Multilayer Dielectric Elastomer Actuators

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Abstract

Dielectric elastomers are a class of electroactive polymers able to exhibit large deformations when electrically stimulated. They are soft, rubber-like materials (hence the term elastomers), and can operate repeatedly and fast at high levels of efficiency. Moreover, dielectric elastomer actuators can easily fabricated in a large variety of shapes and geometries and the induced displacement can be designed to bend, stretch or contract. Since their functional response is similar to biological muscles, dielectric elastomers have been often referred as artificial muscle and have been considered as potential candidates for actuation of smart robotic systems.

The first part of this thesis describes the basic properties of Dielectric Elastomer Actuators. A single-layer geometry was used as a bench test to investigate on the properties of different dielectric elastomers, showing how the choice of the used materials greatly affects the final performances of the actuator. Compliant rubber electrodes were developed using mixtures of conducting carbon black and insulating elastomer above their percolating threshold. It was found that the developed rubber electrodes are not only useful as compliant electrodes for actuation purposes, but find also excellent application in pressure-sensing devices based on capacitive technology. Moreover, the good mechanical stability and adherence of the rubber electrodes to the dielectric elastomer suggested the possibility of developing multilayer dielectric elastomer actuator.

The second part of this thesis focuses on multilayer dielectric elastomer actuators. One practical limit of single-layer actuators is, in fact, the limited amount of force and displacement that it can be developed along the thickness direction. Moreover, one typical problem related to the actuation principle of macroscopic dielectric elastomer actuators is the high voltage
required, usually in the Kilovolt range, that imposes particular care in the insulation of the whole actuator from the surrounding environment. This high actuation voltage, however, can be drastically reduced if a thin film of dielectric elastomer is used. Despite this, the manufacture of a macroscopic stack-like actuator, starting from thin films of dielectric elastomer can present many manufacture difficulties, like the handling and the assembly of the films, the power connection to hundreds or thousands of layers, the presence of defects in one single layer that can cause the complete failure of the whole actuator.

To overcome these limitations, the second part of this thesis describes a novel fabrication process for the manufacture of multilayer DEAs. A semi-automatic machine able to fabricate multilayer DEAs of different shapes and lengths was designed and developed. The proposed fabrication process not only reduced considerably the fabrication time of the single actuator, but also enabled the possibility of reducing the thickness of the dielectric layers, down to 50um.

Finally, the fabricated prototypal actuators, operating at much lower voltages, were tested and characterized in terms of developed stress and strain, and their performances are discussed in relation to the experimental measurements obtained with single-layer actuators.
To my father
Contents

List of Figures vii

1 Introduction 1
   1.1 Why compliant actuators for robotic applications? ................. 1
   1.2 Survey of actuation technologies .................................. 3
      1.2.1 Traditional actuators .......................................... 3
      1.2.2 Introduction to polymeric actuators .......................... 7
         1.2.2.1 Ionic EAPs .................................................. 7
         1.2.2.2 Polyelectrolyte gels ...................................... 7
         1.2.2.3 Conducting polymers ...................................... 9
         1.2.2.4 Ionic Polymers Metal Composite .......................... 11
         1.2.2.5 Electronic polymers ...................................... 12
         1.2.2.6 Electrostrictive polymers ................................ 14
         1.2.2.7 Dielectric Elastomer Actuators ........................... 14
         1.2.2.8 Other EAPs ................................................ 15
         1.2.2.9 Performances of EAPs and comparison between differ-
                        ent actuating technologies ............................. 16
   1.3 Biological muscles as a model for compliant actuators ............ 20

2 Dielectric Elastomer Actuators 25
   2.1 Theory of elasticity applied to elastomers ......................... 25
      2.1.1 Hooke model .................................................. 25
      2.1.2 Neo-Hookean model .......................................... 32
      2.1.3 Mooney-Rivlin model ........................................ 34
      2.1.4 Ogden model ................................................ 36
# CONTENTS

2.1.5 Finite element modeling ........................................... 40
2.2 Dielectric properties of polymers ............................... 44
2.3 Operational principle of dielectric elastomer actuators .......... 48

3 Single Layer Dielectric Elastomer Actuators ............... 65
3.1 Fabrication procedure for single layer DEAs .................... 66
3.2 Dielectric material ...................................................... 67
  3.2.1 Silicone dielectric .............................................. 69
  3.2.2 Acrylic dielectric .............................................. 70
  3.2.3 Dielectric strength .............................................. 72
  3.2.4 Permittivity ...................................................... 76
3.3 Compliant electrodes .................................................... 76
  3.3.1 Types of electrodes .............................................. 78
  3.3.2 Dust electrodes .................................................. 79
  3.3.3 Grease electrodes .............................................. 81
  3.3.4 Rubber electrodes .............................................. 82
  3.3.5 Resistance ....................................................... 86
  3.3.6 Further applications of compliant electrodes ............... 87
3.4 Experimental results .................................................... 88
  3.4.1 Experimental Setup ............................................. 88
  3.4.2 Strain Measurement ............................................. 90
  3.4.3 Response Speed ................................................ 92
  3.4.4 Actuator failure mechanisms .................................. 94
  3.4.5 Reliability tests .............................................. 99

4 Multilayer Dielectric Elastomer Actuators ............... 105
4.1 Fabrication issues ...................................................... 106
4.2 Finite element analysis of multilayer actuators ............... 110
  4.2.1 Unconstrained case ............................................. 110
  4.2.2 Constrained case 1: length actuation mode .................. 117
  4.2.3 Constrained case 2: thickness actuation mode ............... 121
4.3 Setup for the semiautomatic fabrication of multilayer actuators ..... 127
  4.3.1 The winding unit .............................................. 129
  4.3.2 The spraying unit .............................................. 131
List of Figures

1.1 ICub(1), a humanoid robotic platform designed to study the development
of cognitive capabilities in artificial systems and humans. .................. 2
1.2 Examples of state-of-the art robots with advanced force control. From
left to right: the DLR’s humanoid ’Justin’, the DLR-III lightweight arm,
SARCOS and Big-Dog. ..................................................... 4
1.3 Examples of compliant electromechanical actuators: the Series-Elastic
Actuator, the Variable Stiffness Actuator and the AMACS. ............... 4
1.4 The parallel-distributed approach of the DM$^2$ actuator. Each robotic
joint is controlled by two motors: one, with high torque an low operating
frequency is positioned outside the structure of the manipulator; the
other, smaller and operating at high frequency is directly coupled to the
joint. ................................................................. 5
1.5 On the left, examples of of pneumatic actuators based on the McKibben
principle. On the right, a typical actuating configuration for two antag-
onistic pneumatic ’artificial muscles’. .................................. 6
1.6 Bending of a polyacrylic acid gel caused by the swelling of the material
on the anode side (top). .................................................. 8
1.7 Examples of different conducting polymers, with in evidence the sp2-
conjugation in their chemical structure. ................................. 9
1.8 Example of three different configurations of conducting polymer actu-
ators produced by EAMEX Corp. ........................................ 10
1.9 On the left: an adjustable optical aperture fabricated with a conveniently
shaped CP actuator. On the right: a polymeric-actuated image stabilizer
developed by EAMEX Corp. ................................................ 11
1.10 Operating principle of an IPMC actuator. The bending displacement is due to the movement of ions from the anode to the cathode of the device. 13
1.11 A demonstrative application of an IPMC gripper in action. 13
1.12 Comparison between EAPs, natural muscle and other actuation technologies. 17
1.13 Comparison between different actuator technologies in terms of developed strain and active stress. Performances of natural muscles are given as reference. 18
1.14 Comparison between different actuator technologies in terms of specific power and response bandwidth. Performances of natural muscles are given as reference. 19
1.15 Hierarchical structure of a mammalian skeletal muscles 20
1.16 The sarcomere as the functional unit of the muscle. The chemically driven sliding motion of the thick and thin filaments is responsible for the sarcomere contraction. 21
1.17 The amount of total force developed by the sarcomere is composed by a passive contribution (that exists regardless of activation of the muscle) and an active contractile force, that depends on degree of overlap of the contractile filaments and the activation level of their cross bridges. 22
2.1 Schematic representation of the polymeric chains of an elastomer in their relaxed state and under an external stress. The red dots represent the crosslinks between the chains. 26
2.2 Schematic description of the behavior of an elastic material (left) and of a metal spring (right), according to the Hooke model. When the applied force doubles, also the length of the rubber sample/metal spring doubles. 27
2.3 Schematic representation of the uniaxial extension of a piece of rubber. The sample expands along the direction \(\hat{x}_1\) and contracts equally along \(\hat{x}_2\) and \(\hat{x}_3\). 28
2.4 Experimental setup for the measurement of force/strain curves of materials. 30
2.5 Comparison between the stress-strain curves of three different materials: copper (elastic material), glass (elastic material) and rubber (hyperelastic material). 31
LIST OF FIGURES

2.6 Force/strain relationship of a sample of VHB4910 acrylic elastomer (size: 10mm x 1mm x 10cm). The linear Hooke model is able to represent only the first 10% of strain of the curve. ........................................ 31

2.7 Fit of the Neo-Hookean model to the experimental data obtained for a sample of VHB4910 acrylic elastomer. ................................. 33

2.8 Fit of the Mooney-Rivlin model to the experimental data obtained for a sample of VHB4910 acrylic elastomer. ................................. 35

2.9 Fit of the Ogden model to the experimental data obtained for a sample of VHB4910 acrylic elastomer. The model is able to complete capture the behavior of the elastomer in the whole range of strain (max elongation: 500%). ................................................................. 37

2.10 Comparison between different elastic models. The values of the fitted constants and the elastic modulus of a sample of VHB4910 acrylic elastomer are reported. ........................................ 37

2.11 Comparison between the modeled stress/strain relationships of a sample of VHB4910 acrylic elastomer. ................................. 38

2.12 Comparison between the modeled force/strain relationships of a sample of VHB4910 acrylic elastomer. ................................. 39

2.13 Finite element model of a stripe of elastomeric material. ............... 40

2.14 Deformation of a stripe of VHB 4910 acrylic elastomer under uniaxial traction. ................................................................. 41

2.15 Force/displacement plot for the simulated elastomer stripe. ............ 42

2.16 Width of the stripe of VHB4910 elastomer measured along the x-axis, passing through the center of symmetry of the model. The increase in the applied traction force from 0 to 14N causes a reduction of the cross-sectional area of the elastomer stripe. ................................. 43

2.17 (a) In free atoms, electrons can be found only in discrete energy levels. (b) The proximity of near atoms causes small perturbations in their energy states. (c) In solids, energy levels that cease to exist as discrete states and form energy bands. ........................................ 44
2.18 (a) In conductors, the conduction and the valence band partially overlaps, allowing the electrons to freely move from one band to the other. (b) In insulators the valence band is saturated of electrons that cannot move to the conduction band because the large energy gap.

2.19 (a) In the absence of an external electrical field, dielectric dipoles have random orientation. (b) Dipoles align themselves in the direction of the electric field.

2.20 Schematic representation of a planar capacitor.

2.21 A finite element model of a planar capacitor.

2.22 Results for the finite element simulation of the planar capacitor. The calculated electric field inside the dielectric is $2.0e6$ V/m. The calculated capacitance of the device is $2.82pF$.

2.23 Plot of the calculated electric field along the xy cross sectional plane. The maximum value of the electric field, found inside the dielectric, is $2.0e6$ V/m.

2.24 Plot of the calculated Maxwell surface stress tensor on the two electrodes of the capacitor, for an input voltage of 2000V.

2.25 Force/voltage characteristics for the two electrodes of the capacitor. The resulting maximum force is 90mN for an input voltage of 2000V. The two curves have opposite sign because the forces, acting on the two electrodes along the y-axis, have opposite directions.

2.26 Actuation principle of a planar dielectric elastomer actuator.

2.27 A finite element model of a planar dielectric elastomer actuator.

2.28 Subdomain and boundary settings for the electrostatic module.

2.29 Subdomain and boundary settings for the plane strain (smnp) module.

2.30 Subdomain and boundary settings for the moving mesh (ALE) module.

2.31 Intensity of the electric field.

2.32 Profile of the marked boundary at different applied voltages.

2.33 Y-displacement of the marked point versus applied voltage. The maximum displacement is 41um with an applied voltage of 45kV.

2.34 Thickness strain versus electric field. The maximum electrically induced stain is -18% with an applied electric field of 110V/um.
2.35 X-displacement of the marked point versus applied voltage. The maximum displacement is 0.81mm with an applied voltage of 45kV.  

2.36 Horizontal strain versus electric field. The maximum electrically induced stain is +16% with an applied electric field of 110V/um.  

3.1 Fabrication procedure for a single layer dielectric elastomer actuator. (a) The support frame on which the dielectric film is prestrained. (b) The dielectric elastomer is now fixed on the support frame. (c) The dielectric film is masked and the compliant electrodes are deposed on the two sides of the device. (d) Two copper wires are connected to the lateral inactive areas of the electrode using a conductive glue.  

3.2 Examples of silicone (a) and acrylic (b) actuators fabricated using the described procedure, and clamped to their support frame.  

3.3 Different silicone elastomers.  

3.4 On the left: the liquid silicone rubber is poured on the spinning support. On the right: after the curing process, the obtained silicone film is ready to be used as dielectric layer.  

3.5 A roll of 50um thick acrylic film (VHB 9460).  

3.6 Measurements of the dielectric strength of VHB acrylic dielectrics under different prestrain conditions.  

3.7 Measurements of the dielectric strength of silicone elastomers under different prestrain conditions.  

3.8 Breakdown field of prestrained silicone and acrylic elastomers.  

3.9 Breakdown voltage of prestrained silicone and acrylic elastomers.  

3.10 Dielectric constants of various materials.  

3.11 Dielectric constants of different acrylic and silicone elastomers.  

3.12 Schematic representation of the percolation regime in a composite. (a) Isolated conductive particles in the insulating polymer matrix. (b) At higher volume concentrations, the conductive particles start to form a network of interconnections, even if incomplete (c-d) Above the percolation threshold, a complete conductive path allows the current to flow inside the composite.  

3.13 SEM images of Vulcan XC72-R carbon black at different magnifications.
LIST OF FIGURES

3.14 Microscope image of an homogeneous dust electrode sprayed on the top of a VHB 9460 dielectric film. ............................................ 81
3.15 Microscope image of a grease electrode. ................................. 82
3.16 Samples of electroded elastomer films. On the left, 3M’s 4200 polyurethane was used as polymeric matrix for the fabrication of rubber electrodes (the dielectric layer is VHB 4910). On the right, an Ecoflex 00-30 silicone stripe was electroded using a mixture of carbon black and silicone (CAF4 by Rhodosil). ...................................................... 83
3.17 List of silicone and polyurethane elastomers suitable for the fabrication of compliant electrodes. .............................. 83
3.18 Table solvents ............................................................... 85
3.19 The airbrush setup used to fabricate sprayed rubber electrodes. ... 85
3.20 Measurement of the surface resistivity of a sample. .................... 86
3.21 Surface resistivity of a different electrode types, as a function of the carbon black loading .......................... 88
3.22 Schematic representation of a capacitive pressure sensor. When an external pressure is exerted on the sensor, the dielectric layer between the two electrodes is squeezed, with a resulting increase of the measured capacitance. ...................................................... 89
3.23 Elastomeric electrodes can undergo large deformations without losing their conductivity. ............................................. 89
3.24 Example of pressure sensors based on capacitive technology. (a) a pressure-sensitive artificial skin. (b) a fingertip sensor for a humanoid robot. ... 90
3.25 Experimental setup for strain measurements of single layer DEAs. ........ 90
3.26 Screenshot of the analysis software used to extract the geometric informations of the sample from the acquired digital images. ................. 91
3.27 Plot of the strain of single layer actuators, fabricated with different materials and with different applied prestrains. ......................... 92
3.28 Effects of repeated actuation cycles on the performance of different kinds of single layer actuators. ............................ 93
3.29 Time response of single layer actuators fabricated with different materials. 95
3.30 A schematic representation of the two types of electrical breakdown: a) the
presence of defects can locally lower the dielectric strength of the dielectric
elastomer, causing a puncture breakdown. b) if the electrodes are not properly
insulated, a walkaround breakdown can occur through the air, short-circuiting
the device. ............................................................ [95]

3.31 Sequence of frames showing the dielectric failure of a DEA (VHB4910 dielectric,
carbon grease electrodes). 1) the actuator is in its rest state. 2) the device is
powered and reaches its maximum strain. 3) a dielectric breakdown occurs @
122V/um. 4-6) the spark caused by the breakdown perforates the dielectric
and crack propagates through the dielectric, ripping in two the device. ........ [96]

3.32 To prevent walkaround breakdown, safety unelectroded areas must be consid-
ered on the edges of a single layer dielectric elastomer actuators. ............ [97]

3.33 a-b) acrylic DEA under normal operation. c-d) 3D wrinkling patterns caused
by a pull-in instability. .................................................... [98]

3.34 Model of the uniaxially-prestrained single layer actuator used to determine the
pullin threshold. .......................................................... [99]

3.35 Details of the edges of the simulated actuator during pullin. The two pictures
are obtained varying the initial prestrain of the dielectric layer. .............. [100]

3.36 Calculated pullin voltage for a 50um-thick actuator with the increasing of its
elastic modulus. ......................................................... [101]

3.37 Reliability test of single layer DEAs performed at 90% (top), 80% (middle) and
70% (bottom) of the maximum appliable electric field. ...................... [102]

4.1 The typical internal structure of commercial rolled capacitors. ............... [104]

4.2 A multilayer stack configuration obtained by rolling the electroded dielectric
film on a flat removable core. ......................................... [107]

4.3 On the left: the fabrication process for actuating silicone stripes. On the right,
the silicone stripes removed from the baking support and carefully cut. ...... [108]

4.4 Prototypal rolled DEAs fabricated in different shapes and sizes. ........... [109]

4.5 In this configuration, the electroded lateral areas cause an internal stress, pro-
portional to the number of layers, that opposes to the main actuating motion,
limiting the maximum achievable strain. .................................. [109]
4.6 Finite element model of stacked actuator composed by ten layers. The blue areas represent the active zone of the device, while the red areas are not electroded.

4.7 Boundary conditions for the electrostatic domain of the multilayer stack actuator. The green boundaries are put to ground, while on the red boundaries is applied the electric potential.

4.8 Deformation of a multilayer stack actuator at increasing electric fields.

4.9 Electrically induced displacement along the x-axis vs. applied electric field.

4.10 Electrically induced strain along the x-axis vs. applied electric field.

4.11 Electrically induced displacement along the y-axis vs. applied electric field.

4.12 Electrically induced strain along the y-axis vs. applied electric field.

4.13 Model of a multilayer DEA exploiting length actuation mode.

4.14 Deformed model of a multilayer DEA exploiting length actuation mode.

4.15 Developed length strain vs. applied voltage. The curves are obtained for different applied forces (isotonic condition).

4.16 Relative electro-induced strain for different load conditions. The pull-in instability limits the maximum applicable voltage for high values of the externally applied force.

4.17 Model of a multilayer DEA exploiting thickness actuation mode.

4.18 Deformed model of a multilayer DEA thickness length actuation mode.

4.19 Developed thickness strain vs. applied voltage. The curves are obtained for different applied forces (isotonic condition).

4.20 Relative electro-induced strain for different load conditions.

4.21 Model of the thickness actuation mode of a multilayer actuator at which a compressive force is applied.

4.22 The winding unit.

4.23 Details of the automated spraying unit.

4.24 The semiautomatic machine for the fabrication of multilayer elastomeric actuators, in its fume hood (left) and during the operation (right).

4.25 Schematic representation of different parts that constitute the machine for the fabrication of multilayer DEAs.

4.26 The winding unit.

4.27 Details of the automated spraying unit.

4.28 Schematic representation of the parts constituting the praying unit.
4.29 Schematic representation of the parts constituting the masking unit. .......... [133]
4.30 Schematic connection diagram for the electronic control unit. .................. [134]
4.31 The electronic control unit. ......................................................... [135]
4.32 Screenshot of the graphical interface used to monitor the fabrication process of
the multilayer actuators. ................................................................. [138]
4.33 The algorithm used by the machine for the fabrication of electroded elastomer
roll. .......................................................... ................................. [139]
4.34 Example of multilayer roll fabricated through the described process. In this
example five stacks of eight layers each are fabricated simultaneously. .......... [140]
4.35 If \( \alpha_b \) and \( \alpha_w \) are kept constant for each layer \( l \), the resulting stack’s electrodes
will be impossible to contact. .......................................................... [141]
4.36 The linear length of the electrodes is kept constant for all layers, despite of the
different angular position at which the electrodes are fabricated (\( \alpha_{b1} < \alpha_{b7}, \alpha_{e7}
< \alpha_{e1} \)). .......................................................... ................................. [142]
4.37 On the left: An image of the machine during the removal phase of the protective
liner from a previously electroded roll. This two-step fabrication process allows
the examination of the quality of the fabricated electrodes before the removal
of the protective liner (right). .......................................................... [143]
4.38 (a) Occasional defects in one layer of the dielectric elastomer can cause the
premature failure of the actuator. (b) With two (or more) dielectric layers
in series, the probability of having aligned defects that can cause a dielectric
breakdown is small. .......................................................... ................................. [144]
4.39 Schematic representation of a multilayer roll with no interleaved layers (left),
one interleaved layer (middle), two interleaved layers (right). ..................... [145]
4.40 Interleaved layers decrease the probability of breakdown due to the presence of
defects in the dielectric layer. .......................................................... [146]
4.41 On the left: the electroded roll fabricated by the machine. On the right:
one of the fabricated multilayer stacks is cut from the roll. ..................... [146]
4.42 packaging procedure for multilayer actuators: a) The stack is cut from the roll.
b) The edges of the stack are trimmed in order to expose the electrodes. c) The
electrodes are connected together by manually spraying a layer of conductive
elastomer. .......................................................... ................................. [147]
LIST OF FIGURES

4.43 A packaged multilayer actuator, with the power cables connected on the two sides of the stack. .................................................. 147
4.44 On the left: a section of the lateral area of the stack, in which only the positive electrodes are present. On the right: a section of the central area of the stack, in which both positive and negative electrodes are present. .................... 148
4.45 A section of a multilayer stack (30 layers). The thickness of the dielectric layers (VHB 9460 acrylic elastomer) is 50um. .......................... 149
4.46 Schematic representation of a multilayer actuator as a set of capacitors connected in parallel. In the picture one lateral connection is damaged, causing a reduction of the total capacitance of the device. ......................... 149
4.47 Plot of the expected (teoric) and measured capacitance of multilayer devices (area of each single electrode: 1 cm$^2$). ................................. 150
4.48 Statistics of the number of connected electrodes for multilayer stacks of 20 layers each. .................................................. 151
4.49 Strain measurements for multilayer actuators with 100% prestrain. ........ 152
4.50 Strain measurements for multilayer actuators with 200% prestrain. ........ 153
4.51 Strain measurements for multilayer actuators with 400% prestrain. ........ 153
4.52 Dependence of the resistivity of the electrodes on the applied biaxial prestrain. 155
1

Introduction

1.1 Why compliant actuators for robotic applications?

Despite the diverse range of actuation devices developed in the last twenty years, the lack of a technology able to reach the functional performance of the biological muscle still represents the most significant barrier for the development of machines which can compete with natural systems in terms of motion performance, safety and energy efficiency.

Nature has always served humans as a model for mimicking and inspiration [2]. Its highly effective biological mechanisms were refined thru evolution over million of years and offer incredible inspiring models for human innovation. For example, even if wheeled vehicles can travel fast and carry enormous payloads on paved roads, legged creatures can transverse rough terrains and perform functions whose complexity are far beyond the capabilities of an automobile.

Robots represent one of the greatest results of the human desire of emulating nature. Several motivation led to developing effective robots, including the need to perform complex manipulation or repetitive tasks with degree of accuracy, as well as dangerous operations in hazardous environments (e.g. hazardous waste clean up, sweeping mine fields, search an rescue, and more). However, while in automation applications the robot is required to perform only a set of preprogrammed operations in a well known environment, the complexity and unpredictability of natural environment have made it virtually impossible to explicitly program a non industrial robot for every foreseeable circumstances. This requires making robots that are adaptive and capable of learning
1. INTRODUCTION

from their own experience. Moreover this robots are expect to be safe, in the sense that their interaction with the surrounding environment should not constitute a injury risk to humans that share the same working area. The ultimate goal in this sense is the development of robots able to coexist and cooperate with humans, helping people in the tasks of everyday life.

![Figure 1.1: ICub(1), a humanoid robotic platform designed to study the development of cognitive capabilities in artificial systems and humans.](image)

This requires that robots with similar size and mass as the humans also have comparable power, strength, velocity and interaction compliance. This ambitious goal can, however, not be achieved with the existing robot technology, in which the robots are designed primarily as rigid position or torque sources and most interaction skills are imposed by virtue of control software. For example, locomotion devices with conventional actuators cannot elastically store and release energy, in contrast to human locomotion, thus require large actuators and significantly more energy. Also, conventional robots in which interaction is controlled by software only, could not avoid an impact to damage the robot and possibly the human neighbor, as the controller will react with some delay.

Electroactive polymers (EAPs) are increasingly being recognized as an important enabling actuating technology for making biologically inspired devices [3] [4]. While practical and reliability issues have so far limited the development of commercial appli-
1.2 Survey of actuation technologies

1.2.1 Traditional actuators

Traditionally, a number of key actuation technologies (electric, pneumatic and hydraulic) have been widely and effectively used in a variety of application domains from industrial robots to humanoid platforms and exoskeleton systems.

In recent years, electric motor technology in particular has found enormous applications in all aspects of motion operations. From a structural point of view, this technology is characterized by the predominant use of heavy, stiff position/velocity and torque actuation units, coupled with rigid non back-drivable transmission mechanisms. These stiff actuation groups are usually implemented by combining DC Brushed or Brushless or AC drives with planetary or Harmonic Drive Gears and/or timing belts. The resulting stiffness of these actuation units, on the other hand, has also some benefits from the functional point of view, since a rigid transmission allows to achieve high accuracy in positioning and high operating bandwidth. In this sense, conventional industrial automation promoted the development of robots optimized for precision, speed and high levels of repeatably, taking advantage of the fact they are needed to operate within constrained and well defined environments. However, as the areas for technical exploitation have been increased, new demands are placed on the available actuation systems and it has become increasingly clear that this traditional stiff actuation approach has significant performance limitations related to safety, efficiency and ability to interact with uncertain environment. To address these issues, control techniques based on impedance/admittance and joint torque control actuation have been developed to increase the ability of the robots to interact with the physical environment. The DLR’s lightweight robot, the SARCOS hydraulically actuated humanoid robot and the Big-Dog quadruped robot from Boston Dynamics are just few examples of successful implementation of these techniques, that exploit sensory data and advanced software control to make compliant conventionally stiff actuated systems.
1. INTRODUCTION

Figure 1.2: Examples of state-of-the-art robots with advanced force control. From left to right: the DLR's humanoid 'Justin', the DLR-III lightweight arm, SARCOS and Big-Dog.

In addition to force control algorithms, in recent years a wide range of experimental novel compliant actuation systems have been developed. Considering the electric motor technologies, an early development towards the realization of actuator units with inherent compliance is the Series Elastic Actuator (SEA) which employs a fixed compliance element between a high impedance actuator and the load. However, since the compliance of this system relies on a fixed mechanical spring, it is not possible to regulate the stiffness of the controlled joint. To address this limitation, actuation units with the ability to modulate compliance have also been developed (Fig. 1.3).

Figure 1.3: Examples of compliant electromechanical actuators: the Series-Elastic Actuator, the Variable Stiffness Actuator and the AMACS.

These systems include the selective compliant Actuator, the antagonist Actuator with Mechanically Adjustable Series Compliance (AMASC), the biological inspired joint stiffness actuation, the Variable Stiffness Actuator (VSA), the Mechanically Adjustable Compliance and Controllable Equilibrium Position Actuator
1.2 Survey of actuation technologies

(MACCEPA)\(^{15}\), the Actuator with Non-Linear Elastic System (ANLES)\(^{16}\). These variable compliance actuation systems typically employ two actuator units in combination with passive elastic elements to control, independently, the compliance and the equilibrium position of the activated joint. Additionally to this solutions, the Jack spring actuator \(^{17}\) and the Mechanical Compliance Adjuster \(^{18}\) differs from the above, by regulating the joint stiffness by changing the number of active turns of a spring element and controlling the effective length of a compliant element respectively. Due to the two actuators required in these variable stiffness implementations, the mechanical complexity, weight, cost and integration are still open issues in these realizations. An alternative approach towards safe human-robot interaction is the distributed macro-mini actuation approach (DM\(^2\)) \(^{19}\). This actuation method uses an actuation structure consisting of low- and high-frequency parallel actuators to create a unified high-performance and safe robotic system (Fig. 1.4).

![Figure 1.4: The parallel-distributed approach of the DM\(^2\) actuator. Each robotic joint is controlled by two motors: one, with high torque an low operating frequency is positioned outside the structure of the manipulator; the other, smaller and operating at high frequency is directly coupled to the joint.](image)

A further alternative to electric motor based variable compliance actuation is the use of fluidic actuation. In particular, pneumatic actuation (Fig. 1.5) has been employed
in various actuation forms ranging from pneumatic cylinders \cite{20} to flexible muscles, including the McKibben muscle actuator \cite{21}, its variations \cite{22} \cite{23} and chamber structures \cite{24}.

**Figure 1.5:** On the left, examples of pneumatic actuators based on the McKibben principle. On the right, a typical actuating configuration for two antagonistic pneumatic 'artificial muscles'.

Thanks to its inherent compliance, pneumatic actuation has intrinsically higher safety levels and it has been employed in various setups from antagonist configurations \cite{25} \cite{26} to hybrid actuation schemas \cite{27} \cite{28}. In particular, one example of this hybrid systems is represented by a version of the previously mentioned macro-mini actuation concept, in which braided pneumatic muscles actuators are used as the high force, low bandwidth actuator, while a smaller electric motor provides the high-frequency actuation contribution.

In these configurations, independent control of torque and compliance becomes feasible by the co-activation of the antagonistic actuators. However, low bandwidth, friction, nonlinearities and indeed the lack of stiffness are some of the issues which have limited the widespread use of pneumatic actuation in traditional industrial inspired robotic designs.

Besides the described traditional approaches, novel actuation units, based on new actuation materials, have recently emerged. More in particular, *Electro Active Poly-
1.2 Survey of actuation technologies

mers (EAPs) have recently gained increasing interest for robotics, due to their unique
electrical and mechanical properties. An overview of these polymeric actuators is given
in the following section.

1.2.2 Introduction to polymeric actuators

Research on polymer-based actuators is relative young discipline, that sees it first en-
gineering applications in the 1980s. In the last two decades, a multitude of different
polymeric materials have been investigated and many different actuating principles
have emerged. Because of this, a classification of all these different types of electroac-
tive polymers has become necessary. Bar-Cohen (3)(4) distinguishes between two main
categories of electroactive polymers: the ionic EAPs and electronic EAPs. There also
other categories of polymers, excluded from this classification, that exhibit shape or vol-
ume changes when heated (29)(30) or exposed to magnetic fields(31), infrared light (32)
or other kinds of stimuli. Because these materials are not directly responsive to electri-
cal stimulation, they are more generally named smart materials but not electroactive
polymers. The following sections will give a brief overview of the different operating
principles of ionic and electronic EAPs, highlighting their peculiar characteristics, their
strong points and weaknesses.

1.2.2.1 Ionic EAPs

Polymers containing ions are called ionic polymers. This class of EAPs (also called
"wet" EAPs, because they work in aqueous solution) comprises polyelectrolyte gels,
ionic polymers and conducting polymers.

1.2.2.2 Polyelectrolyte gels

First studies on chemically-driven gel actuators were conducted by Osada in the eighties
and were based on cross-linked polyacrylic acid gels (33). These materials were able
to show a large, reversible, increase of volume when moved from an acid solution to a
base. In particular, it was found that when the pH of the solution increases to a value
greater than the acid dissociation constant of the gel, the chains of the polymer becomes
ionized. The resulting ion-ion repulsions cause a great expansion of the polymer chains,
that uncoil themselves to something similar to straight chains (34)(35), with a resulting
1. INTRODUCTION

increase of the gel’s volume. A few drawbacks of these first experimented gels were their weakness (typical strength few hundreds of kPa), their slow response (typically hours), and the chemical actuation, that is typically undesirable because difficult to control and complex to implement.

During the decade between 1985 and 1995, a number of new gel systems with stronger mechanical responses were investigated, and also electrically activated gels were developed. In particular it was found that the electrolysis of an aqueous solution can be used to actuate a gel, producing a bending displacement that is proportional to increase of the pH of the solution at the negative electrode (Fig. 1.6).

Figure 1.6: Bending of a polyacrylic acid gel caused by the swelling of the material on the anode side (top).

Additionally, efforts were made to develop gel actuators able to run as sealed system in absence of external water. Latest advancements include gels with incorporated electrodes of conducting polypyrrole, gel composites with high induced anisotropic conductivity, multilayer stack configurations and polyvinylalcohol gels swollen with dimethylsulfoxide in place of water.

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1The electrolysis of the aqueous solution causes a release of hydrogen in region near the negative electrode, with a corresponding increase of the concentration of $OH^-$ ions.
1.2 Survey of actuation technologies

Recently, most the attention moved from this kind of technology to other types of more promising wet EAPs, especially IPMCs and conductive polymers. In particular, part of the reason for slow progress in gel actuators is the difficulty of building complex structures in soft materials. Additionally, electroactive gels typically show a slow response and hysteretic behavior, aggravated by a difficult control of their level of activation. Finally, the progressive degradation of their actuating performances over time limits the number of possible real applications for this kind of technology.

1.2.2.3 Conducting polymers

Conjugated polymers (CPs) have been investigated as promising materials for actuation for many years (41)(42)(43)(44). Although the term conjugated polymer comprises a large variety of different materials (for example polypyrrole, polyaniline, polythiophene etc.) a common characteristics of all of these polymers is to exhibit a well-defined chemical structure, the sp2-hybridization, that extends along their backbone. Because of these \(\pi\)-bonds, conjugated polymers can be continuously oxidized and reduced and show electronic conductance in one these two forms (Fig. 1.7).

![Figure 1.7: Examples of different conducting polymers, with in evidence the sp2-conjugation in their chemical structure.](image)

A conductive polymer actuator is basically structured as an electrochemical cell, with two electrodes (one of whose is the the conjugate polymer) immersed in a liquid electrolyte. By applying a difference of potential between the electrodes, the ions present in the electrolyte diffuses into/from the conducting polymer, due to the change
1. INTRODUCTION

The resulting intercalation of doping ions and solvent molecules causes a structural and dimensional change in the material that works as an actuator (45)(46)(47). Conductive polymer actuators have been investigated in many different geometries. The bending beam, constituted by a simple bilayer with one active polymer layer attached to an inactive carrier layer (gold, kapton, mylar etc.) is probably the simplest configuration: due to the volumetric expansion/contraction of the CP electrode, the beam bends in the two directions. However, this actuation mode causes a drastic reduction in the actuation force. Because of this, different linear actuation modes were investigated (48)(49). For example, conductive polymer fibers that behave like contractile muscular fibers were developed (50). EAMEX Corp. developed a configuration constituted by bundles of tubular conductive polymer films grown around a metallic coils (51) (Fig. 1.8). In other different configurations developed by EAMEX, the aperture of a camera hole can be adjusted and the distance of a focus lens can be regulated by conveniently-shaped conductive polymer actuators (52) (Fig. 1.9).

Figure 1.8: Example of three different configurations of conducting polymer actuators produced by EAMEX Corp.
1.2 Survey of actuation technologies

Despite the large number of prototypes and demonstrators developed using conducting polymers, however, severe shortcomings limit the practical use of these actuating materials. The first problem is related to the stability of the conducting polymers. It was observed that, due to collateral effects (like internal reorganizations of the material, delaminations in multilayer structures, evaporation of solvents and others), the actuator stop working after few hundreds of cycles. Moreover, despite the large amount of force that they are able to develop, CP actuators are typically slow and their response is usually one order of magnitude slower that natural muscles.

While these limitations have so far prevented conductive polymer actuators to find a commercial market, research on novel materials continues constantly to improve their performances as demonstrated by the researches conducted by Prof. G. Wallace’s group at IPR{sup 1} on ionic liquid electrolytes as ‘the elixir of life’ of CP actuators{sup 53} and the recent breakthrough (33.5% max strain, 6.7MPa max stress) obtained by Prof. K. Kaneto’s group at KIT{sup 2}.

1.2.2.4 Ionic Polymers Metal Composite

The technology regarding Ionic Polymer Metal Composites (IPMCs) is probably the most popular in the field of polymeric actuators, considering the large amount of papers published on this subject. IPMCs were firstly investigated and developed in the early

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{sup 1}Intelligent Polymer Research Institute, Australia

{sup 2}Kyushu Institute of Technology, Japan
ninetieths by the group of Prof. Mohsen Shahinpoor and their operating principle is described in a set of articles of the same author.

An IPMC actuator typically consists of a perfluorinated ion-exchange membrane on the surface of which is deposited a metallic electrode. More in particular, metal ions (platinum/gold) are dispersed on the polymer surface and subsequently reduced to the metal atoms which form the electrode film.

Two base polymers are typically used to produce an IPMC actuator: *Nafion* (perfluorosulphonate, made by DuPont) or *Flemion* (perfluorocarboxylate, made by Asahi Glass, Japan). These base polymers can be considered as fixed matrix of negative ions, in which positive ions are free to move. When a external voltage is applied to the IPMC, an electrical field rises inside the polymer. This causes a movement of the cations along with hydrated water molecules towards the cathode, resulting in a bending of the EAP towards its anode. Because most of the deflection produced by the actuator is due to the movement of the hydrated ions inside the polymer, it is mandatory for the IPMCs to work in an aqueous environment, in order to maintain constant their moisture content for a long time. Moreover, due to its wetness, voltages greater than 1V involve electrolysis during activation, causing degradation of the polymer, heat and release of gases like hydrogen. Finally, it must be noticed that process of diffusion of the ions inside the polymer is typically slow, and this limits the maximum achievable bandwidth that can be obtained with an IPMC actuator.

Because of these limitations, in addition to a poor controllability of the displacement (due to hysteretic effects and degradation of the performances in time) IPMCs are actively investigated in the research labs in a large variety of configurations like benders, grippers, wipers but presently fail in finding a successful industrial application.

### 1.2.2.5 Electronic polymers

In contrast to ionic polymers, the electronic EAPs (also called 'dry', because they work in standard air environment) are actuated by the external presence of an electric field, in a similar manner to piezoelectric ceramics. Compared to wet EAPs, electronic EAPs have two advantages: their chemical stability respect the surrounding environment and their fast response. However, while ionic polymers operate at low voltages, typically
1.2 Survey of actuation technologies

Figure 1.10: Operating principle of an IPMC actuator. The bending displacement is due to the movement of ions from the anode to the cathode of the device.

Figure 1.11: A demonstrative application of an IPMC gripper in action.
1. INTRODUCTION

few volts, electronic EAPs need high operating voltages (up to the kilovolt range), although at much lower currents. Electronic EAPs comprise electrostrictive polymers and dielectric elastomer actuators.

1.2.2.6 Electrostrictive polymers

Significant high-field electrostriction was firstly observed in certain materials by Scheinbeim at Rutgers (68). In particular, it was found that polyvinylidene fluoride and certain polyurethanes exhibit a large electrostrictive response due to a field-induced re-orientation of their internal structure. Novel materials based on polyvinylidene fluoride were developed by Zhang et al. at Pennsylvania State University (69) (70), demonstrating a specific energy density greater than piezoelectric ceramics, and moderate strains, up to 5%. The high efficiency, the moderate force and displacement and their very fast response speed makes electrostrictive polymers suitable for a large variety of industrial applications, like diaphragm actuators, bending beam actuators, linear actuators, electrostatic speakers and ultrasound transducers.

Presently, research on electrostrictive polymers is carried on by many different groups (Auburn University, NASA-Langey etc) and mainly focuses on material improvements. New polymer formulations are expected to improve manufacturability, increase the developed stress/strain and reduce the high electric field necessary to actuation (71) (72).

1.2.2.7 Dielectric Elastomer Actuators

In Dielectric Elastomers, also known as electrostatically stricted EAPs, the actuating response is generated by the electrostatic interaction between unlike charges present on the two opposing sides of a soft rubbery insulator.

When a electric field is applied across an insulator, opposite charges rise on the electrodes and generate an electrostatic attractive force that squeezes the material in the thickness direction, with a resulting areal expansion that depends on the Poisson’s coefficient of the material. This electrostatic force, called Maxwell stress, is experienced by all dielectrics, but becomes significantly relevant if a soft elastomer (a polymer with low modulus of elasticity) is used. Hence the name of Dielectric Elastomers for this family of electroactive polymers that comprises many different materials, like silicones, acrylic rubbers, soft polyurethanes etc.
1.2 Survey of actuation technologies

Dielectric Elastomer Actuators were firstly developed by Pelrine et al. at SRI International in 1995 (73) (74) (75), reporting large actuating strains (in particular cases more than 100%) and specific energy densities greater than any other field-induced actuator technology. Since then, the dramatic growth of interest on DEAs and the large number of research groups in Europe, Asia and US that started to investigate DEs for actuating purposes has resulted in continuous advancement in this technology, such as new polymer formulations with higher dielectric constants (76), (77), (78) new actuator configurations (79) (80) (81) (82) and predictive mathematical models (83) (84) (85).

Dielectric elastomers offer the promise of a fast acting and efficient technology that is also capable of large motions. Between all EAP technologies, DEAs probably represent the devices that most approach the stroke and the capabilities of biological skeletal muscle, and therefore have been considered for possible ‘artificial muscles application’ in robotics and prosthetics. (86) (87)

The fabrication and the electromechanical characterization of Dielectric Elastomer Actuators are the main objective of this thesis. A detailed description of the operating principle of dielectric elastomers is presented in Section 2, while the actuating performances of single-layer and multilayer DEAs are discussed in sections 3 and 4.

1.2.2.8 Other EAPs

Recent advancements in chemistry and nanotechnology allowed the development of novel actuation concepts and new electroactive materials (88) (89) (90) (91). Liquid crystal elastomers(92), carbon nanotubes actuators (93) (32), electrostrictive papers (94), and molecular actuators (95) are only few of the technologies that are currently investigated by the many EAP research groups in the world. In parallel to these efforts, other EAP-related technologies, such as self repairing electrodes(96) or self-assembled conductive nanocomposites(97), offer the potential to improve EAPs’ performances in a near future.

For further informations on the classification of electroactive polymers and a comprehensive review of these novel actuating concepts, the reader is referred to book of Bar-Cohen Electroactive Polymers Actuators as Artificial Muscles - Reality, Potential and Challenges (3) (4)
1. INTRODUCTION

1.2.2.9 Performances of EAPs and comparison between different actuating technologies

As discussed in the previous sections, the term ‘Electroactive Polymers’ comprise a relative high number of different actuating technologies, each of them presenting peculiar characteristics, different actuating modes and possible limitation in their use. In literature, different attempts have been made to compare the performances between different EAPs. An example of these comparison tables, adapted from [98] is reported in Fig[1.12]. This table compares many traditional and polymeric actuators in terms of speed, energy efficiency, generated stress and strain. Fig[1.13] and Fig[1.14] give additional informations about these technologies, giving a qualitative comparison of their performances through different graphical diagrams.

It must be noticed, however, that the experimental measurements for each technology were typically conducted in different conditions, so an accurate quantitative comparison is not possible. Furthermore, the data represented in these tables are based only on material performances, while practical devices often require additional external structures such as power connections, mechanical supports etc. that can somehow limit the performances of the active material. Finally, maximum peaks in terms of developed strain and pressure can be obtained only in particular conditions and sometimes for a limited number of work cycles. In fact, the performances of many types of polymeric actuators (especially ‘wet’ EAPs) are not constant in time: hysteretic effects and progressive degradation of the active material can cause, in particular cases, a reduction of the developed displacement. Because of this, practical EAPs are still unable, in certain instances, to outperform traditional competing technologies, except for certain niche applications.
### 1.2 Survey of actuation technologies

#### Figure 1.12: Comparison between EAPs, natural muscle and other actuation technologies.

<table>
<thead>
<tr>
<th>Actuator Type (specific example)</th>
<th>Maximum Strain (%)</th>
<th>Maximum Pressure (MPa)</th>
<th>Specific Elastic Energy Density (J/g)</th>
<th>Elastic Energy Density (J/lm³)</th>
<th>Coupling Efficiency (%)</th>
<th>Maximum Efficiency (%)</th>
<th>Specific Density</th>
<th>Relative Speed (full cycle)</th>
</tr>
</thead>
<tbody>
<tr>
<td>Electroactive Polymer Artificial Muscle</td>
<td>215</td>
<td>7.2</td>
<td>3.4</td>
<td>3.4</td>
<td>~60</td>
<td>60–80</td>
<td>1</td>
<td>Medium</td>
</tr>
<tr>
<td>Acrylic Siliconne (CF19-2186)</td>
<td>63</td>
<td>3.0</td>
<td>0.75</td>
<td>0.75</td>
<td>63</td>
<td>80</td>
<td>1</td>
<td>Fast</td>
</tr>
<tr>
<td>Electroactive Polymer (P(VDF-TrFE))</td>
<td>4</td>
<td>15</td>
<td>0.17</td>
<td>0.3</td>
<td>5.5</td>
<td>~60</td>
<td>1</td>
<td>Fast</td>
</tr>
<tr>
<td>Electrostatic Devices (Integrated Force Array)</td>
<td>50</td>
<td>0.03</td>
<td>0.0015</td>
<td>0.0015</td>
<td>~60</td>
<td>&gt; 90</td>
<td>1</td>
<td>Fast</td>
</tr>
<tr>
<td>Electromagnetic (Voice Coil)</td>
<td>50</td>
<td>0.10</td>
<td>0.0003</td>
<td>0.0025</td>
<td>n/e</td>
<td>&gt; 90</td>
<td>8</td>
<td>Fast</td>
</tr>
<tr>
<td>Piezoelectric</td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
</tr>
<tr>
<td>Ceramic (PZT)</td>
<td>0.2</td>
<td>110</td>
<td>0.013</td>
<td>0.10</td>
<td>52</td>
<td>&gt; 90</td>
<td>7.7</td>
<td>Fast</td>
</tr>
<tr>
<td>Single Crystal (PZTNaT)</td>
<td>1.7</td>
<td>131</td>
<td>0.13</td>
<td>1.0</td>
<td>61</td>
<td>&gt; 90</td>
<td>7.7</td>
<td>Fast</td>
</tr>
<tr>
<td>Polymer(P(VDF))</td>
<td>0.1</td>
<td>4.8</td>
<td>0.0015</td>
<td>0.0024</td>
<td>7</td>
<td>n/e</td>
<td>1.8</td>
<td>Fast</td>
</tr>
<tr>
<td>Shape Memory Alloy (TiNi)</td>
<td>&gt; 5</td>
<td>&gt; 200</td>
<td>&gt; 15</td>
<td>&gt; 100</td>
<td>5</td>
<td>&lt; 10</td>
<td>6.5</td>
<td>Slow</td>
</tr>
<tr>
<td>Shape Memory Polymer</td>
<td>100</td>
<td>4</td>
<td>2</td>
<td>2</td>
<td>–</td>
<td>&lt; 10</td>
<td>1</td>
<td>Slow</td>
</tr>
<tr>
<td>Thermal (Ceramic)</td>
<td>1</td>
<td>78</td>
<td>0.15</td>
<td>0.4</td>
<td>–</td>
<td>&lt; 10</td>
<td>2.7</td>
<td>Slow</td>
</tr>
<tr>
<td>Electroactive-thermal-conducting polymer (Polymer)</td>
<td>10</td>
<td>450</td>
<td>23</td>
<td>23</td>
<td>&lt; 1</td>
<td>&lt; 1%</td>
<td>~1</td>
<td>Slow</td>
</tr>
<tr>
<td>Mechano-chemical (polymer-gels)</td>
<td>&gt; 40</td>
<td>0.3</td>
<td>0.06</td>
<td>0.06</td>
<td>–</td>
<td>30</td>
<td>~1</td>
<td>Slow</td>
</tr>
<tr>
<td>Membrane electroactive (Pfendel-O. Electra Products)</td>
<td>0.2</td>
<td>70</td>
<td>0.0027</td>
<td>0.0025</td>
<td>–</td>
<td>60</td>
<td>9</td>
<td>Fast</td>
</tr>
<tr>
<td>Natural Muscle (Human Skeletal)</td>
<td>&gt; 40</td>
<td>0.35</td>
<td>0.07</td>
<td>0.07</td>
<td>n/e</td>
<td>&gt; 35</td>
<td>1</td>
<td>Medium</td>
</tr>
</tbody>
</table>

The table compares various actuation technologies in terms of maximum strain, maximum pressure, specific elastic energy density, elastic energy density, coupling efficiency, maximum efficiency, specific density, and relative speed (full cycle), to provide a comprehensive overview of their performance characteristics.
Figure 1.13: Comparison between different actuator technologies in terms of developed strain and active stress. Performances of natural muscles are given as reference.
1.2 Survey of actuation technologies

Figure 1.14: Comparison between different actuator technologies in terms of specific power and response bandwidth. Performances of natural muscles are given as reference.

Source: DARPA and SRI International
1.3 Biological muscles as a model for compliant actuators

As previously mentioned in the previous sections, standard actuating technology, based on electromagnetic motors and fluidic units, lacks of many features that are commonly present in the skeletal mammalian muscles. To better understand the huge gap that prevents our artificial system to emulate the unique structural and functional properties of biological muscles, this section will give a brief overview about the peculiarities of their physiological structure and contractile motion.

First of all, it’s important to put in evidence that a muscle is not a uniform tissue but it’s the result of the hierarchical organization of smaller units: it is thus from the superimposition of the single properties of these units that the overall behavior the muscle originates.

A skeletal muscle is subdivided into parallel bundles of stringlike multinucleated cells, called muscle fibers (Fig. 1.15), whose diameter is typically in the range of 50-100\,\mu m.

![Hierarchical structure of a mammalian skeletal muscle](image)

**Figure 1.15**: Hierarchical structure of a mammalian skeletal muscles
1.3 Biological muscles as a model for compliant actuators

These fibers, in turn, are composed by smaller units, the myofibrils, that consist of longitudinally repeated contractile units called sarcomeres (Fig. 1.16). The sarcomere is the functional contractile unit of the muscle, and it is structured as an interdigitated matrix of thick and thin proteic filaments. The basis for understanding muscle contraction is the sliding filament model, first proposed in the 1950s by Andrew Huxley (99)(100).

In a contracting muscle, adjacent thick and thin filaments slide past each other, propelled by the cyclic interaction between the myosin heads of the thick filaments that bind to specific sites on the actin of the adjacent thin filaments. The filament sliding is chemically driven by adenosine triphosphate (ATP), a molecule consumed by the myosin heads to reversibly deform and attach to the binding site of the actin filament, forming a cross bridge.

![Image](image-url)

**Figure 1.16:** The sarcomer as the functional unit of the muscle. The chemically driven sliding motion of the thick and thin filaments is responsible for the sarcomer contraction.

The total force output that can be measure at the extremities (tendons) of a muscles derives from the superimposition of a passive and an active contribution. The passive force exists regardless the level of activation of the muscle, starting at the rest length.
1. INTRODUCTION

$L_0$ of the muscle and increasing exponentially as progressive lengthening of the muscle stretches the connectin filaments that tether the thick filaments between the Z disks (Fig. 1.17). The active force, on the contrary, depends by the degree of activation of its muscular fibers (i.e. the force produced by each cross bridge) and the degree of overlap between thick and thin filaments. When the sarcomere is stretched beyond the length at which the thick and thin filaments overlap, the myosin heads are too far from the binding site to form cross bridges and no active force develops (Fig. 1.17a). As the filaments overlap, the number of binding site increases linearly and the developed force increases as well (Fig. 1.17b). Around the muscle’s optimal length, the level of force remains constant because no myosin heads are present in the central portion of the thick filaments (Fig. 1.17c). Further reduction in length cause a progressive overlap of the thin filaments and a consequent reduction of the available binding sites. The resulting active force thus decreases (Fig.1.17d).

![Figure 1.17](image-url)

**Figure 1.17:** The amount of total force developed by the sarcomere is composed by a passive contribution (that exists regardless of activation of the muscle) and an active contractile force, that depends on degree of overlap of the contractile filaments and the activation level of their cross bridges.

Another important consideration on the activation of muscles, is the recruiting strategy. Muscle are composed of different contractile fibers, that can be categorized in
1.3 Biological muscles as a model for compliant actuators

three different types: the slow-twitch fibers (type I), the fast fatigue resistant (type IIA) and the fast fatigable (type IIB). The three types of fibers are characterized by different types of myosin, and are thus able to form cross bridges in different ways. Type I fibers can produce small amounts of tension for long periods and can produce relatively small amounts of force. Type II fibers, on the contrary, produce force more effectively at rapid shortening velocities but are less fatigue resistant. In order to minimize the consumption of energy, muscular fibers are recruited in a fixed order, from weakest to strongest. This means that when only small amounts of force are required, only type I fibers are activated. As more force is required, fast fatigue resistant and then fast fatigable units are activated.

As discussed in this section, muscles are complex hierarchical structures. While their operating principle, based on sliding, chemically driven filaments, is almost impossible to replicate in our artificial system, polymeric actuators are able to emulate other actuating characteristics, like the linear contraction and the intrinsic compliance. The actuation response of dielectric elastomer actuators, in particular, is composed by a non-linear elastic passive behavior (somewhat analogous to the passive relationship between the sarcomere length and its tension) and an electrically-controllable active force. Another similarity between biological muscles and polymeric actuators is the kind of motion that they are able to produce. In fact, while electromagnetic actuators are typically able to generate only rotational torque, a large range of motions, such as bending, linear contraction and areal expansion can be obtained with polymeric devices. Once again, dielectric elastomers are, between all EAP technologies, the most similar to biological muscles in terms of motion type (contraction/expansion), developed stress/strain and response time.

Dielectric elastomers represents a possibility to emulate, from the functional point of view, the behavior of biological muscles. The operating principle of dielectric elastomer actuators and the performances of single and multilayer fabricated with different materials are discussed in the following chapters.
1. INTRODUCTION
Dielectric Elastomer Actuators

The actuation principles of dielectric elastomers are discussed in this chapter, and their electromechanical properties are analyzed as the superimposition of different contributes (i.e. elastic forces and electrostatic interactions).

The chapter is structured as follows: section 2.1 gives a synthetic overview of the theory of elasticity applied to elastomers. Since dielectric elastomer actuators are basically constituted by soft, compliant rubbers, the basic concepts introduced in this section will help to explain the mechanical properties of these elastic materials.

In section 2.2, the dielectric properties of insulating elastomers are presented, and the origin of the Maxwell stress, which is responsible of the actuation of DEAs, is discussed.

Finally the combination of the two effects, the passive restoring force (due to the elastic behavior of the elastomer) and the active electrostatic force (due to the Maxwell stress induced in the dielectric) are combined in section 2.3 to derive the fundamental equation of dielectric elastomer actuators.

2.1 Theory of elasticity applied to elastomers

An elastomer is a polymer characterized, at room temperature and standard pressure, by the property of elasticity (i.e. the ability to deform under an external stress and recover its original shape when the stress is removed).

At the molecular level, the elasticity of a polymer derives from the ability of its polymeric chains to reconfigure themselves to distribute an externally applied stress.
2. DIELECTRIC ELASTOMER ACTUATORS

(Fig. 2.1). The presence of chemical crosslinks (i.e. covalent or ionic bonds between different chains) limits the mobility of the individual chains and allows them to recover their initial configuration when an external stress is removed \((101)(102)\). The chemical structure of the chains and their length is not only responsible of the elasticity of a polymer but also of its viscosity. In particular the interaction (friction) between moving chains causes an energy dissipation whose macroscopic effect results in a viscous behavior of the polymer \((103)(104)(105)\). Hence, the term viscoelasticity is commonly used when referring to elastomers exhibiting viscous behavior (like in the case of VHB acrylic rubbers used to fabricate the dielectric elastomer actuators described in this thesis, see Section 3.2).

![Schematic representation of the polymeric chains of an elastomer in their relaxed state and under an external stress. The red dots represent the crosslinks between the chains.](image)

**Figure 2.1:** Schematic representation of the polymeric chains of an elastomer in their relaxed state and under an external stress. The red dots represent the crosslinks between the chains.

2.1.1 Hooke model

The simplest model describing the elastic properties of an elastomer is the *Hooke model*. To introduce the Hooke model, let’s consider a sample of material, of length \(l_0\) clamped at its extremities and stretched by an external force \(f\) (Fig. 2.2 left).

The Hooke model is linear and states that the amount of elastic force \(f\) generated at the extremities of a sample of a rubber material is proportional to its deformation (change of length \(\Delta l\)). According to the Hooke model, an elastic material thus behaves
2.1 Theory of elasticity applied to elastomers

analogously to a mechanical spring stretched by an external load (Fig. 2.2 right).

\[ f = k\Delta l = k(l - l_0) \]  \hspace{1cm} (2.1)

\[ \sigma \equiv \frac{f}{A} \]  \hspace{1cm} (2.2)

where \( A \) is the cross-sectional area of the sample under analysis. For the strain, we have:

\[ \varepsilon \equiv \alpha - 1 \]  \hspace{1cm} (2.3)

where \( \alpha \) is called stretch ratio and is defined as the ratio between the stretched length of the sample and its original length:

\[ \alpha \equiv \frac{l}{l_0} \]  \hspace{1cm} (2.4)

Let’s now extend the Hooke model in a three dimensional space, by considering the block of rubber material in Fig. 2.3.
2. DIELECTRIC ELASTOMER ACTUATORS

Figure 2.3: Schematic representation of the uniaxial extension of a piece of rubber. The sample expands along the direction $\hat{x}_1$ and contracts equally along $\hat{x}_2$ and $\hat{x}_3$. 
2.1 Theory of elasticity applied to elastomers

Let’s call \(x_1 x_2 x_3\) the three dimensions of the sample, along the three directions \(\hat{x}_1 \hat{x}_2 \hat{x}_3\). If the sample is uniaxially stretched along the direction \(\hat{x}_1\), we will also have an elastic deformation in the other two directions, that can be expressed as:

\[
x'_1 = \alpha_1 \cdot x_1 \\
x'_2 = \alpha_2 \cdot x_2 \\
x'_3 = \alpha_3 \cdot x_3
\]

Because the volume of the rubber is constant during the deformation:

\[
x'_1 \cdot x'_2 \cdot x'_3 = x_1 \cdot x_2 \cdot x_3 = V \\
\alpha_1 \cdot \alpha_2 \cdot \alpha_3 = 1
\]

Additionally, if the rubber deforms with rotational symmetry without shear:

\[
\alpha_2 = \alpha_3 \quad \Rightarrow \quad \alpha_1 = \frac{1}{\alpha_2^2} \quad \Leftrightarrow \quad \alpha_2 = \frac{1}{\sqrt{\alpha_2}}
\]

Let’s now define the Young modulus (also called tensile modulus or modulus of elasticity) as the slope at the very beginning of the stress-strain curve:

\[
\frac{\partial \sigma}{\partial \varepsilon} \rightarrow E \quad \text{for} \quad \varepsilon \rightarrow 0 \quad (2.5)
\]

Using the above definition we have:

\[
\frac{\partial}{\partial \varepsilon} \frac{kl_0}{A_0} (\varepsilon^2 + \varepsilon) = \frac{kl_0}{A_0} (2\varepsilon + 1) \rightarrow \frac{kl_0}{A_0} \quad \text{for} \quad \varepsilon \rightarrow 0 \quad \Rightarrow \quad E = \frac{kl_0}{A_0}
\]

That means that, if the original length, the cross-sectional area and the force constant are known, we can get an estimate of the tensile modulus of \(E=kl_0/A_0\). It must be noticed, however, that the Hooke model can be considered a good approximation of the elastic behavior of the elastomer only for a limited range of strain. In fact, despite to materials like steel, carbon fiber, glass etc, for which the Young’s modulus is essentially constant over a large range of strain \(^1\) elastomers are typically non linear materials (Fig. 2.5) and their stress/strain relationship can be generally described as:

\[
\sigma_{\text{elastic}} = Y(\varepsilon_z)\varepsilon_z \quad (2.6)
\]

\(^1\)Such materials are called linear elastic materials or Hookean materials
2. DIELECTRIC ELASTOMER ACTUATORS

The generic $Y(\varepsilon_z)$ relationship of an elastomeric material can be derived from the corresponding force/strain curve, which can be experimentally measured by stretching a sample of material at a constant rate of motion and measuring the resulting force at the extremities of the sample with a load cell (Fig. 2.4).

Fig. 2.6 represents the typical force/strain curve of an elastomeric material. It can be noticed that the curve is highly non-linear and that after an oblique flex at about 50% of strain, the elastomer exhibits a strain hardening: the Hooke model is thus able to describe well only a very small region at the beginning of the curve, corresponding to a strain of about 10%.

To overcome these limitation, more advanced model needs to be used when dealing with non-linear rubbers. These models, often referred as hyperelastic models, can be classified as:

1. Model based on the phenomenological descriptions of observed behavior: in these models the material behavior is described by means of a strain energy density function, from which the stress-strain relationships are derived.
2.1 Theory of elasticity applied to elastomers

Figure 2.5: Comparison between the stress-strain curves of three different materials: copper (elastic material), glass (elastic material) and rubber (hyperelastic material).

Figure 2.6: Force/strain relationship of a sample of VHB4910 acrylic elastomer (size: 10mm x 1mm x 10cm). The linear Hooke model is able to represent only the first 10% of strain of the curve.
2. DIELECTRIC ELASTOMER ACTUATORS

- Mooney-Rivlin
- Ogden
- Polynomial
- Yeoh

2. Mechanistic models deriving from arguments about underlying structure of the material

- Neo-Hookean
- Arruda-Boyce (8 chain model)

3. Hybrids of phenomenological and mechanistic models

- Gent

While for a exhaustive discussion on this topic the reader is referred to [106], the following sections will give a brief overview of three of these hyperelastic models: the Neo-Hookean model, Mooney-Rivlin model, and the Ogden model.

2.1.2 Neo-Hookean model

The Neo-Hookean model can be considered as an extension from one dimension to three dimensions of the stress response of an elastic body. For a Neo-Hookean solid we have that the total stress can be expressed as:

\[ T = -pI + GB \]

where: \( p \) is the pressure, \( I \) the identity tensor, \( G \) is a constant named shear modulus and \( B \) is the Finger tensor, that in the case of uniaxial extension is diagonal [107] and can be expressed as:

\[ B = \begin{bmatrix} \alpha_1^2 & 0 & 0 \\ 0 & \alpha_1^{-1} & 0 \\ 0 & 0 & \alpha_1^{-1} \end{bmatrix} \]

According to these definitions, the force acting on the extremities of the stretched elastomer sample is:
2.1 Theory of elasticity applied to elastomers

\[ f = A \left[ G(\varepsilon + 1) - \frac{1}{(\varepsilon + 1)^2} \right] \]

The value of the constant \( G \) can be simply obtained by fitting the above equation to the experimental force/strain curve of sample of known cross-sectional area \( A \) (Fig. 2.7).

![Figure 2.7: Fit of the Neo-Hookean model to the experimental data obtained for a sample of VHB4910 acrylic elastomer.](image)

The stress/strain relationship is calculated as:

\[ \sigma = \left[ G(\varepsilon + 1)^2 - \frac{1}{(\varepsilon + 1)} \right] \]

The resulting tensile modulus can be thus calculate as:

\[ E = \frac{\partial}{\partial \varepsilon} \left[ G(\varepsilon - 1) - \frac{1}{(\varepsilon - 1)^2} \right] \rightarrow 3G \quad \text{for} \quad \varepsilon \rightarrow 0 \]
2. DIELECTRIC ELASTOMER ACTUATORS

2.1.3 Mooney-Rivlin model

The Mooney-Rivlin model derives a stress-strain relationship of an elastomer not from a mechanical description of the phenomena but from a strain energy minimization analysis. The strain energy function is defined as:

\[ \Phi = C_1(I_1^B - 3) + C_2(I_2^B - 3) \]

where:

\[ I_1^B = \alpha_1^2 + \alpha_2^2 + \alpha_3^2 \]
\[ I_2^B = \frac{1}{\alpha_1^2} + \frac{1}{\alpha_2^2} + \frac{1}{\alpha_3^2} \]

The stress is then obtained as the derivative of the energy function with respect to the extension ratio:

\[ T_{ii} = \alpha_i \frac{\partial \Phi}{\partial \alpha_i} - p \]

The result can be written in tensor notation using:

\[ \mathbf{T} = -p\mathbf{I} + 2C_1\mathbf{B} + 2C_2\mathbf{B}^{-1} \]

It must be noticed that the Mooney-Rivlin model is a two-parameters generalization of the Neo Hookean model and that this latter can be obtained for particular values of the two constants \((C_2=0, 2C_1=G)\). The force acting on the extremities of the stretched elastomer sample is:

\[ f = A \left( 2C_1 - \frac{2C_2}{(\varepsilon + 1)} \right) \left( (\varepsilon + 1) - \frac{1}{(\varepsilon + 1)^2} \right) \]

The value of the constants \(C_1\) and \(C_2\) can be obtained by fitting the above equation to the experimental force/strain curve of a sample of known geometry (Fig. 2.8).

The calculated resulting stress is:

\[ \sigma = \left( 2C_1 - \frac{2C_2}{(\varepsilon + 1)} \right) \left( (\varepsilon + 1)^2 - \frac{1}{(\varepsilon + 1)} \right) \]

The elastic modulus can be thus calculated as:

\[ E = \frac{\partial}{\partial \varepsilon} \left( 2C_1 - \frac{2C_2}{(\varepsilon - 1)} \right) \left( (\varepsilon - 1) - \frac{1}{(\varepsilon - 1)^2} \right) \to 3(2C_1 - 2C_2) \quad \text{for} \quad \varepsilon \to 0 \]
2.1 Theory of elasticity applied to elastomers

Figure 2.8: Fit of the Mooney-Rivlin model to the experimental data obtained for a sample of VHB4910 acrylic elastomer.
2. DIELECTRIC ELASTOMER ACTUATORS

2.1.4 Ogden model

Like the Mooney-Rivlin model, also the Ogden model is derived from an evaluation of a strain-energy function \[109\]. This time, the strain energy function is defined as:

\[
\Phi = \sum_{i=1}^{n} \mu_i \Phi(k_i)
\]

where:

\[
\Phi(k_i) = \frac{\alpha_1^k + \alpha_2^k + \alpha_3^k + 3}{k}
\]

and \( n \) represents the number of terms used by the model (\( n_{\text{max}} = 3 \)). For a two-terms Ogden model we have that the elastic force can be written as:

\[
f = A \left[ \mu_1 \left( \alpha_1^k - \alpha_1^{1-k_1} \right) + \mu_2 \left( \alpha_1^{k_2} - \alpha_1^{-1/2 \cdot k_2} \right) \right]
\]

The value of the four constants \( \mu_1, \mu_2, k_1 \) and \( k_2 \) can be obtained by fitting the above equation to the experimental force/strain curve of sample of known cross-sectional area \( A \) (Fig. 2.9).

The stress/strain relationship is calculated as:

\[
\sigma = \mu_1 \left( \alpha_1^{k_1-1} - \alpha_1^{-1/2 \cdot k_1} \right) + \mu_2 \left( \alpha_1^{k_2-1} - \alpha_1^{-1/2 \cdot k_2} \right)
\]

and for the elastic modulus:

\[
E = \frac{\partial}{\partial \varepsilon} \left[ \mu_1 \left( \alpha_1^{k_1-1} - \alpha_1^{-1/2 \cdot k_1} \right) + \mu_2 \left( \alpha_1^{k_2-1} - \alpha_1^{-1/2 \cdot k_2} \right) \right] \rightarrow \mu_1 k_1 + \mu_2 k_2 \quad \text{for} \quad \varepsilon \rightarrow 0
\]

Figure 2.11 and Figure 2.12 shows a comparison of the stress/strain and force/strain relationships between all the presented models. It can be noticed that the Ogden model, using four parameters, represents the best description of the experimental data. A comparison between the described models and the fitted values\(^1\) of the used constants are reported in Fig. 2.10.

\(^1\)The reported values refer to a sample of VHB4910 acrylic elastomer (dimensions: (w)10mm x (t)1mm x (l)10cm).
2.1 Theory of elasticity applied to elastomers

Figure 2.9: Fit of the Ogden model to the experimental data obtained for a sample of VHB4910 acrylic elastomer. The model is able to completely capture the behavior of the elastomer in the whole range of strain (max elongation: 500%).

<table>
<thead>
<tr>
<th>Constants</th>
<th>Young Modulus</th>
<th>Stress</th>
<th>Force</th>
</tr>
</thead>
<tbody>
<tr>
<td>Hooke</td>
<td>E 1.17 Mpa</td>
<td>kU/Aα</td>
<td>kU/Aβ (e^α+e^β)</td>
</tr>
<tr>
<td>Neo-Hookean</td>
<td>G 0.41 kPa</td>
<td>3G</td>
<td>G (e^-1/α)</td>
</tr>
<tr>
<td></td>
<td>E 1.22 Mpa</td>
<td></td>
<td>AG (e^-1/α^2)</td>
</tr>
<tr>
<td>Mooney-Rivlin</td>
<td>C1 133 kPa</td>
<td>3(2C1-2C2)</td>
<td>(2C1+2C2)/α (e^-1/α)</td>
</tr>
<tr>
<td></td>
<td>C2 -47.3 kPa</td>
<td></td>
<td>A(2C1+2C2)/α (e^-1/α^2)</td>
</tr>
<tr>
<td></td>
<td>E 1.32 Mpa</td>
<td></td>
<td></td>
</tr>
<tr>
<td>Ogen</td>
<td>u1 745 kPa</td>
<td>u1k1k2+u2</td>
<td>u1(α+1+α-1β)+u2(α-1-α+β)</td>
</tr>
<tr>
<td></td>
<td>u2 650 kPa</td>
<td></td>
<td>A[u1(α+1-α-1β)+u2(α-1-α+β)]</td>
</tr>
<tr>
<td></td>
<td>k1 1.10</td>
<td></td>
<td></td>
</tr>
<tr>
<td></td>
<td>k2 4.42</td>
<td></td>
<td></td>
</tr>
<tr>
<td></td>
<td>E 1.7 Mpa</td>
<td></td>
<td></td>
</tr>
</tbody>
</table>

Figure 2.10: Comparison between different elastic models. The values of the fitted constants and the elastic modulus of a sample of VHB4910 acrylic elastomer are reported.
2. DIELECTRIC ELASTOMER ACTUATORS

Figure 2.11: Comparison between the modeled stress/strain relationships of a sample of VHB4910 acrylic elastomer.
2.1 Theory of elasticity applied to elastomers

Figure 2.12: Comparison between the modeled force/strain relationships of a sample of VHB4910 acrylic elastomer.
2. DIELECTRIC ELASTOMER ACTUATORS

2.1.5 Finite element modeling

The presented models can be used to perform a finite element simulation of a piece of elastomeric rubber. Fig. 2.13 represents a stripe of elastomeric material, modeled using the structural mechanics analysis module (smsld) of Comsol Multiphysics. The model is discretized using a mesh of 10000 tetrahedral elements, for each of whose the numerical solver computes the resulting stress, according to the material properties, the boundary conditions (constrains) and the externally applied loads. The stripe is divided into three sections: the two lateral sections (in blue) are not deformable, and two opposite forces are applied along the z-axis. On the contrary, the middle section (in green) is modeled as a deformable hyperelastic material (VHB 4910), whose parameters are obtained from the table in Fig. 2.10. The overall dimensions of the elastic section are: (w)10mm x (t)1mm x (l)50mm, while the applied force on the two extremities is increased from 0 to 7N. The results of the finite element simulation are presented in Fig. 2.14: the elastomer stripe undergoes a deformation of 71%, under a maximum applied force of 14N. The resulting force/displacement plot in the z-direction is presented in Fig. 2.15 while the width of the stripe (along the x-direction) is shown in Fig. 2.16. The outputs of the simulation match with the experimental results obtained from a sample of VHB acrylic elastomer. Finite element analysis is again performed later on this chapter to model a working dielectric elastomer actuator.

Figure 2.13: Finite element model of a stripe of elastomeric material.
2.1 Theory of elasticity applied to elastomers

Figure 2.14: Deformation of a stripe of VHB 4910 acrylic elastomer under uniaxial traction.
Figure 2.15: Force/displacement plot for the simulated elastomer stripe.
2.1 Theory of elasticity applied to elastomers

Figure 2.16: Width of the stripe of VHB4910 elastomer measured along the x-axis, passing through the center of symmetry of the model. The increase in the applied traction force from 0 to 14N causes a reduction of the cross-sectional area of the elastomer stripe.
2. DIELECTRIC ELASTOMER ACTUATORS

2.2 Dielectric properties of polymers

A dielectric material is an insulator, i.e. a material in which there is no electrical conduction. The band theory of solids gives an explanation about the different electrical behavior of conductors and insulators \(^{110}\). In free atoms, the electrons can be found only in certain discrete states called energy levels (Fig. 2.17a). If several atoms are brought together into a molecule, small perturbations in their energy levels can be observed (Fig. 2.17b). When a large number of atoms are brought together to form a solid, the difference in energy between these perturbed states becomes very small, so the levels may be considered to form continuous energy bands (Fig. 2.17c). However, some intervals of energy contain no energy states, no matter how many atoms are aggregated. These regions are called energy gaps (or forbidden bands) and their width is proportional to the amount of energy required by an electron to jump from the lower energy band to the upper one.

In solids, a large amount of energy bands and energy gaps are typically present, but only the interaction between the uppermost energy band in which electrons are typically present (valence band) and the first empty band at higher energy (conduction band) are responsible to determine their electronic properties. In conductors the valence band partially overlaps with the conduction band, so that the electrons are free to move from one band to the other (Fig. 2.18a). On the contrary, if the energy gap is large, the electrons are not able to move from the valence band to the conduction band and the solid is therefore an insulator (Fig. 2.18b).

Figure 2.17: (a) In free atoms, electrons can be found only in discrete energy levels. (b) The proximity of near atoms causes small perturbations in their energy states. (c) In solids, energy levels that cease to exist as discrete states and form energy bands.
2.2 Dielectric properties of polymers

**Figure 2.18:** (a) In conductors, the conduction and the valence band partially overlaps, allowing the electrons to freely move from one band to the other. (b) In insulators the valence band is saturated of electrons that cannot move to the conduction band because the large energy gap.

Let’s now focus on the behavior of an insulating material when in the presence of an external electric field. A typical model of a dielectric regards the material as composed of small dipoles, which are electrically neutral, but posses internal charge separation \( \text{[III]} \). When no external electric field is present, these molecular dipoles have random orientation. However, in the presence of an electric field, the tendency of the dielectric dipoles is to align themselves in the direction of the electric field, such that their positive end points toward the lower potential (Fig. 2.19).

The physical quantity that describes how an dielectric medium is affected by an electric field is called permittivity and can be represented as:

\[
\epsilon = \epsilon_0 \epsilon_r
\]  

(2.7)

where \( \epsilon_0 = 8.854 \cdot 10^{-12} \text{ F/m} \) is a constant, called permittivity of free space (or vacuum permittivity or electric constant) and \( \epsilon_r \), called relative dielectric constant, represents the ratio between the electric flux density in the dielectric and in vacuum\(^1\).

Let’s now consider the example in Fig. 2.20, representing a planar capacitor constituted by two electrode plates of area \( A \), separated by a dielectric material of thickness \( z \). If a voltage \( V \) is applied to the capacitor, charges appear on the surface of the two electrodes. The amount of charge stored by the capacitor is:

\[
Q = C \cdot V
\]  

(2.8)

\(^1\)The electric flux density (also called electric displacement) represents a measure of the strength of an electric field generated by a free electric charge, and corresponds to the number of electric lines of force passing through a given area. The electric flux density is expressed through the equation \( D = \epsilon E \).
2. DIELECTRIC ELASTOMER ACTUATORS

Figure 2.19: (a) In the absence of an external electrical field, dielectric dipoles have random orientation. (b) Dipoles align themselves in the direction of the electric field.

where $C$ is called capacitance and, in the case of a planar capacitor is:

$$C = \varepsilon_0 \varepsilon_r \frac{A}{z} \quad (2.9)$$

The electrostatic energy stored in the capacitor is:

$$U = \frac{1}{2} E^2 A z \quad (2.10)$$

where $E = \frac{V}{z}$ is the intensity of the electric field inside the planar capacitor.

Figure 2.20: Schematic representation of a planar capacitor.

Let’s now evaluate the Maxwell force generated by the electrostatic attraction between the opposite charges present on the capacitor’s plates. We have:

$$f = -\frac{dU}{dz} \quad (2.11)$$
From the Eq. (2.10) and Eq. (2.11) we obtain:

\[ f = -\frac{1}{2} \epsilon_0 V^2 \frac{d(\frac{A}{z^2})}{dz} \]  \hspace{1cm} (2.12)

If we consider a standard capacitor with rigid electrode plates, A is constant and thus:

\[ \frac{d(\frac{A}{z})}{dz} = A \frac{d(\frac{1}{z})}{dz} = -A \frac{1}{z^2} \]

We can thus derive the expression of electrostatic attraction force:

\[ f = \frac{1}{2} \epsilon_0 \epsilon_r A \frac{V^2}{z^2} = \frac{1}{2} \epsilon_0 \epsilon_r A E^2 \]  \hspace{1cm} (2.13)

The Maxwell force is therefore proportional to the dielectric constant of the material and to square of the applied electric field.

The intensity of the electric field and the corresponding Maxwell force can be computed for a generic geometry using a finite element analysis. A simple example of such a kind of analysis is performed using the electrostatics (emes) module of Comsol Multiphysics. Figure 2.21 represents the model used for the simulation: a solid block (in green), surrounded by air, represents the dielectric layer of a planar capacitor. A constant electric potential is applied between the bottom and the upper face of the capacitor, and the resulting electric field inside the dielectric layer is therefore calculated, together to the Maxwell stress acting on the two electrodes. The following parameters are used for the simulation:

- Size of the dielectric layer: 2mm x 2mm x 50um
- Relative permittivity of the dielectric layer: 3.2
- Relative permittivity of the surrounding air: 1
- Applied electric potential: 100V

The results of the performed simulation are presented in Fig. 2.22. The electric field inside a ‘slice’ of dielectric is analyzed, returning a value of 2.0e6 V/m, the same value that can be obtained by considering the simple formula of \( \frac{V}{z} \), valid for parallel plates capacitors. Additionally, the model returns a calculated capacitance of 2.82pF, the same value obtained from Eq. (2.9), thus confirming the accuracy of the model. Finally, the Maxwell stress acting on the two electrodes of the capacitor is
2. DIELECTRIC ELASTOMER ACTUATORS

Figure 2.21: A finite element model of a planar capacitor.

computed, solving the problem for a parametric input voltage. The plot presented in Fig.2.24 shows the intensity of the Maxwell stress on the two faces of the capacitor, for an input voltage of 2000V. The resulting pressure is 22.6kPa. The Maxwell stress is then integrated across the surface of the two electrodes, and the resulting force/voltage characteristic is calculated for the two electrodes in Fig.2.25. The plot evidences the quadratic dependency of the Maxwell force from the applied voltage, and the obtained numerical results match with the ones predicted by Eq.2.13.

2.3 Operational principle of dielectric elastomer actuators

As previously mentioned, dielectric elastomers are a subclass of electronic EAPs able to produce large deformations when an external electric field is applied. To understand the operating principle of a DEA, let’s consider again the parallel plate capacitor example but, this time, let’s put between the two electrode plates a material that is at the same time a good dielectric and an elastomer. As previously explained, elastomers are incompressible materials; this means that if an external force is applied, they will deform by maintaining constant their volume. This constrain can be expressed mathematically as:

\[ V \equiv A \cdot z = \text{const} \quad \Leftrightarrow \quad \frac{dV}{dz} = 0 \]

Let’s now calculate the derivative \( \frac{d(Az)}{dz} \) using the incompressibility constraint:
2.3 Operational principle of dielectric elastomer actuators

Figure 2.22: Results for the finite element simulation of the planar capacitor. The calculated electric field inside the dielectric is $2.0 \times 10^6$ V/m. The calculated capacitance of the device is $2.82 \text{pF}$.
2. DIELECTRIC ELASTOMER ACTUATORS

Figure 2.23: Plot of the calculated electric field along the xy cross sectional plane. The maximum value of the electric field, found inside the dielectric, is $2.0 \times 10^6$ V/m.

Figure 2.24: Plot of the calculated Maxwell surface stress tensor on the two electrodes of the capacitor, for an input voltage of 2000V.
Figure 2.25: Force/voltage characteristics for the two electrodes of the capacitor. The resulting maximum force is 90mN for an input voltage of 2000V. The two curves have opposite sign because the forces, acting on the two electrodes along the y-axis, have opposite directions.
2. DIELECTRIC ELASTOMER ACTUATORS

\[
\frac{d(Vz^2)}{dz} = \frac{d(Vz^2)}{dz} = -2Vz = -2A z^2
\]

Consequently, the Eq. (2.13) presented in the previous section becomes:

\[
f = \frac{\epsilon_0 \epsilon_r AE^2}{z^2}
\]

Equation 2.14 describes the Maxwell force calculated under the hypothesis of having an incompressible elastomeric dielectric. We can notice that this force is exactly twice than the value calculated previously (Eq. 2.13), this because under the Maxwell force, the dielectric elastomer shrinks along the thickness direction and simultaneously expands the area of its electrodes, as result of its incompressibility.

In order to remove from Eq. 2.14 any reference to the sample geometry, we can calculate the Maxwell stress by using the definition stated in Eq. 2.2:

\[
\sigma = \epsilon_0 \epsilon_r E^2
\]

Figure 2.26 represents a typical dielectric elastomer actuator in its simpler configuration: a single layer of elastomeric material, with the two faces covered by compliant electrodes. When an electric potential is applied to the electrodes, the Maxwell pressure squeezes the dielectric elastomer, causing a simultaneous reduction of its thickness and an increase of its area. It must be noticed that, due to incompressibility of the elastomer, the compliance of the electrodes is a key property to obtain large deformations. In fact, if the electrodes are stiffly bonded to the dielectric, the elastomer will be unable to expand itself in the direction perpendicular to the applied electric field and the thickness strain will result diminished (see Section 3.3).

Figure 2.26: Actuation principle of a planar dielectric elastomer actuator.
2.3 Operational principle of dielectric elastomer actuators

Let’s now consider the combination of the effects of the elasticity of the dielectric material and the Maxwell pressure. As discussed in section 2.1, the elastic force of the elastomer opposes to an external stress. In equilibrium condition, the maxwell force and the elastic force are thus balanced:

\[ F_{\text{maxwell}} = F_{\text{elastic}} \]

or, using the stress notation:

\[ \sigma_{\text{maxwell}} = \sigma_{\text{elastic}} \] (2.16)

By substituting the Eq.2.15 and the general stress/strain relationship (Eq.2.6) in Eq. 2.16 we have:

\[ \epsilon_0 \epsilon_r \frac{V^2}{d^2} = Y(\varepsilon)\varepsilon \] (2.17)

Equation 2.17 describes the equilibrium state of a dielectric elastomer actuator in absence of an external load. The non linearity represented by \( Y(\varepsilon) \), however, makes difficult to calculate the static strain \( \varepsilon \) of the actuator. One possibility is to evaluate \( \varepsilon \) using the models presented in Section 2.1. For example, in the simple case of the Neo-Hookean model we can use:

\[ \sigma_{\text{elastic}} = \left[ G(\varepsilon + 1) - \frac{1}{(\varepsilon + 1)^2} \right] \]

However, in the case of complex geometries, different from the planar configuration presented in this section, only finite element analysis can be used to evaluate the strain of a dielectric elastomer actuator.

The previously reported simulations, the structural mechanics analysis (smdl) and electrostatic analysis (emes), can be in fact combined in Comsol multiphysics, in order to predict the deformation of the dielectric layer according to the computed Maxwell stress.

A finite element model of a planar dielectric elastomer actuator is presented in Fig 2.27. In order to reduce the computational complexity of the analysis, the model is reduced to a two-dimensional problem, taking advantage of the symmetry of the device. Only a slice along the xy plane is thus analyzed, but the results are calculated...
2. DIELECTRIC ELASTOMER ACTUATORS

considered a finite thickness along the z direction. This operation is performed by using the plane strain (smnp) module. The sizes for the modeled actuator are:

- global length of the actuator in the x direction, considering the central electroded area and the lateral edges (which is not electroded): 12mm.
- length of the electroded area = 10mm.
- thickness of the dielectric layer (along the y axis) = 0.5mm.
- width of the actuator (along the z axis) = 10mm.

Since this time the simulation includes two different physics domains, the mechanical analysis and the electrostatic analysis, the model is much more complex and requires the application of boundary conditions for both the domains, in addition to a description of how these two domains interact. This operation is performed in Comsol Multiphysics using the ALE (arbitrary Lagrangian-Eulerian) description, which is a technique to simulate deforming computational domains. By adding the ALE application mode to the model it is possible to introduce a moving mesh frame in which formulate the physics equations. Then, the software automatically transforms the equations and uses them to calculate results both in the reference frame and the moving frame.

Fig 2.28 shows the subdomain and boundary condition applied to the dielectric elastomer model in the electrostatic (emes) domain. The actuator is modeled as dielectric film ($\epsilon_r = 3.2$), surrounded by air ($\epsilon_r = 1$). The following condition are applied to the boundaries of the dielectric:

- boundary 1 (blue): electric potential, $V = V_{in}$
- boundary 2 (green): ground, $V = 0$
- all exterior boundaries (red): zero charge/symmetry, $n \cdot D = 0$
- all interior boundaries (black): continuity, $n \cdot (D_1 - D_2) = 0$

In the structural analysis domain (plane strain module), the actuator is modeled as an hyperelastic material (modulus of elasticity = 1.55Mpa), surrounded by air (Fig 2.29). For the boundaries we have:
2.3 Operational principle of dielectric elastomer actuators

Figure 2.27: A finite element model of a planar dielectric elastomer actuator.

Figure 2.28: Subdomain and boundary settings for the electrostatic module.
2. DIELECTRIC ELASTOMER ACTUATORS

- boundary 1 (blue): no constraints, the applied load corresponds to the calculated Maxwell force.

- boundary 2 (green): no constraints, the applied load corresponds to the calculated Maxwell force.

- interior boundary 3 (pink): the boundary is constrained in the y direction.

- all exterior boundaries (red): the boundaries are fixed in both x and y directions.

- all interior boundaries (black): the boundaries are free to move.

**Figure 2.29:** Subdomain and boundary settings for the plane strain (smnp) module.

In order to connect the structural and the electrostatic domains, a moving mesh description is added to the model (Fig. 2.30). The subdomain are divided in two groups: the free displacement areas (pink) and the physics induced displacement area (white). The following boundary conditions are then applied:

- exterior boundaries (green): no mesh displacement, \( dx=dy=0 \).

- interior boundaries (blue): the mesh displacement is calculated using the results of the structural analysis, \( dx=u, \ dy=v \).

- other boundaries (black): no boundary conditions are applied.

The result of the finite element simulation are presented in Fig. 2.31 et seq. The applied voltage is controlled parametrically, raising from 0 to 45kV, and the resulting electric field and induced displacement is computed along the planar and the thickness directions. The maximum thickness strain is of -18\% (Fig. 2.34), for an induced electric field of 107V/um. The corresponding displacement of the actuator along the
2.3 Operational principle of dielectric elastomer actuators

thickness direction is of 0.09mm (Fig. 2.33). The actuator also expands along the planar direction, as predicted by the Maxwell equation, with a quadratic tendency respect to the applied electric field. The maximum planar strain is +16% (Fig. 2.36), corresponding to an elongation of 1.6mm (Fig. 2.35).

One final consideration about the performances of the simulated device regards the applied voltage. It must be noticed, in fact, that a maximum applied voltage of 45kV is uncomfortably high for a real application, rising issues about the insulation level of the driving electronics and the safety of the fabricated device. However since the Maxwell stress depends on the induced electric field, the two parameters, the applied voltage and the thickness of the dielectric layer, can be tuned in order to obtain the same induced stress at much lower voltages. For example if the thickness of the dielectric layer is reduced from 0.5mm to 0.05mm, the same electrically induced strain can be obtained with an applied voltage of 4.5kV. Obtaining practical dielectric elastomer actuators that work at lower voltages is thus possible, and the minimum available dielectric thickness depends only the chosen fabrication process.\footnote{Spin coating technique have been demonstrated successful in obtaining silicone films of few micrometers of thickness, thus micro DEAs, working in the few hundred volt range, can be practically fabricated.}
2. DIELECTRIC ELASTOMER ACTUATORS

Figure 2.31: Intensity of the electric field.
2.3 Operational principle of dielectric elastomer actuators

Figure 2.32: Profile of the marked boundary at different applied voltages.
Figure 2.33: Y-displacement of the marked point versus applied voltage. The maximum displacement is 41\textmu m with an applied voltage of 45\text{kV}.
Figure 2.34: Thickness strain versus electric field. The maximum electrically induced strain is -18% with an applied electric field of 110V/um.
Figure 2.35: X-displacement of the marked point versus applied voltage. The maximum displacement is 0.81mm with an applied voltage of 45kV.
2.3 Operational principle of dielectric elastomer actuators

Figure 2.36: Horizontal strain versus electric field. The maximum electrically induced stain is +16% with an applied electric field of 110V/um.
3

Single Layer Dielectric Elastomer Actuators

In the previous chapter the basic operating principle of a dielectric elastomer actuator was described.

However, no indications were given about how practical dielectric elastomers can be fabricated and which kind of materials can be used to maximize the actuating performances of the device.

Considering materials, it can be easily guessed that the deformation of dielectric elastomer actuator strongly depends on the choice of the dielectric elastomer but also on the nature the fabricated compliant electrodes. Literature reports a large number of polymers, tested alone or combined in composites, as base materials for the fabrication of DEAs: silicones, polyurethanes, nitrile rubbers, acrylics just to cite a few examples. However, since these materials are often used in different acting configuration, whose geometry also affects the performances of the device, a comparison of their electrical and mechanical properties is difficult. Hence the need to test these materials under identical conditions, using a simplest available actuating configuration: the parallel plate capacitor.

In this chapter the electromechanical characterization of prototypal DEAs, fabricated using different kinds of elastomers and electrode materials, is presented. A single-layer configuration was chosen for the prototypal devices: this choice is justified by the simplicity of this geometry, that reduces the complexity of the fabrication, and allows an easier comparison of the performances of different materials.
3. SINGLE LAYER DIELECTRIC ELASTOMER ACTUATORS

The chapter is thus organized as follows: Section 3.1 describes more in detail the configuration adopted for the fabricated single-layer devices. The following sections, 3.2 and 3.3, focus on material issues, highlighting how the different choices of materials constituting the dielectric layer and the compliant electrodes can effect the behavior of the polymeric actuators. Finally the performances of prototypal single-layer DEAs are presented in Section 3.4, and guidelines for the fabrication of multi-layer actuators are discussed.

3.1 Fabrication procedure for single layer DEAs

The single layer dielectric elastomer actuators described in this thesis have been fabricated using the procedure summarized in the following points:

1. Preparation of the dielectric layer.
2. Prestrain of the dielectric film on a support frame.
3. Deposition of the compliant electrodes on the dielectric layer.
4. Connection of the electrodes to the power supply.

The first fabrication step is the preparation of the dielectric layer. The exact procedure can vary depending by the chosen kind of elastomer but typically consists in mixing a base liquid prepolymer with a catalyst that activates the crosslinking process. The liquid mixture is then poured in a flat mould (for film with thickness >500um) or spinned (to obtain thin films with thickness down to few microns). The polymer is finally cured in an oven and removed from the mould (or spinning support).

The prestrain of the dielectric film has two different functions: the first one is to ensure that the film remains taut on the support frame (Fig.3.1b), avoiding the bulging of the device during the actuation. This is particularly important because if the deformation of the film is not planar, it can affect the measurements of the strain during the experiments. For this purpose, a uniaxial prestrain <20% is generally applied to the dielectric film before gluing it to its support frame (Fig.3.1b). The second function of the prestrain is to reduce the thickness of the dielectric layer and to increase its breakdown strength. In this latter case, large biaxial prestrains are needed, typically greater than 100%. More details on this point are given in section 3.2.3.
3.2 Dielectric material

It must be also noticed that the choice of the material used as dielectric affects the bonding strength of film to the support frame. VHB acrylic elastomer, for example, has excellent bonding properties, and no additional glue needs to be used to stick it on its support frame. On the contrary, additional care is required for silicone devices, since silicone firmly sticks only to silicone. In this case, special glues\(^1\) and conveniently shaped frames have to be used, in order to maintain a firm bonding even in the case of large prestrains.

The third fabrication step is the deposition of the compliant electrodes on the two faces of the dielectric film (Fig.3.1c). A masking tape is typically used to cover the parts of the dielectric film that must remain not conductive after the application of the electrodes. In particular, not conductive safety-areas must be be considered on the sides of the film, in order to avoid the rise of sparks that may connect the two electrodes through the air.

The last fabrication step is the connection of the electrodes to the power supply (Fig.3.1d). This operation is performed by connecting two small-gauge copper wires (or two thin aluminum foils) to the inactive lateral area of the actuator, using an electrically conductive glue\(^2\).

Examples of single layer actuators fabricated accordingly to the described procedure are shown in Fig.3.2. It can be noticed the simplicity of this geometry that allows an easy measurement of the lateral strain of actuator by simple optical inspection (see section 3.4.1).

The following sections will discuss in greater detail the dielectric properties of the acrylic/silicone elastomers and the fabrication procedure for compliant electrodes.

---

\(^1\)An example of suitable glue is SILPOXY by Smooth-on, used to fabricate the silicone actuators described in this thesis.

\(^2\)For silicone actuators, a custom conductive glue was prepared by adding carbon black nano-powder (Vulcan XC-72R by Cabot Corp.) to a standard silicone glue (CAF4 by Rhodosil). Further details are given in Section 3.3.
3. SINGLE LAYER DIELECTRIC ELASTOMER ACTUATORS

Figure 3.1: Fabrication procedure for a single layer dielectric elastomer actuator. (a) The support frame on which the dielectric film is prestrained. (b) The dielectric elastomer is now fixed on the support frame. (c) The dielectric film is masked and the compliant electrodes are deposited on the two sides of the device. (d) Two copper wires are connected to the lateral inactive areas of the electrode using a conductive glue.

Figure 3.2: Examples of silicone (a) and acrylic (b) actuators fabricated using the described procedure, and clamped to their support frame.
3.2 Dielectric material

As previously described (section 2.3), the deformation of the polymeric actuator depends on the balance between an active force, (represented by the Maxwell stress) and a passive restoring force (that is related to the elastic properties of the dielectric material). Both of these two forces are dependent on the intrinsic properties of the dielectric material: the Maxwell stress depends on the permittivity of the dielectric and its breakdown strength (that limits the maximum applicable electric field), while the restoring force depends on the Young Modulus of the material.

It’s thus evident that choice of the dielectric material will be dictated by these parameters: in particular, according to Equations 2.15 and 2.6, the ‘optimal’ dielectric must have:

- High electric permittivity: at a fixed value of the electric field, the Maxwell pressure is directly proportional to the permittivity of the material, therefore at higher values of the dielectric constant correspond higher actuating pressures.

- High dielectric strength: higher electric fields can be applied to materials with higher breakdown strength, resulting in an increase of the maximum Maxwell pressure that can be developed by the actuator.

- Low modulus of elasticity: 'soft' elastomers oppose less mechanical resistance to the Maxwell pressure, resulting in an increase of the maximum displacement of the actuator.

In this thesis two different families of elastomers have been experimented as actuating dielectrics: the **silicones** and the **acrylics**. The following sections will discuss more in the detail the peculiarities of these two classes of elastomers, and will give an insight of their electromechanical properties.

3.2.1 Silicone dielectric

Silicones (also known as **polysiloxanes**), are inorganic polymers based on a silicon-oxygen backbone, with functional organic groups (R) attached to the silicon atoms. The chemical formula of silicones can be thus described as: \([R_2SiO]_n\), where R represents organic groups such as methyl, ethyl, and phenyl. By varying these functional groups and the length of the -Si-O- chain, silicones can be synthesized with a wide variety of properties and compositions, with consistencies that vary from liquid to gel to rubber
3. SINGLE LAYER DIELECTRIC ELASTOMER ACTUATORS

to hard plastic. Between the large variety of useful properties of silicones, we can enumerate:

- Thermal stability: dielectric and mechanical properties silicone are stable over a wide operating range of -100 to +250 °C.
- Elasticity and Flexibility: commercially available silicones can be found with tensile strengths ranging from few hundred of kPa up to several MPa.
- High gas permeability and hydrophobicity.
- Low Toxicity: many silicones are compatible for biomedical applications.
- Excellent resistance to oxygen, ozone, UV radiation a many chemical substances (excluding organic solvents).
- Nonstick property.
- Good electrical insulation and high breakdown strength.

Different kinds of silicones were experimentally evaluated, in order to select the most suitable materials for the fabrication of single layer actuators (Fig. 3.3). Two materials in particular were chosen: Ecoflex 00-30 by Smooth-on and Sylgard 184 by Dow Corning. These silicones are composed by two liquid parts (a base prepolymer and a crosslinker) that must be mixed in a well defined ratio (10:1 for Sylgard 184, 1:1 for Ecoflex 00-30) in order to activate the curing process. The mixture is then vigorously stirred and placed in a vacuum chamber for few minutes, in order to remove the entrapped air bubbles. The compound is finally poured in the mould, or spinned onto a glass surface (1700 rpm, 30s) in order to obtain a thin film of material (Fig. 3.4). The process is completed by placing the mould/spinning support in a oven for 1 hour at 125°C. Measurements of the dielectric strength of the fabricated silicone films are reported in the following sections.

3.2.2 Acrylic dielectric

The first reference to an acrylic elastomer used as a dielectric for actuation purposes comes from the paper of Pelrine (74). In his experiments, Pelrine showed that an acrylic adhesive tape, manufactured by 3M and known with the commercial name of
3.2 Dielectric material

<table>
<thead>
<tr>
<th>Name</th>
<th>Shore A</th>
<th>Viscosity</th>
<th>Tensile strength</th>
<th>Elongation at break</th>
<th>Curing time</th>
</tr>
</thead>
<tbody>
<tr>
<td>Ecotex 09-60</td>
<td>09-60</td>
<td>6000 cps</td>
<td>2.7 MPa</td>
<td>900%</td>
<td>18 min</td>
</tr>
<tr>
<td>Ecotex 09-30</td>
<td>09-30</td>
<td>3000 cps</td>
<td>1.20 MPa</td>
<td>900%</td>
<td>3 hrs</td>
</tr>
<tr>
<td>Ecotex 09-10</td>
<td>09-10</td>
<td>14000 cps</td>
<td>0.80 MPa</td>
<td>800%</td>
<td>30 min</td>
</tr>
<tr>
<td>Dragon Skin</td>
<td>A-10</td>
<td>20000 cps</td>
<td>3.27 MPa</td>
<td>1000%</td>
<td>6 hrs</td>
</tr>
<tr>
<td>Smooth-Sil 920</td>
<td>A-20</td>
<td>20000 cps</td>
<td>3.29 MPa</td>
<td>&gt;700%</td>
<td>2 hrs</td>
</tr>
<tr>
<td>Smooth-Sil 920</td>
<td>A-40</td>
<td>20000 cps</td>
<td>4.13 MPa</td>
<td>&gt;700%</td>
<td>24 hrs</td>
</tr>
<tr>
<td>Silastic 106</td>
<td>A-24</td>
<td>80000 cps</td>
<td>4.29 MPa</td>
<td>110%</td>
<td>24 hrs</td>
</tr>
<tr>
<td>Silastic 106</td>
<td>A-24</td>
<td>60000 cps</td>
<td>7.4 MPa</td>
<td>110%</td>
<td>24 hrs</td>
</tr>
</tbody>
</table>

**Figure 3.3:** Different silicone elastomers.

**Figure 3.4:** On the left: the liquid silicone rubber is poured on the spinning support. On the right: after the curing process, the obtained silicone film is ready to be used as dielectric layer.

VHB 4910, was able to develop high strains and pressures under exceptionally high electric fields. This because the extremely high dielectric strength of that dielectric elastomer. The discovery of the electroactive properties of this acrylic elastomer gave a tremendous boost to the research on dielectric elastomer actuators, and many research groups started to investigate on acrylic-based DEAs just because the relatively simple process needed to convert this commercially available material in a performing polymeric actuator.

In the last ten years, numerous attempts have been made to improve the performances of DEAs by identifying new elastomers [77] or developing novel composites [76][112][113] or new polymer chemistries or formulations [78]. However, results obtained so far have not been significantly better than those reported in earlier literature, and VHB elastomer can be still considered one the most performing available materials for DEAs.

The VHB acrylic film is commercialized by 3M as roll of elastomeric material protected by a plastic layer (Fig.3.5). Three kinds of acrylic elastomers have been experimented in this thesis: VHB4910, VHB4905, VHB9460, with thicknesses of 1mm,
3. SINGLE LAYER DIELECTRIC ELASTOMER ACTUATORS

0.5mm and 500um respectively. This means that at least the first two VHB tapes must be prestrained in order to reduce the thickness of the elastomer and thus decrease the actuation voltage to an acceptable range (<10kV).

At room temperature\(^1\) the VHB is extremely sticky and this make extremely easy to glue the acrylic film on its support frame. Because of this, however, the material must be protected from dust that can accidentally stick on the film before the deposition of the electrodes.

Other two significant properties of VHB acrylic elastomer are its temperature-dependent viscoelastic behavior and its high breakdown strength. While for the first property the reader is referred to (115)(116)(117), the dielectric strength of the VHB elastomer is discussed in detail in the following section and compared to the measurements obtained for silicone-based dielectrics.

![Figure 3.5: A roll of 50um thick acrylic film (VHB 9460).](image)

3.2.3 Dielectric strength

The dielectric strength of an insulating material is the maximum electric field strength that it can withstand intrinsically without breaking down, i.e. without experiencing failure of its insulating properties. The band theory introduced in section 2.2 gives an explanation of the physics behind the dielectric breakdown phenomena. In insulators,

\(^1\)The bonding strength of VHB elastomer increases with time and is also dependent on the applied pressure (114) (VHB tapes are also commonly referred as pressure sensitive tapes). The VHB elastomer completely looses its stickiness at ≈-40°C which is the glass transition temperature for this polymer.
3.2 Dielectric material

electrons that occupy the valence band cannot freely move to conduction band because a large energy gap separates the two bands. But if an electric field of sufficient energy is applied, electrons can gain enough energy to jump to the conduction band, thus making a current able to flow inside the dielectric. The dielectric strength of a material is therefore related to its chemical structure: the larger is the energy gap, the higher is electric field needed to cause the dielectric breakdown of the insulator.

In dielectric elastomer actuators, the dielectric strength of the material plays an important role in determining the final performance of the device: it represents in fact the ultimate electric field that can be applied to the actuator without damaging it. The table in Fig.3.6 and Fig.3.7 shows the measured breakdown values for different kinds of dielectrics. In particular, three acrylic dielectrics (VHB 4910, VHB 4905 and VHB 4960) and two silicone elastomers (Ecoflex 00-30, Sylgard 184) were tested. For each kind of elastomer, 20 samples were tested, by depositing compliant electrodes on the top of them and rising the applied voltage until electrical breakdown occurs. Moreover, each sample was tested at different prestrain levels, in order to investigate the dependency of the dielectric strength from the stretch ratio. The following considerations can be derived from the results presented in the two tables:

• At no prestrain, all the acrylic and silicone elastomers have comparable dielectric strengths ($\approx 30\text{ V/um}$).

• The breakdown strength of the tested dielectric elastomers increases at higher prestrain values.

• Sprayed carbon black electrodes does not damage the insulation strength of the acrylic dielectric. This is confirmed by the experiment #19, where aluminum foils were applied to the dielectric films and used as electrodes. The resulting dielectric strength was comparable to the one measured in the devices fabricated by spraying carbon black electrodes.

• The maximum dielectric strength of the VHB material remains constant even if two acrylic films are stacked one on the top of the other to form a thicker dielectric (experiment #20). In fact, even if occasionally defects of the dielectric lowered the insulation strength of few samples, the maximum measured breakdown strength was $35\text{ V/um}$, approximatively the same value reported for the devices fabricated
using one single dielectric layer. The measured value can be thus considered as the intrinsic breakdown strength of the material and it is not influenced by the occasional presence of defects inside the dielectric.

Literature reports different explanations about the increase of the breakdown strength in prestrained dielectric elastomers (74)(118)(119)(120). In (121), it is argued that the breakdown strength across a polymer chain is higher than along it, due to a higher collision cross section between avalanching electrons and polymer atoms. The pre-strain thus uncurls the elastomer chains and aligns them on a grid perpendicular to the applied electric field, whose higher cross-section impedes the accelerating charges to start a breakdown avalanche. It must noticed, however, that this dependency of the breakdown strength from the prestrain is not a phenomenon restricted to dielectric elastomers only, but it can be found also in hard polymers. For example, the breakdown strength of thin-films of PTFE, mylar and kapton is higher than the one reported for the same materials in bulk form (122).

The breakdown strength of the analyzed dielectric elastomers is presented as a function of the applied prestrain in Fig. 3.8 and Fig. 3.9.

<table>
<thead>
<tr>
<th>MATERIALS</th>
<th>THICKNESS PRESTRAIN</th>
<th>THICKNESS AFTER PRESTRAIN</th>
<th>MAX VOLTAGE</th>
<th>MAX BREAKDOWN (V/m)</th>
</tr>
</thead>
<tbody>
<tr>
<td>VHB 4910 - FURANO ELECTRODES</td>
<td>1000</td>
<td>1000</td>
<td>1000</td>
<td>-</td>
</tr>
<tr>
<td>VHB 4006</td>
<td>1000</td>
<td>1000</td>
<td>500</td>
<td>-</td>
</tr>
<tr>
<td>VHB 4005</td>
<td>1000</td>
<td>1000</td>
<td>250</td>
<td>-</td>
</tr>
<tr>
<td>VHB 4905</td>
<td>1000</td>
<td>1000</td>
<td>125</td>
<td>700</td>
</tr>
<tr>
<td>VHB 4910</td>
<td>1000</td>
<td>1000</td>
<td>62.5</td>
<td>9719</td>
</tr>
<tr>
<td>VHB 4905</td>
<td>1000</td>
<td>1000</td>
<td>50.0</td>
<td>4900</td>
</tr>
<tr>
<td>VHB 4005</td>
<td>1000</td>
<td>1000</td>
<td>25.0</td>
<td>-</td>
</tr>
<tr>
<td>VHB 4005</td>
<td>1000</td>
<td>1000</td>
<td>125.0</td>
<td>6525</td>
</tr>
<tr>
<td>VHB 4905</td>
<td>1000</td>
<td>1000</td>
<td>67.0</td>
<td>4095</td>
</tr>
<tr>
<td>VHB 4910</td>
<td>1000</td>
<td>1000</td>
<td>50.0</td>
<td>3965</td>
</tr>
<tr>
<td>VHB 4005</td>
<td>1000</td>
<td>1000</td>
<td>31.3</td>
<td>3297</td>
</tr>
<tr>
<td>VHB 4905</td>
<td>1000</td>
<td>1000</td>
<td>15.0</td>
<td>1993</td>
</tr>
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<td>1000</td>
<td>1000</td>
<td>31.3</td>
<td>1480</td>
</tr>
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<td>1000</td>
<td>25.0</td>
<td>1400</td>
</tr>
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<td>1000</td>
<td>1000</td>
<td>15.0</td>
<td>1200</td>
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<td>1000</td>
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<td>1095</td>
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<td>7.0</td>
<td>1050</td>
</tr>
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<td>VHB 4905</td>
<td>1000</td>
<td>1000</td>
<td>50.0</td>
<td>1750</td>
</tr>
<tr>
<td>VHB 4905</td>
<td>1000</td>
<td>1000</td>
<td>100.0</td>
<td>2712</td>
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</tbody>
</table>

Figure 3.6: Measurements of the dielectric strength of VHB acrylic dielectrics under different prestrain conditions.

74
3.2 Dielectric material

<table>
<thead>
<tr>
<th>MATERIALS</th>
<th>THICKNESS</th>
<th>PRESTRAIN</th>
<th>THICKNESS AFTER PRESTRAIN</th>
<th>MAX VOLT (KV)</th>
<th>MAX BREAKDOWN (V/mm)</th>
</tr>
</thead>
<tbody>
<tr>
<td>ECOFLEX 0030 - CAF ELECTRODES</td>
<td>500</td>
<td>100%</td>
<td>500</td>
<td>755</td>
<td>49.8</td>
</tr>
<tr>
<td>ECOFLEX 0030</td>
<td>500</td>
<td>200%</td>
<td>250</td>
<td>755</td>
<td>50.6</td>
</tr>
<tr>
<td>SYLVANIC 104</td>
<td>500</td>
<td>400%</td>
<td>125</td>
<td>625</td>
<td>36.6</td>
</tr>
<tr>
<td>SYLVANIC 104</td>
<td>500</td>
<td>600%</td>
<td>62.5</td>
<td>3725</td>
<td>59.6</td>
</tr>
<tr>
<td>SYLVANIC 104</td>
<td>500</td>
<td>800%</td>
<td>62.5</td>
<td>3725</td>
<td>76.6</td>
</tr>
</tbody>
</table>

Figure 3.7: Measurements of the dielectric strength of silicone elastomers under different prestrain conditions.

![Dielectric Strength Measurements](image)

Figure 3.8: Breakdown field of prestrained silicone and acrylic elastomers.

![Breakdown Field](image)

Figure 3.9: Breakdown voltage of prestrained silicone and acrylic elastomers.
3. SINGLE LAYER DIELECTRIC ELASTOMER ACTUATORS

3.2.4 Permittivity

It can be noticed from Eq. 2.15 that the Maxwell stress is proportional to the square of applied electric field through a constant, called permittivity, that is an intrinsic property of the dielectric material.

Typical values for dielectric elastomers are in the range of 2.0-8.0, a value that can be considered low if compared to dielectric constants of certain ceramic powders (Fig. 3.10).

<table>
<thead>
<tr>
<th>Material</th>
<th>Relative Permittivity</th>
<th>Material</th>
<th>Relative Permittivity</th>
</tr>
</thead>
<tbody>
<tr>
<td>Vacuum</td>
<td>1 (by definition)</td>
<td>Diamond</td>
<td>6.5-10</td>
</tr>
<tr>
<td>Air</td>
<td>1.00054</td>
<td>Salt</td>
<td>3.16</td>
</tr>
<tr>
<td>Teflon</td>
<td>2.1</td>
<td>Silicon</td>
<td>11.98</td>
</tr>
<tr>
<td>Polyethylene</td>
<td>2.26</td>
<td>Methanol</td>
<td>30</td>
</tr>
<tr>
<td>Polystyrene</td>
<td>2.4-2.7</td>
<td>Water</td>
<td>80.1</td>
</tr>
<tr>
<td>Carbon disulfide</td>
<td>2.6</td>
<td>Titanium dioxide (TiO₂)</td>
<td>98.173</td>
</tr>
<tr>
<td>Silicone</td>
<td>3.8</td>
<td>Strontium titanate (SrTiO₃)</td>
<td>310</td>
</tr>
<tr>
<td>Paper</td>
<td>3.6</td>
<td>(Li,Na)(Zr,Ti)(PO₄)</td>
<td>600-9000</td>
</tr>
<tr>
<td>Silicate Oxide</td>
<td>3.7</td>
<td>Ca₃(CO₃)₂</td>
<td>9000-10000</td>
</tr>
<tr>
<td>Glass</td>
<td>4.7</td>
<td>Barium titanate (BaTiO₃)</td>
<td>1250-10000</td>
</tr>
<tr>
<td>Acrylate VHB</td>
<td>4.7</td>
<td>Magnesium Niobate (FVH)</td>
<td>26000</td>
</tr>
</tbody>
</table>

Figure 3.10: Dielectric constants of various materials.

Literature reports many different attempts to increase the permittivity of dielectric elastomers using ceramic fillers [76] or blends of conductive polymers [78]. It must be noticed, however, that for such kind of high permittivity elastomers, a corresponding increase of the dielectric losses is typically observed. This is an unwanted effect, because the dielectric losses reduce the capability of the modified-elastomer to tolerate high electric fields, resulting in a decreased dielectric strength [112] [113].

In this thesis, no attempts were made to modify the permittivity of the used commercial dielectric elastomers. The dielectric constants of the acrylic and silicone elastomers employed for the fabrication of single layer DEAs is reported in the following table (Fig. 3.11).

3.3 Compliant electrodes

As mentioned in section 2.3, the ability of a dielectric elastomer actuator to generate large deformations relies on the compliance of its electrodes, that must be able to follow the strain of the dielectric layer without losing their conductivity or constraining its
3.3 Compliant electrodes

<table>
<thead>
<tr>
<th>Material</th>
<th>Relative Permittivity</th>
</tr>
</thead>
<tbody>
<tr>
<td>YHE 4910</td>
<td>4.7</td>
</tr>
<tr>
<td>YHE 4905</td>
<td>4.7</td>
</tr>
<tr>
<td>YHE 9460</td>
<td>4.6</td>
</tr>
<tr>
<td>ECOFLEX 0030</td>
<td>3.2</td>
</tr>
<tr>
<td>SYLGARD 184</td>
<td>2.8</td>
</tr>
<tr>
<td>SYLGARD 186</td>
<td>2.6</td>
</tr>
</tbody>
</table>

Figure 3.11: Dielectric constants of different acrylic and silicone elastomers.

movement. If a stiff electrode is used, in fact, the resulting displacement will be greatly reduced (or completely absent) because its tensile strength will oppose the electrically-induced planar expansion of the dielectric film.

Several techniques, materials and fabrication procedures can be used to fabricate compliant electrodes. While compliant electrodes obtained by using commercial conductive greases [123][119][77] are often referred in literature, advanced fabrication techniques have also been developed in the last few years. Ion implanted electrodes [124][125], wrinkled polymeric electrodes [126] and conveniently-structured gold electrodes fabricated by sputtering [75][127] are just few examples. All of this new techniques, however, are complex and expensive, and have the major drawback of being applicable only for the fabrication of small devices. Their use is thus limited to the field of microactuators/microsensors.

In this thesis, compliant electrodes have been fabricated from a binary mixture of an elastomer and a conductive (nano)filler (e.g. example silicone rubber made conductive by the addition graphite powder) that is subsequently sprayed or brushed on the surface of the dielectric film. This procedure has the double advantage of being inexpensive respect to the previously presented techniques, and, more importantly, of being able to fabricate large electroded areas.

The following sections will discuss more in detail the fabrication procedure of compliant electrodes and their conductive and mechanical properties as well.

---

1An automated airbrushing process is presented in Section 4.3 for the fabrication of compliant electrodes of multilayer DEAs.
3. SINGLE LAYER DIELECTRIC ELASTOMER ACTUATORS

3.3.1 Types of electrodes

One of the probably oldest and best-known methods of making a material electrically conductive is to make a composite, loading it with a conductive filler. Many conductive fillers (e.g. metal (nano)particles, metal-coated-glass and carbon fibers) are commercially available; graphite or carbon powder, in particular, is a typically used in EMI shielding applications because its cost effectiveness.

Numerous factors can affect the properties of a conductive polymer composite, in particular the conductivity of the filler particles, the loading level, and the particle shape.

As might be expected, the resistivity of the composite is closely related to the resistivity of the filler. For example, carbon black, which in pure powder form has a volume resistance of approximately 0.1 Ω-cm, can be used to produce polymer composites with resistivity down to 1 Ω-cm. In comparison, silver powder with has a typical resistance of $1.5 \times 10^{-6}$ Ω-cm and it can be thus used to fabricate polymer composites with volume resistances as low as $10^{-2}$ Ω-cm.\(^{128}\)

The effect of filler loading on the composite resistivity follows a nearly universal pattern, characterized by a sudden drop in resistivity when the filler loading reaches a critical point, called percolation threshold. This phenomenon is explained with the help of sketch presented in Fig. 3.12. At low filler loadings (Fig. 3.12a), the filler particles can be considered as conductive islands in a sea of electrically insulating resin. The resistance of the composite is not affected by the presence of the filler particles, because electrons moving through the composite still encounter the insulating polymer. As the filler volume increases, the conductive particles become more and more crowded, and start to come into contact with each other, forming small, incomplete networks (Fig. 3.12b). Finally, at the percolation threshold, the filler particles form a complete conducting network, and the electron can now pass through the composite without encountering the high-resistance polymer resin (Fig. 3.12c). Additional filler loading beyond the percolation threshold does not greatly reduce the resistance of the composite (Fig. 3.12d).

The shape of the particle plays a critical role in the determination of the percolation threshold. The more structured or elaborated is the surface of the particle, the more likely it is to come in contact with a neighbor particle and form a percolative network.
Fillers constituted by spherical particles, can require as much as 40% loading in order to reach the percolation threshold. Carbon-black particles are more irregularly shaped and have high surface area to volume ratio. These moderately structured fillers can require 5-35% loading to reach the percolation threshold. It must be noticed, moreover, that due to the typical steepness of the percolation curve it can be difficult to precisely control the level of conductivity of the fabricated composite. Finally, highly shaped fillers such as carbon nanotubes may be present in as little as a few percent by volume in order to achieve low resistance.

For the fabrication of dielectric elastomer actuators, different kind of compliant electrodes were studied and experimentally tested. Dust, grease and rubber electrodes, in particular, were experimentally evaluated and have been demonstrated to be effective for actuation purposes, although each of them present peculiar characteristics, as described in the following section.

### 3.3.2 Dust electrodes

This kind of electrodes is probably the simplest to fabricate, because only conductive carbon powder is used, without the additional support of any polymeric matrix. Vulcan XC72-R, a carbon black from Cabot corporation, was chosen as the conductive medium. Its appearance is deep black, and dust-like, with an extremely low density (about
3. SINGLE LAYER DIELECTRIC ELASTOMER ACTUATORS

100g/l). SEM analysis confirms the irregularly-shaped nature of this material, with long structures branching out from the main body of the particle (Fig. 3.13).

![SEM images of Vulcan XC72-R carbon black at different magnifications.](image)

Figure 3.13: SEM images of Vulcan XC72-R carbon black at different magnifications.

In order to apply the conductive material to the dielectric layer, carbon black powder was first of all dissolved in an appropriate solvent (e.g. acetone, ethanol, toluene or heptane). However, if the mixture is left to itself, the carbon powder sinks to the bottom of the vessel, even if shaken or stirred vigorously. In order to appropriately disperse the powder in the solvent, an ultrasound treatment is therefore necessary. About ten minutes of treatment with a 100Watt ultrasonic homogenizer are typically required to force the carbon black in suspension, without any subsequent precipitation.

After the preparation of the conductive solution, the carbon black can be easily transferred to a VHB acrylic film using an airbrush (Fig. 3.14). It must be noticed that in this operation, the amount of the sprayed material must be reduced as much as possible to prevent the solvent to wet the film. In fact, if the solvent evaporates slowly, the VHB film will swell as consequence of the absorption of part of the solvent. The effect is particular evident if acetone, toluene or heptane is used, while it’s less significant using ethanol.

Dust electrodes are extremely compliant: since no polymeric matrix is used to keep bound together the carbon particles (which are only sticked to the dielectric film), no additional tensile stress is added to the electroded elastomer.
3.3 Compliant electrodes

![Microscope image of an homogeneous dust electrode sprayed on the top of a VHB 9460 dielectric film.](image)

**Figure 3.14:** Microscope image of an homogeneous dust electrode sprayed on the top of a VHB 9460 dielectric film.

### 3.3.3 Grease electrodes

Dust electrodes have the drawback of being applicable only to sticky dielectric films, hence this kind of electrodes can be used to fabricate acrylic actuators but not, for example, silicone ones.

A more stable compliant electrode is obtained if carbon black is dispersed in another material that keeps it bonded to the dielectric film. For example, a conductive grease can be obtained by dispersing the carbon powder in a silicone (PDMS) oil. As for the dust electrodes, also in this case we first need to dissolve the carbon black in a solvent, using an ultra sound treatment. The choice of the possible solvents is however limited to only those solvents able to disperse both carbon powder and silicone oil, such as toluene, tetrahydrofuran, heptane, trichloroethylene etc. Acetone or ethanol, on the contrary, can dissolve carbon black powder but not PDMS oil, and thus cannot be employed for the fabrication of grease electrodes. Regarding the choice of the PDMS oil, different silicone prepolymers were tested. Sylgard 184 (by Dow Corning) and Ecoflex (by Smooth-On) are two silicone kits, composed by a prepolymer and a crosslinker that have to be mixed together to obtain a platinum assisted polymerization. Grease electrodes have been fabricated by using only the PDMS oil, without adding the catalyst to the prepolymer. In order to decrease the viscosity of the fluid, the same solvent used to dissolve the carbon black has to be added to the PDMS. The PDMS and CB solution are then mixed together and stirred thoroughly to obtain an homogeneous solution. The
3. SINGLE LAYER DIELECTRIC ELASTOMER ACTUATORS

container in finally placed in a fume hood to allow the solvent to evaporate. Depending by the viscosity of the silicone oil and the evaporation rate of the solvent, this process can take from few hours to one full day.

Finally, after the complete evaporation of the solvent, the conductive grease is manually brushed on the masked dielectric film (Fig. 3.15). Grease electrodes can be successfully employed to fabricate both silicone and acrylic actuators.

Figure 3.15: Microscope image of a grease electrode.

3.3.4 Rubber electrodes

Dust and grease electrodes have the drawback of not being mechanical stable. Carbon powder, in fact, can be removed from the dielectric film by simple scratching, while grease electrodes can be easily spread on unelectroded areas of the device during the handling of the actuator.

For this reasons, rubber electrodes that strongly bonds to the dielectric film were developed (Fig. 3.16). A rubber electrode is composed by a mixture of carbon black and a crosslinked elastomer. Of course the choice of the appropriate electrode elastomer depends on the nature of the dielectric film. For example, a conductive silicone can be used as electrode only if the dielectric film is silicone too. In any other case, (e.g. silicone electrode on acrylic dielectric) the bond between the electrode and the dielectric film will be very weak, and it will be possible to peel away the electrode from the dielectric film.
3.3 Compliant electrodes

Figure 3.16: Samples of electroded elastomer films. On the left, 3M’s 4200 polyurethane was used as polymeric matrix for the fabrication of rubber electrodes (the dielectric layer is VHB 4910). On the right, an Ecoflex 00-30 silicone stripe was electroded using a mixture of carbon black and silicone (CAF4 by Rhodosil).

To fabricate rubber electrodes, carbon black powder, previously dispersed in a proper solvent, is mixed to the elastomer prepolymer, before the curing process. Different commercial elastomers have been experimentally investigated. Fig.3.17 gives a comprehensive overview of the different materials that have been successfully used to fabricate rubber electrodes.

<table>
<thead>
<tr>
<th>Name</th>
<th>Brand</th>
<th>Family</th>
<th>Type</th>
<th>Notes</th>
</tr>
</thead>
<tbody>
<tr>
<td>CAF 4</td>
<td>Rhodosil</td>
<td>Silicone</td>
<td>1-component</td>
<td>1-component, moisture RTV Cure</td>
</tr>
<tr>
<td>SilPoxy</td>
<td>Smooth-on</td>
<td>Silicone</td>
<td>1-component</td>
<td>1-component, moisture RTV Cure</td>
</tr>
<tr>
<td>Ecoflex 30</td>
<td>Smooth-on</td>
<td>Silicone</td>
<td>2-component</td>
<td>1:1 Mix Ratio, Heat Cure</td>
</tr>
<tr>
<td>Silgard 164</td>
<td>Dow Coming</td>
<td>Silicone</td>
<td>2-component</td>
<td>1:10 Mix Ratio, Heat Cure</td>
</tr>
<tr>
<td>VytaFlex 10</td>
<td>Smooth-on</td>
<td>Polyurethane</td>
<td>2-component</td>
<td>1:1 Mix Ratio, RTV Cure</td>
</tr>
<tr>
<td>VytaFlex 30</td>
<td>Smooth-on</td>
<td>Polyurethane</td>
<td>2-component</td>
<td>1:1 Mix Ratio, RTV Cure</td>
</tr>
<tr>
<td>UreBond 2</td>
<td>Smooth-on</td>
<td>Polyurethane</td>
<td>2-component</td>
<td>1:1 Mix Ratio, RTV Cure</td>
</tr>
<tr>
<td>4200 FC</td>
<td>3M</td>
<td>Polyurethane</td>
<td>1-component</td>
<td>1-component, moisture RTV Cure</td>
</tr>
<tr>
<td>5200 FC</td>
<td>3M</td>
<td>Polyurethane</td>
<td>1-component</td>
<td>1-component, moisture RTV Cure</td>
</tr>
<tr>
<td>SilkaFlex</td>
<td>Silka</td>
<td>Polyurethane</td>
<td>1-component</td>
<td>1-component, moisture RTV Cure</td>
</tr>
</tbody>
</table>

Figure 3.17: List of silicone and polyurethane elastomers suitable for the fabrication of compliant electrodes.

The materials listed in this table can be subdivided in two different categories, according to their chemical nature: silicones and polyurethanes. Silicone electrodes can be applied to silicone dielectrics only. Polyurethanes, on the contrary, can be applied to acrylic and polyurethane dielectrics.

Silicones and polyurethanes can be in turn classified as one-component or two-component elastomers. One-component elastomers are typically easier to process, be-
cause they don’t need the addition of a catalyst and polymerize quickly in presence of an external factor like temperature, humidity, UV light etc. On the contrary, two-component elastomers are composed by a pre-polymer and a catalyst that must be mixed in a proper ratio before adding the CB solution. If electrodes are fabricated by spraying, one-component elastomers are generally preferable, because their faster curing time. On the contrary, best results for mould-casted or brushed electrodes are obtained with two-components elastomers.

Depending on the viscosity of the liquid elastomer before the curing process, it may be necessary to thin the compound, using an additional solvent, before the addition of the carbon black dispersion. It’s thus important that:

- The solvent used to thin the elastomer prepolymer doesn’t modify the physical properties of the elastomer, altering the curing process.

- The carbon powder is dispersed in a solvent that is compatible with the elastomer that we are going to use (preferably the same solvent used to thin the elastomer prepolymer).

Fig. 3.18 represents a compatibility chart for different elastomers/solvents combinations. It can be noticed that carbon black Vulcan XC72 can be easily dispersed in most of solvents with the help of a proper ultrasound treatment. Therefore, the choice of the solvent basically depends only on the choice of polymer. In fact, while silicone-based elastomer can be typically dispersed in polar organic solvents only, for polyurethane-based elastomers a wider range of solvents can be used.

Another consideration regards the mechanical properties of rubber electrodes. In general, all the listed elastomers used to fabricate rubber electrodes have higher modulus of elasticity respect to the dielectric elastomer on which they are applied. However, if spraying technique is used, electrodes with thickness ranging from 5 to 50um can be obtained, an thus they do not significantly constrain the dielectric layer. It must be noticed however, that the dielectric layer cannot be stretched more then ultimate elongation of the electrodes, in order to prevent the ripping of the polymeric matrix and the loosing of conductivity of the electrodes. For this reason, single layer actuators were typically fabricated by applying the required prestrain before the deposition of rubber electrodes (Fig. 3.19).
3.3 Compliant electrodes

**Figure 3.18:** Table solvents

**Figure 3.19:** The airbrush setup used to fabricate sprayed rubber electrodes.
3. SINGLE LAYER DIELECTRIC ELASTOMER ACTUATORS

One final consideration regards the possibility of taking advantage of the sticky nature of the not-fully cured rubber elastomer to glue together several dielectric layers in a stack configuration. Polyurethane electrodes, in particularly, were demonstrated to be extremely effective in gluing acrylic VHB elastomers, and were thus employed for the fabrication of multilayer actuators, as discussed in Chapter 4.

3.3.5 Resistance

In order to evaluate the quality of the fabricated electrodes, the surface resistivity of the compliant electrodes was determined.

The surface resistivity is, by convention, the insulation resistance reported to a square measurement area (unit: Ohm/square). This way, the value of resistivity measurement is not influenced by the geometry of the sample and the configuration of the electrodes. The insulation resistance of a conductive material is obtained from the measurement of the resistance which the material opposes to a flow of electrical current. This resistance is the ratio of the potential gradient (volts) parallel to the current direction against the intensity of the current (amperes) flowing in the material between the measurement electrodes placed on its surface (Ohm law). By convention, the insulation resistance reported to a square measurement area is called surface resistivity. The resistance measurement is performed using an electrode system made of two conductive paint lines as described in the international norm IEC 167 (Fig. 3.20).

![Figure 3.20: Measurement of the surface resistivity of a sample.](image)

In order to avoid any influence of the electrode configuration, the measurement of the insulation resistance between the two electrode is reported to a square measurement area and expressed in ohm/square. The following equation is then applied:
3.3 Compliant electrodes

\[ SR = \frac{R \cdot L}{d} \]

Where \( SR \) is the surface resistivity (expressed in Ohm/square), \( R \) is the average of the resistance measurements (Ohms), \( L \) is the length of the conductive paint lines (cm) and \( d \) is the distance between the electrodes (cm).

Fig. 3.21 shows the surface resistivity of different types of electrodes as a function of carbon black loading. In particular, five types of electrodes are evaluated:

- Carbon black with ECOFLEX30 silicone elastomer
- Carbon black with CAF4 silicone elastomer
- Carbon black in SYLGARD 184 silicone oil
- Carbon black in 3M's 4200 polyurethane elastomer
- Carbon black in Vytaflex10 polyurethane elastomer

The plot shows that, even if the analyzed composites present different percolation thresholds, the resistivity values for carbon loadings > 25%wt are all similar. It was also observed that the mechanical properties of the rubber electrodes have no evident degradations for carbon loadings up to 35%wt. For this reason, rubber electrodes with carbon loadings of 30%wt were used for the fabrication of single-layer and multilayer dielectric elastomer actuators.

3.3.6 Further applications of compliant electrodes

The employment of conductive silicones and polyurethane mixtures as compliant electrodes is not limited to only dielectric elastomer actuators applications. One example is represented by pressure sensing devices, based on capacitive technology. Capacitive sensors are able to detect an external pressure in terms of change of the measured capacitance between a fixed and a movable electrode. Figure 3.22 represents a simple capacitive sensor constituted by an array of fixed electrodes, a soft dielectric layer, and a top electrode that acts as ground plane. When an external pressure is exerted on the sensor, the top electrode come closer to the bottom one, squeezing the dielectric layer. In this way, the smaller gap between the two electrodes causes an increase of the capacitance that is measured by an electronic circuit. Since the top electrode undergoes
3. SINGLE LAYER DIELECTRIC ELASTOMER ACTUATORS

Figure 3.21: Surface resistivity of different electrode types, as a function of the carbon black loading.

large deformations when an object is pressed against the sensor (Fig. 3.23), compliant rubber electrodes are highly recommended in this kind of application.

For further details on the employment of compliant electrodes in pressure sensing devices, please refer to (129) (130), where two different applications, a fingertip for a humanoid robot (Fig. 3.24a) and a pressure-sensitive artificial skin (Fig. 3.24b), have been developed using rubber electrodes fabricated with the previously described techniques.

3.4 Experimental results

3.4.1 Experimental Setup

Strain measurements were performed using an optical setup, composed by a high-resolution camera (Hamamatsu C4742-95) and an analysis program written in Labview (Fig. 3.25). The program filters the images taken by the camera and extracts the profile of the actuator, returning a measurement of the distance between the two edges of the actuator or, alternatively, its areal expansion (Fig. 3.26). A 10 Watt switching
Figure 3.22: Schematic representation of a capacitive pressure sensor. When an external pressure is exerted on the sensor, the dielectric layer between the two electrodes is squeezed, with a resulting increase of the measured capacitance.

Figure 3.23: Elastomeric electrodes can undergo large deformations without losing their conductivity.

Figure 3.24: Example of pressure sensors based on capacitive technology. (a) a pressure-sensitive artificial skin. (b) a fingertip sensor for a humanoid robot.
mode amplifier (Spellman MPS10P1024) was used to generate voltages up to 10 Kilovolts (maximum current: 1mA). The high voltage output of the amplifier was linearly controlled through a reference signal (0-10V) generated by the Labview software. In this way it is possible to measure the strain of the actuator, by applying different voltage profiles to the device and synchronizing the acquisition of the images on them.

**Figure 3.25:** Experimental setup for strain measurements of single layer DEAs.

### 3.4.2 Strain Measurement

Fig. 3.27 represents the transverse strain response of different single layer actuators as function of the applied voltage. Both acrylic (VHB 4910) and silicone (Ecoflex 00-30) dielectrics were tested at different prestrain values. VHB 4910 devices were tested with prestrains of 200%, 400%, 800%, corresponding to a reduction of thickness from 1mm to 500um, 250um and 125um respectively. For silicone actuators, the original thickness of the dielectric film was 400um, and the devices were tested with prestrains of 200% and 400%, corresponding to a resulting thickness of 200um and 100um respectively. Rubber electrodes were employed in both acrylic and silicone devices. For acrylic actuators, 3M’s 4200 polyurethanes was used as polymeric matrix for the fabrication of compliant...
3.4 Experimental results

Figure 3.26: Screenshot of the analysis software used to extract the geometric informations of the sample from the acquired digital images.

Two important aspects are put in evidence in the plot of Fig. 3.27. First, the strain has a quadratic dependency from the applied voltage. This characteristic obviously derives from the quadratic dependency of the Maxwell stress from the induced electric field. Second, actuators with higher prestrains exhibit higher actuation strains in comparison to similar actuator with lower prestrain. This because, as described in section 3.2.3, prestraining the film not only decrease the thickness of dielectric, but also increase its dielectric strength, resulting in an actuator able to withstands higher electric fields and thus generating higher Maxwell stress.

Fig. 3.28 shows the behavior of different kinds of single layer DEAs during repeated actuation cycles. In particular, the plot compares the measurement of the strain of the actuator at a defined actuation cycle, respect to the strain obtained during the first measurement. The obtained value will be thus greater than 100% or smaller than 100% depending if the strain capabilities of the actuator increased or reduced during the repeated actuation cycles. From the plot it can be noticed that:

electrodes. On the contrary, electrodes of silicone actuators were fabricated using CAF-4 silicone.
3. SINGLE LAYER DIELECTRIC ELASTOMER ACTUATORS

- The two silicone actuators are the most stable in their performance, since the measured value of strain after 500 actuation cycle is still the same then the one measured at the beginning of the experiment.

- A creep effect dominates the behavior of the three VHB 4910 acrylic actuators with polyurethane electrode. The measured strain increases, in fact, during repeated actuation cycles. The effect is more evident for prestrained actuators.

- One acrylic actuator was fabricated using grease electrodes. For this actuator, an apparently huge increase of strain is reported, due to the migration of the grease material during the actuation. Since the conductive grease has poor mechanical adhesion to the dielectric film, repeated actuation cycles cause its spreading on the surface of the actuator, resulting in an apparent increase of strain.

- One acrylic actuator was fabricated using dust electrodes. For this actuator, it was measured a decrease of strain during repeated actuation. This effect is opposite to the one previously reported: repeated actuation cycle cause a progressive degradation of the dust electrodes, resulting in a decrement of the actuation strain.

From the presented plot it can concluded that rubber electrodes are mandatory to obtain stable values of strain during repeated actuation, and that silicone Ecoflex 00-30 elastomer show smaller creep effect respect to the acrylic VHB 4910.

3.4.3 Response Speed

The response time of a Dielectric Elastomer Actuator is influenced by three main factors:

- The mechanical properties of the actuator (viscous losses)
- The capacitance of actuator
- The response speed of the control electronic

The mechanical properties of the used materials greatly influence the response speed of the actuator. Elastomeric materials are in fact structured as a tangle of curled chains, whose sliding movement is is subject to viscous friction. Different elastomers
3.4 Experimental results

**Figure 3.27:** Plot of the strain of single layer actuators, fabricated with different materials and with different applied prestrains.

**Figure 3.28:** Effects of repeated actuation cycles on the performance of different kinds of single layer actuators.
3. SINGLE LAYER DIELECTRIC ELASTOMER ACTUATORS

are thus characterized by different viscoelastic behaviors, depending on the length of the polymeric chains and their chemical structure. This effect is particularly evident in VHB acrylic elastomers, whose viscous losses are typically one order of magnitude higher than silicone elastomers.

The capacitance of the actuator (i.e. the capacitance between the two electrode plates) can also affect the response type of the device, but for single layer actuators this value is typically smaller than 0.1nf, and the resulting low-pass effect can be neglected.

Finally, it must be noticed that response time of actuator can be also limited by the response speed on its electronic controller. In particular, switching mode amplifiers operate at tens of kilohertz and the pulsing nature of the high voltage output is smoothed by the use of an output capacitor. Because of this, the rise/fall time of the amplifier is practically limited to a value that depends on the capacitance of its output stage. For the MPS10P1024 amplifier used in the strain measurements, the rise/fall time was about 100 ms.

The response time of single actuators fabricated with acrylic and silicone elastomers are presented in Fig. 3.29. A square pulse is applied to the actuator, and the resulting strain is measured for the whole duration of the pulse. The plot is normalized respect to the maximum strains that each kind of actuator was able to develop. In this experiment, silicone actuators demonstrated faster response time (≈ 200ms) respect to acrylic actuators (≈ 600ms).

3.4.4 Actuator failure mechanisms

During its operation, a DEA can be accidentally pushed beyond its physical limits, leading to a temporary inability to produce mechanical work or, in the worst case, to a permanent damage. This section discusses the different mechanisms, both electrical and mechanical in nature, that can cause the failure of dielectric elastomer actuators.

One failure mechanism is the mechanical tearing of the dielectric layer. This typically happen when the elastomer is stretched beyond its maximum elongation or when a crack present at the edges propagates through the whole film, ripping the actuator in two. This type of failure was observed in both Ecoflex 30 silicone actuators and VHB 4910 acrylic actuators. For this kind of actuators the maximum elongation at break was 800% (Tear Strength: 6.65 N/mm) and 700% (Tear Strength: 5.86N/mm) respectively. In order to limit as much as possible the propagation of the cracks, reinforcements can
3.4 Experimental results

Figure 3.29: Time response of single layer actuators fabricated with different materials.

be applied on the edges of the dielectric film, for example gluing an additional stripe of acrilic tape (in the case of VHB actuators). For silicone actuators, instead, reinforced films can be obtained by pouring additional silicone on the edges or applying thin lines of silicone glue (and eventually refining the edges with a clean cut, made with scalpel).

Another typical failure mechanism of a dielectric elastomer actuator is the electrical breakdown. Two types of breakdown can be typically observed: the puncture breakdown and the workaround breakdown (Fig. 3.30).

Figure 3.30: A schematic representation of the two types of electrical breakdown: a) the presence of defects can locally lower the dielectric strength of the dielectric elastomer, causing a puncture breakdown. b) if the electrodes are not properly insulated, a workaround breakdown can occur through the air, short-circuiting the device.
The puncture is the most dangerous type of breakdown for a DEA. As described in section 3.2.3, every insulating material owns a well-defined dielectric strength, and electric fields of greater intensity cause an irreparably failure of its insulating properties. Additionally, the presence of occasional defects inside the elastomer can locally lower its dielectric strength, resulting in areas that are more exposed to potential puncture breakdowns (Fig. 3.30).

When a puncture breakdown occurs, the dielectric material is completely burned by the electrical discharge and air goes to occupy the areas that were previously insulated by the dielectric. Since the dielectric strength of the air is much lower than the one of the dielectric elastomer\textsuperscript{1}, further breakdowns will occur in the same place at much lower voltages, making the actuator unusable. Moreover, if the actuator is subject to high prestrain ratios, the puncture of the dielectric will act as a nucleation point for cracks, whose propagation causes the ultimate tearing of the dielectric elastomer (Fig. 3.31).

The risk of puncture breakdown can be limited by measuring the dielectric strength of the chosen elastomer and keeping the actuation voltage below a safety threshold (e.g. 75% of the measured breakdown strength).

\textbf{Figure 3.31:} sequence of frames showing the dielectric failure of a DEA (VHB4910 dielectric, carbon grease electrodes). 1) the actuator is in its rest state. 2) the device is powered and reaches its maximum strain. 3) a dielectric breakdown occurs @ 122V/\textmu m. 4-6) the spark caused by the breakdown perforates the dielectric and crack propagates through the dielectric, ripping in two the device.

The workaround breakdown, on the contrary, is caused by the insulating failure of the air that separates the electrodes on the sides of the actuator. In fact, if the

\textsuperscript{1}The dielectric strength of dry air is about 3Kv/mm, ten times lower than a typical silicone elastomer and about one hundred of time lower than a prestrained VHB4910 film.
edges of the two electrodes are too close and separated by air only, a spark can rise, connecting the electrodes through an air path (Fig. 3.30b). This kind of breakdown can be prevented by an appropriate design of the geometry of the actuator, taking in account safety unelectroded areas on its sides (Fig. 3.32).

The third failure mode of a DEA is the pull-in instability. This phenomena appears when the equilibrium condition (Eq. 2.16) between the Maxwell pressure and elastic restoring force of the elastomer cannot be reached because, as the thickness of the dielectric layer reduces, the electrically induced Maxwell force increases faster than the opposing elastic force \((131)(132)(115)\). In this case the dielectric film collapses into 3D wrinkling patterns, causing a mechanical tearing of the actuator or a dielectric breakdown (Fig. 3.33).

The pullin phenomena is common in electrostatically actuated MEMS devices (where air gap is typically used as dielectric insulator) and the pull-in voltage is one typical parameter. The pullin voltage for a dielectric elastomer actuator can be determined once again with the help of finite element analysis. For the simulation, the model of Section 2.3 is again considered, and an uniaxial prestrain of 20% is applied along the x-direction (Fig. 3.34). The applied voltage is then raised in small steps until the solution ceases to converge. The found critical voltage represents the pullin threshold and, for the simulated single layer actuators, 0.5mm thick, is \(\approx 60.000\text{V}\) (Fig. 3.35). One inter-

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Figure 3.32: To prevent walkarounds breakdown, safety unelectroded areas must be considered on the edges of a single layer dielectric elastomer actuators.

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3.4 Experimental results
3. SINGLE LAYER DIELECTRIC ELASTOMER ACTUATORS

Figure 3.33: a-b) acrylic DEA under normal operation. c-d) 3D wrinkling patterns caused by a pull-in instability.

Interesting aspect highlighted by the simulations is that the pull-in more easily occurs at the edges of the electrodes. In these areas, in fact, the induced electric field raises from 120V/µm up to 250V/µm, causing a local increase of the Maxwell stress and an abnormal thinning of the dielectric layer. Moreover, it must be noticed the pullin voltage is strictly dependent on the stress-strain curve of the dielectric elastomer, so actuators fabricated with different materials exhibit different pullin voltages. The dependence of the pullin voltage on the elasticity modulus of the dielectric layer, estimated through finite element simulation, is presented in Fig. 3.36.

In the performed experiments, pull-in instability was observed only in acrylic actuators and at large values of prestrain (>600%). In unprestrained actuators, in fact, puncture breakdown typically occurs at lower electric fields respect those that can trigger a pullin instability. Analogously, silicone actuators have lower breakdown strength respect to acrylic ones, and pullin instability was never observed, also in prestrained actuators. In order to avoid a potential electromechanical instability in prestrained acrylic actuators, the maximum applied electric field during the experiments was limited to a
safety value of 100V/um, even if higher electric fields could be applied.

\begin{figure}
\centering
\includegraphics[width=\textwidth]{figure3_34}
\caption{Model of the uniaxially-prestrained single layer actuator used to determine the pullin threshold.}
\end{figure}

\subsection*{3.4.5 Reliability tests}

Reliability tests of single layer DEAs were performed by repeatedly cycling the fabricated devices at different actuation levels. In particular, Ecoflex 30 and VHB 4910 DEAs were tested at different prestrain levels, with an applied voltage equal to the 90\%, 80\% and 70\% of the measured mean breakdown strength (refer also to section 3.2.3). For each actuator type, 20 devices were tested for a maximum number of 1000 actuations (duration of the single actuation cycle: 10 second, 50\% duty cycle). Fig. 3.37 reports the results of these experiments.

As shown by the plot, decreasing the applied voltage increases the life time of the actuators: the maximum reliability was obtained for applied voltages smaller than the 70\% of the measured mean breakdown strength. In these condition, all the tested actuators were able to successfully to survive to 1000 actuation cycles. An heuristic explanation of this behavior considers the migration of impurities inside the polymeric matrix during repeated actuation, whose alignment generates weak points with locally lower breakdown strength.

One final consideration on this topic regards the quadratic dependency of the
Figure 3.35: Details of the edges of the simulated actuator during pullin. The two pictures are obtained varying the initial prestrain of the dielectric layer.
3.4 Experimental results

**Figure 3.36:** Calculated pullin voltage for a 50um-thick actuator with the increasing of its elastic modulus.
3. SINGLE LAYER DIELECTRIC ELASTOMER ACTUATORS

Figure 3.37: Reliability test of single layer DEAs performed at 90% (top), 80% (middle) and 70% (bottom) of the maximum appliable electric field.
3.4 Experimental results

Maxwell stress from the applied electric field. This means that if the applied voltage is halved, the electrically induced deformation it reduced to one-fourth. The reduction of the applied voltage is therefore a great limitation respect to the potential actuating performances, but it is a necessary precaution to safeguard the lifespan of the actuator.
3. SINGLE LAYER DIELECTRIC ELASTOMER ACTUATORS
Multilayer Dielectric Elastomer Actuators

One problem related to the actuation principle of macroscopic dielectric elastomer actuators is the high voltage required, typically in the Kilovolt range, that imposes particular care in the insulation of the whole actuator from the surrounding environment. This high actuation voltage, however, could be drastically reduced if a thin film of dielectric elastomer is used. For example, let’s consider an electric field of 15V/um, a typical actuation value for non prestrained actuators. This electric field will result in applied voltage of 9000V for 0.6 mm thick dielectric layer and 750V for a 50um thick dielectric film. This trivial calculation shows how much is preferable to have an actuator made of thin dielectric layers.

Unfortunately, the fabrication of a macroscopic stack-like actuator, starting from thin-films of dielectric elastomer can present many manufacture difficulties. The first difficulty is the handling and the assembly of the films. On the contrary of many strong polymers that are commonly used as thin films in the electronic and textile industry (like Kevlar, Polyimides etc.), the soft elastomer used for the fabrication of DEAs (i.e. silicones, acrylic rubber etc.) are quite fragile and can be easily broken during handling, especially films with thickness smaller than 100um. This fact makes difficult the development of a fabrication process in which complex mechanical manipulations of the thin film are performed, like in the case of folded actuators. Other techniques, like the controlled deposition and spinning layer by layer of curable prepolymer, can present different difficulties. First of all, the presence of a small fabrication defect in one
4. MULTILAYER DIELECTRIC ELASTOMER ACTUATORS

layer can cause cumulative errors that can compromise the deposition of all following layers, leading to the failure of the whole actuator. Moreover, spinning processes are effective only for a limited number of fabricated layers since, as the stack grows in size, the uniformity of the fabricated layer becomes difficult to control. In particular the fabrication of uniform stacks of several cm of length, starting from deposited layers of few microns can be really challenging and requires extremely accurate machines. Finally, the discontinuous nature of an actuator composed by hundreds of layers of deposited dielectric can make the electrodes complex to contact.

All these considerations are both valid for multilayer stacks used to produce contractile motion (the stacking is used to increase the displacement of the actuator) and expanding motion (the stacking is used to increase the force of the actuator). To try to overcome to these manufacture difficulties, a different configuration was investigated, that allows the semi-automatic fabrication of a multilayer stack actuator.

4.1 Fabrication issues

Due to mechanical fragility of a thin elastomeric film, a procedure that avoids articulated mechanical manipulations and potentially harmful stress is needed. One first, simple idea to produce multilayer actuators comes from the fabrication process of rolled-type capacitors (Fig. 4.1). In fact, if an electroded dielectric film is wound on a flat removable core, a device functionally equivalent to a multilayer stack can be obtained (Fig. 4.2).

The complete fabrication process for such devices is described in Fig. 4.3. The process starts with the fabrication of long stripes of electroded dielectric elastomer. The liquid prepolymer (i.e. silicone) is poured in the mould and cured in an oven (for Ecoflex 0030: 150°C for 10 mins). The obtained silicone film is then masked (using a polyester tape) and airbrushed to fabricate the first electrode layer. An intermediate dielectric layer is subsequently poured, cured in the oven and again masked. This time, however, the mask is laterally shifted, in order to have electrodes not completely aligned, but exposing a part of the lateral conductive areas. In this way, when the multilayer actuator is rolled, it will be possible to contact the alternate electrodes on the two sides of the device. Finally, the actuating stripes are removed from their support, cut along the edges of the electrodes, and rolled in the way shown in Fig. 4.2. The process is
4.1 Fabrication issues

Figure 4.1: The typical internal structure of commercial rolled capacitors.

Figure 4.2: A multilayer stack configuration obtained by rolling the electroded dielectric film on a flat removable core.
4. MULTILAYER DIELECTRIC ELASTOMER ACTUATORS

similar also for acrylic VHB actuators: in this last case, since the elastomer is already available as dielectric tape, it will be only necessary to fabricate the masked electrodes and to glue together the two dielectric layers, taking advantage of the sticky nature of the VHB films.

Example of actuators fabricated in accordance with the described procedure are shown in Fig. 4.4. For these actuators, a maximum lateral strain of $\approx 5\%$ was measured, a value considerably lower respect to strains obtained for single layer devices. These poor performances can be explained by carefully examining the geometry of this actuating configuration. As the size of the roll grows up, the amount of material that does not produce useful mechanical work increases. The electroded sides of the stack, in fact, will generate a compressive force that opposes to the overall contraction motion along the main axis (Fig. 4.5). This configuration sets thus a practical limit on the maximum number of layers that can be present in the rolled actuator. This rolled geometry can be however optimized in order to obtain multilayer stacks that do not suffer of the previously described problem. In particular, if the electrodes are fabricated

\begin{figure}[h]
\centering
\includegraphics[width=\textwidth]{figure4.png}
\caption{On the left: the fabrication process for actuating silicone stripes. On the right, the silicone stripes removed from the baking support and carefully cut.}
\end{figure}
4.1 Fabrication issues

during the winding process, it’s possible to obtain a geometry in which the lateral areas are not electroded and can be mechanically removed after the winding phase. The proposed methodology, implemented with an ad-hoc built winding machine, is described in section 4.3.

Figure 4.4: Prototypal rolled DEAs fabricated in different shapes and sizes.

Figure 4.5: In this configuration, the electroded lateral areas cause an internal stress, proportional to the number of layers, that opposes to the main actuating motion, limiting the maximum achievable strain.
4. MULTILAYER DIELECTRIC ELASTOMER ACTUATORS

4.2 Finite element analysis of multilayer actuators

Finite element analysis can be used to investigate the actuating properties of multilayer dielectric elastomer stacks and to obtain a prediction of their behavior during operation. In the following sections different models will be presented, corresponding to different actuating configurations. For every actuation mode, the electrically induced strain of the actuator is computed parametrically, respect to increasing applied voltages and external loads.

4.2.1 Unconstrained case

The unconstrained case is the simplest case, in which the multilayer device is free to move in any direction without any externally applied loads. A finite element model of ten-layers dielectric elastomer actuator is presented in Fig. 4.6. The model of the multilayer actuator is derived from the one presented in section 2.3: the single layer actuator is replicated ten times along the y-direction, and the edges of layers are bonded together. The boundary conditions for the structural analysis domain and the moving mesh domain are analogous to the ones described in the case of the single layer actuator. Fig.4.7 shows the boundary conditions for the electrostatic domain. The electrodes of the odd layers are put to ground, while the electrodes of the even layers are connected to the applied input voltage through a lateral connection. The model of the multilayer stack is thus electrically equivalent to a set of capacitors connected in parallel.

The solution of the finite element analysis is presented in Fig. 4.8. In this unconstrained case, no loads are applied to the actuator and its displacement in both length and thickness directions is computed for increasing applied voltages. The plots presented in Fig.4.9 and Fig.4.10 show respectively the electrically induced displacement and strain along the x-axis (elongation of the actuator). The maximum computed strain is +9% for an applied electric field of 80V/μm. The corresponding contraction of the actuator along the y-axis is reported in Fig.4.11 and Fig.4.12. In this case, the computed contraction of the multilayer stack is -9%, for an applied electric field of 80V/μm.
4.2 Finite element analysis of multilayer actuators

**Figure 4.6:** Finite element model of stacked actuator composed by ten layers. The blue areas represent the active zone of the device, while the red areas are not electroded.

**Figure 4.7:** Boundary conditions for the electrostatic domain of the multilayer stack actuator. The green boundaries are put to ground, while on the red boundaries is applied the electric potential.
Figure 4.8: Deformation of a multilayer stack actuator at increasing electric fields.
4.2 Finite element analysis of multilayer actuators

Figure 4.9: Electrically induced displacement along the x-axis vs. applied electric field.
Figure 4.10: Electrically induced strain along the x-axis vs. applied electric field.
4.2 Finite element analysis of multilayer actuators

Figure 4.11: Electrically induced displacement along the y-axis vs. applied electric field.
Figure 4.12: Electrically induced strain along the y-axis vs. applied electric field.
4.2 Finite element analysis of multilayer actuators

4.2.2 Constrained case 1: length actuation mode

The model of a multilayer device that actuates along its length direction is presented in Fig. 4.13. The model is derived from the unconstrained case, but, this time, one of the two lateral edges is fixed, and on the other is applied a defined force (isotonic condition). The applied force elongates the actuator along its length direction and generates an uniaxial prestrain. When the voltage is applied, the actuator is squeezed, resulting in a further increase of the length-elongation. The device thus elongates under activation in an opposite way respect to biological muscles which contract when stimulated. The developed model computes resulting displacement for a range of input voltages and applied forces (Fig. 4.14).

![Figure 4.13: Model of a multilayer DEA exploiting length actuation mode.](image)

The computed strain of the actuator under different load conditions and applied voltages is presented in Fig. 4.15. With no applied voltage, the actuator is subject only to the externally applied traction force, and it elongates along the x-axis. The maximum strain is +65% with an applied force of 0.5N for 50um thick-layers (the corresponding stress is 1N/mm$^2$). When the voltage is applied to the actuator, the electrically induced Maxwell pressure squeezes the layers of the actuators, and the strain of the actuator along the x-axis increases from 0% to 12% with no applied load and from 65% to 67.5% with 0.5N of applied force. Fig. 4.16 compares the the relative electrically induced strains respect to the different load conditions. It can be noticed
that the developed Maxwell force increases as the applied external load increases. This an obvious consequence of the fact that the distance between the electrodes of every single layer is reduced by the elongation of the actuator. However, when the thickness of the layers becomes sufficiently small, a pullin instability occurs and the solution cease to converge. This voltage thresholds depends on the applied external load, as shown by the breaks of the isotonic voltage/strain curves in Fig 4.16.

Figure 4.14: Deformed model of a multilayer DEA exploiting length actuation mode.
4.2 Finite element analysis of multilayer actuators

Figure 4.15: Developed length strain vs. applied voltage. The curves are obtained for different applied forces (isotonic condition).
Figure 4.16: Relative electro-induced strain for different load conditions. The pullin instability limits the maximum applicable voltage for high values of the externally applied force.
4.2 Finite element analysis of multilayer actuators

4.2.3 Constrained case 2: thickness actuation mode

In thickness actuation mode, the electrically induced Maxwell stress is directly exploited to produce a contraction motion. The finite element model of a multilayer device that actuates along its thickness direction is presented in Fig. 4.17 and Fig. 4.18. In this model, the top edge of the actuator is fixed, while on the bottom edge is applied a traction force (weight) that deforms the actuator along its thickness direction. When a voltage is applied to the device, the induced Maxwell stress, make the multilayer stack to contract, opposing the external traction force. Because the contraction motion, the resulting behavior of the actuator is much more similar to the behavior of biological muscles in this case respect to the length actuation mode. The developed model computes resulting displacement of the actuator for an externally applied force (isotonic condition), by varying its input voltage.

![Model of a multilayer DEA exploiting thickness actuation mode.](image)

**Figure 4.17:** Model of a multilayer DEA exploiting thickness actuation mode.

The computed strain of the actuator under different load conditions and applied voltages is presented in Fig. 4.19. With no applied voltage, the actuator is stretched along the y-axis by the external load. The maximum strain is +51% with an applied pressure of 2.5N/mm². When the voltage is applied to the actuator, the electrically induced contractile motion opposes the externally applied force and the passive
4. MULTILAYER DIELECTRIC ELASTOMER ACTUATORS

strain reduces from 51% to 48% at maximum load, while with no load the contraction of the multilayer stack is 8.5%.

Fig. 4.20 compares the relative electrically induced strains respect to the different load conditions. It can be noticed that the developed Maxwell force reduces as the applied external load increases. In fact, the distance between the electrodes of every single layer increases as the increasing externally applied force elongates the actuator along the thickness direction. The resulting Maxwell stress is thus reduced.
4.2 Finite element analysis of multilayer actuators

Figure 4.19: Developed thickness strain vs. applied voltage. The curves are obtained for different applied forces (isotonic condition).

Figure 4.20: Relative electro-induced strain for different load conditions.
A special subcase of the thickness actuation mode is represented by multilayer actuators at which are applied compressive loads (Fig. 4.21). The finite element model is identical to the one previously described, except for the direction of the externally applied load, that this time squeezes the actuator along the thickness direction. The externally applied force and the Maxwell stress have thus the same direction, and the resulting strain derives by the sum of these two forces.

The computed strain of the actuator under different compressive loads and applied voltages is presented in Fig. 4.22. With no applied voltage, the actuator is compressed along the y-axis by the external load. The maximum strain is 32% with an applied pressure of 1.5 N/mm². When the voltage is applied to the actuator, the electrically induced contractile motion adds up to the externally applied force and the compressive strain increases from 32% to 35%.

Fig. 4.23 compares the relative electrically induced strains respect to the different load conditions. It can be noticed that the developed Maxwell force increases as the applied external load increases. The reason is similar to the one present in the length actuation mode: the compressive loads squeezes the multilayer stack, decreasing the thickness of the elastomer layers, thus increasing the applied electric field. A pull-in effect can also be noticed for highly-compressed actuators.
4.2 Finite element analysis of multilayer actuators

Figure 4.21: Model of the thickness actuation mode of a multilayer actuator at which a compressive force is applied.

Figure 4.22: .
Figure 4.23:
4.3 Setup for the semiautomatic fabrication of multilayer actuators

In this section, the setup used for the fabrication of multilayer dielectric elastomer actuators is presented (Fig. 4.24). The machine is composed by several functional units (Fig. 4.25), whose functions are described individually in the following sections. In particular, it is possible to distinguish:

- The winding unit
- The spraying unit
- The masking unit
- The electronic control unit

In addition, a control software, developed in C++ and running on a standard PC, is connected through a CAN bus with the electronic control unit and allows the monitoring of the fabrication process.

Figure 4.24: The semiautomatic machine for the fabrication of multilayer elastomeric actuators, in its fume hood (left) and during the operation (right)
Figure 4.25: Schematic representation of different parts that constitute the machine for the fabrication of multilayer DEAs.
4.3 Setup for the semiautomatic fabrication of multilayer actuators

4.3.1 The winding unit

The winding unit is a mechanism composed by several mechanical parts, whose functions are to: unroll the raw dielectric elastomer spool, provide a support during the fabrication of the compliant electrodes and remove the protective liner from the elastomeric tape. With reference to Fig. 4.26, the parts that compose the winding unit are:

- The rotating support (1) that allows the loading of the spool of raw dielectric elastomer (2) into the machine. An adjustable friction clutch (3) is used to keep the tension of tape fixed during the fabrication process.

- Two pulleys (4-5) are used to properly align the elastomeric tape with the masking unit.

- The dielectric elastomer film is wound on the rubber-covered mandrel (6), that is controlled by a DC motor, provided with an incremental encoder\(^1\). During the fabrication phase, the acrylic VHB elastomer sticks to the main mandrel, while the liner, removed from the dielectric film, is pulled by a second mandrel (7).

- The idler pulley (8) is mounted on spring-loaded support that presses the dielectric film on the mandrel (6), in order to improve its adhesion to the mandrel.

- The liner the is peeled from the dielectric tape after its adhesion to the mandrel (6) by a metal blade (9). A metal spring is used to push the metal blade against the mandrel.

- The removed liner is collected by the mandrel (7), that is controlled by another DC motor\(^2\). This motor also controls the advancement of the tape at the beginning of the process, when the dielectric elastomer is still not in contact with the main mandrel (6).

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\(^1\) Faulhaber, series 3242 mininmotor, IE2512 encoder, 246:1 planetary gearhead

\(^2\) Faulhaber, series 2342 mininmotor, IE2512 encoder, 66:1 planetary gearhead
Figure 4.26: The winding unit
4.3 Setup for the semiautomatic fabrication of multilayer actuators

4.3.2 The spraying unit

The spraying unit is composed by an automatic airbrush (model AJUF by Paasche) placed at an adjustable distance from a mask (Fig. 4.27). Unlike the typical manual airbrushes for decorative works (used for the fabrication of single layer actuators in section 3.3), this type of automatic airbrush doesn’t have a trigger, and the opening/closing of the atomizing needle is controlled pneumatically. In this way, when no pressured air is present in the system, the atomizing needle is closed, and no fluid flows out the airbrush. When the pressured air is present, the atomizing needle is opened, and the fluid is sprayed. In this way the airbrush can be simply turned on and off by controlling an electrovalve (model A638, by Camozzi pneumatica) that open and closes the flow of the pressured air, while the amount of sprayed fluid and the atomizing pressure are regulated independently by two knobs on the airbrush.

![Figure 4.27: Details of the automated spraying unit.](image)

Connected to the automatic airbrush, a supply tank contains the fluid that will be sprayed by the airbrush to fabricate the compliant electrodes. The structure of this supply system is represented in Fig. 4.28. An electronic pressure regulator (model ITV2030, by SMC) controls the pressure that acts on the top of the supply tank,
4. MULTILAYER DIELECTRIC ELASTOMER ACTUATORS

pushing down the fluid. If no pressured air is present in the tank, the system acts as a standard gravity feed airbrush, in which the spraying fluid flows down from the reservoir to the atomizing needle. However this is possible only when thin fluid are sprayed. On the contrary, if thick fluids or pastes are used, a pressure has to be applied inside the tank to help the flow on the material inside the airbrush. The right value of the tank’s pressure needed to feed the airbrush is determined experimentally (typically around 1-1.5 bars) and can be adjusted by the control software during the fabrication process.

Figure 4.28: Schematic representation of the parts constituting the praying unit.

4.3.3 The masking unit

The unit is composed by a mask located between the automatic airbrush and the elastomeric tape on which the compliant electrodes will be sprayed. The distance between the airbrush and the mask is typically 10-20cm and is regulated by a screw (Fig 4.29).
The optimal distance depends on the pressure of atomizing air, the viscosity of the electrode material and the amount of sprayed fluid, and is determined experimentally. An indicator of a good spraying distance is the appearance of the sprayed electrode. If the airbrush is too near, too much fluid will be sprayed on the target and the electrode will be wet, resulting in a longer curing time. On the contrary if, the airbrush is too far, only a small amount of material will reach the target and the electrode will result inhomogeneous, with a semi-transparent appearance.

**Figure 4.29**: Schematic representation of the parts constituting the masking unit.

While the distance between the airbrush and the mask is regulated off-line (as previously described) and then fixed during the fabrication process, the distance between the mask and the target is continuously controlled by a DC motor during the fabrication process. In fact, as the radius of the fabricated roll increases, the mask has to be raised in order to maintain a minimal distance from the elastomeric tape (Fig.4.29). This distance (≈ 0.5mm) is regulated to minimize the shadowing effect of the sprayed fluid under the mask.
Finally, it must be noticed that the sprayed material can accumulate on the mask’s surface during the fabrication process. For this reason, the mask is covered by a layer of a nonstick silicone that prevents the adhesion of the fluid and the potential obstruction of the hole in the spraying mask.

4.3.4 The electronic control unit

The electronic control unit (Fig. 4.31) is composed by two control boards (MC4 board by Microsystem srl) and a power supply board (MCP board by Microsystem srl) (fig. 4.31). The two control boards are based on the Motorola 56807 DSP and each of them is able to control four DC brushed motors through four amplified PWM channels. The connection diagram of the control unit is shown in Fig. 4.30.

![Schematic connection diagram for the electronic control unit.](image)

According to the schematic, the control board 1 controls the two winding motors
and the motor which control the height of the mask on the dielectric film, while the control board 2 controls the activation of the airbrush’s electro valve and regulates the pressure inside of the fluid tank. A PID control algorithm is implemented on the DSP firmware in order to accurately control the three DC motors, whose angular positions are detected through magnetic encoders directly integrated in the motors.

Both the two MC4 boards are connected to the same power MCP board, that regulates the supply voltage. A large capacitor is used to limit the current peaks that might occur when the DC motors are switched on.

A CAN bus is used to communicate with the boards: a standard personal computer, on which the control software runs, sends through the CAN bus\footnote{A custom protocol was developed in order to allow the bidirectional communication between the control boards and the machine.} the operative commands and receives informations about the status of the machine.

\begin{figure}[h]
\centering
\includegraphics[width=\textwidth]{figures/figure431.png}
\caption{The electronic control unit.}
\end{figure}
4. MULTILAYER DIELECTRIC ELASTOMER ACTUATORS

4.3.5 The control software

The electronic control box is connected to a standard personal computer through a CAN interface. A graphical interface was developed in C++ in order to control the parameters of fabrication process and monitor its status (Fig. 4.32). The main parameters are:

- Number of layers: it’s the number of the layers that constitute the stack. The minimum value for this parameter is three, that corresponds to one active electrode layer and two protective insulation layers on the extremities of the stack. The maximum number of layers is limited by the maximum thickness of the stack, that is 1cm (that corresponds to 200 layers for the 50um-thick VHB 9460 elastomer).

- Number of devices: it’s the number of stacks fabricated simultaneously in the same roll. The minimum number of stacks is one, while the maximum number depends by the size (length) of the single device. A graphic representation of the roll is shown in the control panel, allowing the user to determine the optimal number of devices that can be fabricated on the same roll.

- Actuator length/shift: it represents the linear length (expressed in mm) of the electroded area. At the extremities of this area, the electrode is shifted, according to an alternate pattern, in order to allow the lateral connection of the electrodes.

- Tape/liner thickness: it’s the thickness (in microns) of the dielectric elastomer tape and of the protective liner. The control software takes in account these parameters in order to determine the height of the airbrush’s mask at each turn of the main mandrel.

- Empty layers: it’s the number of additional dielectric layers that can be interleaved between two electrodes. For additional information see Section 4.3.2

- Spray speed/rotation speed: It’s used to control the speed of the winding motors during the deposition of the electrodes.

The graphical interface also allows to monitor the current status of advancement of the fabrication process, by showing: the number of the current layer under fabrication, the
time required to complete the fabrication process, the position of the motors, the air pressure inside the fluid tank etc.

4.4 Fabrication process

The fabrication process of the actuator is divided into two different steps. The first step is the fabrication of the multilayer stack using the semi-automatic machine described in section 4.3. The second step is the fabrication of the lateral contacts and the packaging of the stack into the final actuator.

4.4.1 Fabrication of the multilayer stack

The fabrication process of the multilayer stack actuator is described schematically by the algorithm depicted in Fig. 4.33.

First of all, a spool of raw dielectric tape is loaded in the machine and the fluid reservoir of the airbrush is replenished with the conductive liquid elastomer. The first part of the spool is typically constituted of protective liner only, so the liner is advanced until the section of elastomer begins. The fabrication process is then started by setting the desired parameters in the GUI, and clicking on the button start. The subsequent operations are completely automatized by the machine: the liner is peeled from the elastomeric dielectric by the liner-collecting mandrel, while the dielectric elastomer sticks on the main mandrel. At regular intervals, corresponding to pre-calculated angular position of the motors, the airbrushed is activated and the compliant electrodes are deposited onto the dielectric film. When one layer is complete, the mask is raised (according to the new radius of the roll), and the next layer is fabricated. Depending if the layer’s number is odd or even, an appropriate shift is applied to the angle at which the electrode is supposed to be fabricated, in order to obtain an alternate pattern that allows the subsequent connection of the electrodes (see Section 4.4.3).

It is important to notice that, in order to obtain stacks that can be properly cut from the multilayer roll, the positions of the starting and ending points of the electrodes must be calculated layer by layer according to a well-defined algorithm. To better explain this concept, let’s consider the example shown in Fig. 4.34, in which multiple stacks are fabricated simultaneously at different angular positions of the roll. Let’s call:
4. MULTILAYER DIELECTRIC ELASTOMER ACTUATORS

Figure 4.32: Screenshot of the graphical interface used to monitor the fabrication process of the multilayer actuators.
4.4 Fabrication process

Figure 4.33: The algorithm used by the machine for the fabrication of electroded elastomer roll.
4. MULTILAYER DIELECTRIC ELASTOMER ACTUATORS

- \( s_i \), with \( i=1,2,...,n_{\text{stacks}} \), the generic stack of the roll \( n_{\text{stacks}}=5 \) in the example of fig. 4.34.
- \( l_j \), with \( j=1,2,...,n_{\text{layers}} \) the generic layer of the roll \( n_{\text{layers}}=8 \) in the example of fig. 4.34.
- \( r_0 \) the radius of the support pulley (no dielectric layers).
- \( r_j \), with \( j=1,2,...,n_{\text{layers}} \) the radius of the roll at the generic layer \( l_j \).

![Diagram of multilayer roll](image)

**Figure 4.34:** Example of multilayer roll fabricated through the described process. In this example five stacks of eight layers each are fabricated simultaneously.

Now let’s consider the first stack, called \( s_0 \). During the rotation of the mandrel, the machine fabricates the negative electrode on the layer \( l_0 \), beginning at the angular position \( \alpha_b \) and ending at \( \alpha_e \). The positive electrode, instead, will be fabricated on the next layer \( l_1 \), starting at the position \( \alpha_b + \gamma / 2 \) and ending at \( \alpha_e + \gamma / 2 \), where \( \gamma \) represents the shift angle between the electrodes (necessary for contacting the opposite electrodes) (fig. 4.34). According to the finite thickness of the dielectric layer, the radius \( r_j \) of the roll will increase each turn. For this reason, if \( \alpha_b \) and \( \alpha_e \) are kept constant, the electrodes will increase in length at each turn, resulting in a stack whose electrodes are impossible to contact (fig. 4.35).

To obviate this problem, the control software recalculates the values of \( \alpha_b, \alpha_e \) and \( \gamma \) for each stack layer \( s_i \) and for layer \( l_j \), in order to keep constant not the angles, but
the length of the electrodes (Fig. 4.36). In particular, by knowing the user-defined parameters:

- $t$: thickness of the used dielectric tape.
- $e$: linear length of the fabricated electrodes.
- $o$: linear shift between positive and negative electrodes.

we can calculate the radius $r_j$ of the generic layer $l_j$:

$$ r_j = r_0 + t \ast j $$

(where $r_0$ is radius of the support pulley)

and the starting/ending angular position of each electrode:

$$ \alpha_b = \delta - [e \ast 360/(r_j \ast 6.28)]/2 + \gamma_j/2 + 360 \ast j $$

$$ \alpha_e = \delta + [e \ast 360/(r_j \ast 6.28)]/2 + \gamma_j/2 + 360 \ast j $$

where $\delta$ specifies the position of the stack $s_i$ in the roll and is calculated as:

$$ \delta = 360/n_{stacks} \ast i $$
and $\gamma_j$ is the desired linear shift of the electrodes $o$, adjusted to the current radius of the roll $r_j$:

$$\gamma_j = o \times 360 / (r_j \times 6.28)$$

**Figure 4.36**: The linear length of the electrodes is kept constant for all layers, despite of the different angular position at which the electrodes are fabricated ($\alpha_{b1} < \alpha_{b7}, \alpha_{e7} < \alpha_{e1}$).

Using these simple equations, the starting and ending angles of the sectors to be electroded are determined, and the process is repeated until the last layer is reached.

An alternative procedure to the one described in this section, consists in splitting the fabrication of the roll in two phases. In the first phase, the electrodes are fabricated on the top of the dielectric elastomer, but the liner is not removed, and the elastomer together to the protective liner is wound on the main mandrel. In the second phase of this process, the electroded roll is removed from the main mandrel and again processed by the machine, but this time no electrodes are deposited on the elastomer, the liner is removed, and the final roll (equivalent to the one fabricated with the previously described process) is obtained (Fig. 4.37). One advantage of this fabrication process is the possibility of examining the quality of the fabricated electrodes, in order to tune parameters such as the amount of sprayed material or the winding speed.
Figure 4.37: On the left: An image of the machine during the removal phase of the protective liner from a previously electroded roll. This two-step fabrication process allows the examination of the quality of the fabricated electrodes before the removal of the protective liner (right).
4. MULTILAYER DIELECTRIC ELASTOMER ACTUATORS

4.4.2 Interleaved dielectric layers

The presence of defects in the raw elastomer tape can significantly lower the breakdown strength of the dielectric and eventually cause the premature failure of the actuator. Fig. 4.38a shows the case in which a puncture breakdown occurs due to the presence of a defect inside the dielectric film.

![Figure 4.38](image)

**Figure 4.38:** (a) Occasional defects in one layer of the dielectric elastomer can cause the premature failure of the actuator. (b) With two (or more) dielectric layers in series, the probability of having aligned defects that can cause a dielectric breakdown is small.

However, if two dielectric layers are stacked together (Fig. 4.38b), the probability of having two defects placed on top of each other is small, thus reducing the probability of burning the actuator at defects.

The principle can be extended for the fabricated multilayer actuators described in this chapter, by applying interleaved dielectric layers during the winding process of the elastomeric roll. This concept is described schematically in Fig. 4.40 where standard actuators (no interleaved layers) and multilayer stacks with one and two interleaved layers are represented.

The graphical control interface allows to set the number of interleaved layer by simply changing the corresponding parameter. Multilayer DEAs with one and two interleaved layers were fabricated in this way and tested for dielectric breakdown. The plot presented in Fig. 4.40 show the typical breakdown voltage for a set of tested VHB 9460 multilayer actuators with and without interleaved layers. In this test, no prestrain was applied to the dielectric film. It can be noticed that, although the maximum dielectric strength (35kV/mm) is not affected by the number of interleaved layers, the number of defective devices (i.e. device with breakdown strength ≤ 30kV/mm) is greatly reduced by the introduction of interleaved layers. The drawback of this solution,
4.4 Fabrication process

Figure 4.39: Schematic representation of a multilayer roll with no interleaved layers (left), one interleaved layer (middle), two interleaved layers (right).

however, is the increase of the applied actuation voltage, from 1750V (no interleaved layers) to 3500V (one interleaved layer) and 5250V (two interleaved layers).

4.4.3 Packaging of the actuator

After having reached the desired number of layers, the manufactured roll is kept to rest until the sprayed conductive electrodes are fully cured. The duration of the curing process varies depending on the materials used as dielectric elastomer substrate and as compliant electrodes. At least few hours of rest are advisable in order to achieve a full cure of the polymer and a good mechanical bonding between the layers. The roll manufactured in the first step of this process is then cut radially in order to obtain the number of multilayer stack actuators decided during winding phase (Fig. 4.41). In fact, as previously discussed, multiple stacks can be fabricated simultaneously during the winding phase of the dielectric elastomer. The sides of each stack (Fig. 4.42a) are then trimmed in order to properly exposes the edges of the electrodes (Fig. 4.42b). Finally the two lateral sides of the multilayer stack are airbrushed with the same conductive elastomer used for the electrodes fabrication, in order to connect all the electrodes to the positive and negative power supply cables (Fig. 4.42c). The actuator is finally packaged by spraying a thin layer of insulating elastomer on its surface, in order to completely insulate the device from the external environment and to protect the connections of the electrodes from accidental damage (Fig. 4.43).
4. MULTILAYER DIELECTRIC ELASTOMER ACTUATORS

**Figure 4.40:** Interleaved layers decrease the probability of breakdown due to the presence of defects in the dielectric layer.

**Figure 4.41:** On the left: the electroded roll fabricated by the machine. On the right: one of the fabricated multilayer stacks is cut from the roll.
Figure 4.42: packaging procedure for multilayer actuators: a) The stack is cut from the roll. b) The edges of the stack are trimmed in order to expose the electrodes. c) The electrodes are connected together by manually spraying a layer of conductive elastomer.

Figure 4.43: A packaged multilayer actuator, with the power cables connected on the two sides of the stack.
4. MULTILAYER DIELECTRIC ELASTOMER ACTUATORS

4.5 Experimental Results

4.5.1 Connection of the electrodes to the lateral supply rail

One of the most important issues for this kind of multilayer actuators with discontinuous structure, is the interconnection between the electrodes and the lateral feeding line. If one or more electrodes are not properly powered, in fact, the relative layers will not be actuated, causing a reduction of the performances of the whole device. For this reason, it’s important to evaluate the quality of the lateral connections to understand if all the actuating layers are effectively connected to the power line. A first visual inspection at the optical microscope is presented in Fig. 4.44 and Fig. 4.45. In these images, representing two different sections of a multilayer actuator, it’s possible to notice that a clean cut, made with a sharp scalpel, is sufficient to expose the edges of the electrodes that must be contacted by the supply rails.

![Image](image_url)

**Figure 4.44:** On the left: a section of the lateral area of the stack, in which only the positive electrodes are present. On the right: a section of the central area of the stack, in which both positive and negative electrodes are present.

It’s impossible however to determine by visual inspection if all the electrodes have been connected after the fabrication of lateral feeding lines, since the sprayed conductive layer hides the underlying electrodes. The number of the effectively connected layers can be however estimated by measuring the capacitance of the whole device. In fact, since the stack is electrically equivalent to a set of capacitors connected in parallel,
4.5 Experimental Results

Figure 4.45: A section of a multilayer stack (30 layers). The thickness of the dielectric layers (VHB 9460 acrylic elastomer) is 50um.

The total capacitance between the two feeding lines is expected to be a function of the number of layers of the device (Fig. 4.46)

Figure 4.46: Schematic representation of a multilayer actuator as a set of capacitors connected in parallel. In the picture one lateral connection is damaged, causing a reduction of the total capacitance of the device.

For an ideal capacitor constituted by two planar, parallel conductive plates, we have:

\[ C = \varepsilon_0 \varepsilon_r \frac{A}{d} \]

so, if we ignore the border effects on sides of the electrodes, we can approximate the total capacitance of a multilayer stack composed by \( n \) layers electrically connected in parallel:
4. MULTILAYER DIELECTRIC ELASTOMER ACTUATORS

\[ C_{\text{stack}} = \epsilon_0 \epsilon_r \frac{A \cdot n}{d} \]

Figure 4.47 shows the experimental measurements of the capacitance of different devices (from 1 to 20 layers) respect to the theoric capacitance calculated from the equation presented above. According to the plot, the measured capacitance is about 10% lower than the theoric value, a value that can be explained considering the uncertainty on the effective size of the electrodes (that are sprayed through a mask, and thus present shaded edges). This hypotesys is confirmed also by the fact that, for larger electrodes, the uncertainty on their effective size is smaller and the difference between the expected and measured capacitance decreases as well.

![Figure 4.47: Plot of the expected (teoric) and measured capacitance of multilayer devices (area of each single electrode: 1 cm²).](image)

The plot of the measured capacitance presented above is obtained as a mean between all the samples with capacitance that differs no more than 15% from the teoric capacitance value. However, few samples presented a measured capacitance 20% lower than the teoric values. These anomalous values of capacitance can be explained if we consider the possibility of having one or more unconnected electrodes that lower the overall capacitance of the device. By considering the ratio between the expected and measured capacitance, we can determine the number of the effectively connected
4.5 Experimental Results

electrodes. Fig. 4.48 shows a statistic analysis performed on 40 fabricated samples, indicating that the number of devices with one or more damaged power connections is about 20% of the total. On these ‘defective’ devices, a further experiment was performed by cutting a thin stripe of material on the side of the actuator and spraying a new feeding line to reconnect the missing electrodes. The measured capacitance of all the defective devices ‘repaired’ in this way was in the expected range, thus confirming the hypothesis of a fabrication defect during the first connection attempt.

Due to the low number of defective samples, no further attempts were done in this thesis to improve the connecting procedure.

4.5.2 Strain measurements

Strain measurements were performed using the same technique presented in Section 3.4.2 i.e. using a digital camera to optically detect the displacement of the actuator. Fig. 4.49, Fig. 4.50 and Fig. 4.51 show the transverse strain for actuators constituted by 5, 10, 15 and 20 layers of VHB 9460 dielectric at different prestrain values (respectively 100%, 200%, 400%). Rubber electrode were fabricated using a mixture of carbon black and polyurethane (3M’s 4200). According to the presented plot, the prestrain affects the performances of the actuators, increasing the maximum applicable
4. MULTILAYER DIELECTRIC ELASTOMER ACTUATORS

electric field (this effect was already observed in single layer actuators, see section [3.2.3]). Moreover, it can be noticed that the actuation strain developed by multilayer actuators decreases as the number of layers increases. An hypothesis on this undesired behavior regards the progressive hardening of the devices due to the presence of an increasing number of polyurethane layers. Since the elastic modulus of the 4200 polyurethane is greater than the one of the VHB 9460, electrodes with range of thickness comparable to the dielectric layer can limit the electrically induced strain. Finally it can be noticed that the maximum prestrain applied to multilayer DEAs was smaller respect to the case of single layer actuators and that the maximum applied electric filed was thus limited to 70V/um. The reasons for this limitation are explained in the following section.

![Graph](image_url)

Figure 4.49: Strain measurements for multilayer actuators with 100% prestrain.

4.5.3 Considerations on the prestrain of multilayer actuators

Prestraining the actuator is a useful technique to simultaneously decrease the thickness of the dielectric layer and increase its dielectric strength (Section [3.2.3]). In this way, the performances of single layer actuators can be greatly enhanced, because higher electric fields can be applied to the device, resulting in an increase of the developed Maxwell stress (Section [3.4.2]).

However, multilayer actuators imply fabrication issues and consideration that are different respect to the case of single layer devices. In fact, while single layer actuators
4.5 Experimental Results

Figure 4.50: Strain measurements for multilayer actuators with 200% prestrain.

Figure 4.51: Strain measurements for multilayer actuators with 400% prestrain.
4. MULTILAYER DIELECTRIC ELASTOMER ACTUATORS

can be easily prestretched on a support frame and then electroded with a manual airbrush, the electrodes of multilayer actuators are deposited between the dielectric layers during the fabrication of the roll, and the device can be stretched only at the end of the process, when the stacks are cut from the support. The resistance of the electrodes of the two kinds of devices is extremely different. The electrodes of single layer devices are not prestrained and thus maintain high level of conductivity. On the contrary, for multilayer actuators the prestrain is applied also to the rubber electrodes, resulting in an increase of their resistivity. Fig. 4.52 shows the dependence of the resistivity of the rubber electrodes on the applied biaxial prestrain. According to the presented plot, the nominal resistance for unstrained rubber electrodes, constituted by a mixture of 3M’s 4200 polyurethane and carbon black (28% loading) is 5KΩ. This resistivity rises to 1MΩ when the electrode is biaxially prestrained to 200% x 200%. Finally, the electrode looses most of its conductivity when a biaxial prestrain of 300% x 300% is applied (measured resistivity greater than 7MΩ). Because of this severe limitation on the maximum applicable prestrain, it was not possible for multilayer DEAs to exploit the same high breakdown strengths of single layer actuators, resulting in a reduced actuation performance.
4.5 Experimental Results

Figure 4.52: Dependence of the resistivity of the electrodes on the applied biaxial pre-strain.
4. MULTILAYER DIELECTRIC ELASTOMER ACTUATORS
Conclusion

Dielectric elastomers are a promising class of electroactive polymers able to exhibit large deformations when electrically stimulated. Their intrinsic compliance is particularly attractive for robotic application, in which safe interaction between the machine and the working environment is a primary issue. This cannot be typically obtained with existing actuating technology, since the robots are designed primarily as rigid position or torque sources and most interaction skills are imposed by virtue of control software. On the contrary, dielectric elastomers actuators are fabricated from soft, rubber-like materials (hence the term elastomers), and are thus intrinsically compliant; moreover, despite to other classes of EAPs, DEAs can operate repeatedly in non-aqueous environment, with fast activation speeds and high levels of efficiency.

The operating principle of DEAs is described in the first part of this thesis. In particular it is shown how the actuating behavior of the device depends on the balance between the passive restoring force of the elastomer and the active electrically induced Maxwell stress. This latter depends on the square of the applied voltage and on the dielectric constant of the material. Other factors, such as the intrinsic dielectric strength of elastomer and the presence of occasional defect in the insulating layer, affects the maximum electrical field that can applied to the actuator without experiencing the breakdown of the dielectric. Because all of these properties (i.e. the dielectric strength, the permittivity, the elastic modulus) are intrinsic properties of the chosen dielectric elastomer, an analysis of the performances of different materials must be done using a simple actuating configuration as reference setup.
5. CONCLUSION

A single-layer geometry was used as test-bench to evaluate the actuating performances of DEAs fabricated using different techniques and materials. More in particular, both the mechanical and the dielectric properties of silicone (ECOFLEX-30) and acrylic (3M VHB) elastomers were experimentally investigated. Different types of compliant electrodes were also investigated: dust electrodes, grease electrodes and rubber electrodes. It was found that sprayed rubber electrodes, fabricated using mixtures of conductive carbon black and different insulating elastomers above the percolation threshold, offer the best mechanical stability. Moreover compliant rubber electrodes can find useful application in in pressure-sensing devices based on capacitive technology.

The actuating performances of silicone and acrylic single-layer actuators were measured and compared. High values of electrically-induced stress/strain were observed for prestrained acrylic devices, due to an higher dielectric strength that allows the the material to withstand intense electric fields. On the contrary, silicone actuators were characterized by lower breakdown strengths and lower actuating strains, even if their response was faster than the one observed for acrylic actuators. For VHB actuators, in fact, the viscous losses of the acrylic elastomer limit the maximum achievable bandwidth.

VHB acrylic elastomer is commercially available in long rolls of raw material, with thickness ranging from 1mm (VHB 4910) down to 50um (VHB 9460). This suggested the possibility of developing a novel semi-automated fabrication process, able to process the raw dielectric elastomer and fabricate multilayer stacked actuators. To accomplish this goal, a programmable machine able to deposit compliant rubber electrodes on the top of the processed dielectric film was designed and developed.

A graphical control interface was also developed, in order to monitor all the fabrication parameters in an easy and convenient way. In particular, this novel fabrication process allows the fast, simultaneous fabrication of many actuators, whose sizes and geometries can be selected from the control interface.

Fabricated multilayer actuators were visually inspected under the microscope, to verify the quality of the fabrication process. It was found that, while the internal structure of the electrodes virtually presents no defects, a critical issue is the lateral connection of the electrode to the common supply rail. Since the capacity of the device is proportional to the number of the effectively connected layers, its measurement was adopted as a diagnostic criterion to determine the presence of unconnected layers.
inside the actuator. It was also demonstrated that defective actuators can be repaired by simply cutting away the damaged lateral connections and spraying a new layer of conductive elastomer.

The actuating performances of multilayer actuators fabricated from a 50um-thick film of acrylic elastomer (VHB 9460) are presented in the last part of this thesis. For these devices it was not possible to apply large biaxial prestrains like in the case of single layer actuators, since the resistivity of the prestrained electrodes raises too much. Because of this, lower electric fields were generally applied, in order to prevent a premature electrical breakdown of the multilayer actuators. Moreover, it was found that the actuating strain decreases proportionally to the number of the layers of the device. A possible interpretation of this unexpected behavior considers the elastic modulus of the fabricated polyurethane electrodes, that is higher respect to the one of the dielectric elastomer. Further investigation is however required to explain this behavior, by testing different electrode materials as rubber electrodes. Presently, only 3M’s 4200 polyurethane was considered successful in gluing together the layers of the stacked actuators, so it was not possible to determine if the same effect was also present with electrodes fabricated from different, more compliant elastomers.

Additionally, in this thesis only multilayer acrylic actuators were investigated. However, the developed semi-automatic fabrication process is generally able to process other kinds of raw dielectric elastomer. Silicone actuators are particular attractive, because they can operate at higher speed respect to acrylic one (although their lower dielectric strength limits the maximum achievable strain). However, since the machine is able to process only film-shaped materials, long rolls of silicone elastomer must be firstly fabricated with a rotocuring process. This last technique will also enable the employment of custom-formulated elastomers whose electromechanical properties can be tuned to maximize the performances of the fabricated multilayer actuators. This is an open field of research.
5. CONCLUSION
References


REFERENCES


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