Z., von Lockette, P. "Achieving Complex Microarchitectural Nanocomposites Via Non-Uniform Multi-Field Processing."

Submitted

 <u>Erol, A</u>, Frecker, M., von Lockette, P. "1D Modeling of Multi-Field, Multi-Layer, and Segmented Origami-Inspired Devices with Multi-Objective Optimization for Shape, Work, and Cost." Submitted to a special issue of *Smart Materials and Structures*, (2019). **Invited submission**.

Published

1. <u>Erol, A</u>., Ahmed, S., Ounaies, Z., von Lockette, P. "A Microstructure-Based Approach to Modeling Electrostriction that Accounts for Variability in Spatial Locations of Domains." *Journal of the Mechanics and Physics of Solids*, (2019).