# Mechanics of Soft Actuators

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Mechanics of Soft Actuators

A dissertation presented by

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to

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Abstract

This dissertation explores the mechanics of three types of soft actuators experimentally, and theoretically: (i) dielectric elastomer transducers; (ii) fluid-driven actuators; (iii) paper actuators. The dissertation develops new control elements for these actuators, which extend their function, and enable the design of devices with entirely new functions.

A stretchable, transparent conductor is introduced, which is based on an ionically conducting hydrogel. This conductor can operate at frequencies above 10 kHz, voltages above 10 kV without electrochemical reactions, and has a superior optical transmittance compared to stretchable, electronic conductors of comparable sheet resistance. A transparent dielectric elastomer loudspeaker is demonstrated, that generates sound across the entire audible frequency range (20 Hz to 20 kHz). The acoustic properties of the transparent loudspeaker are characterized inside an impedance tube for low frequency active noise cancellation. A linear model is developed, which describes sound reflection, transmission, and generation by the transparent loudspeaker. In an active noise cancellation experiment with feedforward control, which is based on the linear model, the sound transmission loss across the membrane improves compared to passive sound absorption by a factor of 3. The durability of dielectric elastomer transducers under electromechanical actuation is investigated, when ionic liquids are used as the conductors. Because
ionic liquids diffuse into the dielectric membrane, the capacitance of the transducers changes with time, which can be described with a linear diffusion model. The larger the increase in capacitance, the shorter is the lifetime of the transducers.

An entirely soft valve for the autonomous control of pneumatic, soft devices is developed. The valve uses the snap-through instability of an approximately hemispherical membrane to switch air-flow through the valve. In a feedback circuit, the hysteresis of the snap-through instability generates oscillations of pressure using air from a source of constant pressure. Its use for soft robots is demonstrated: (i) in a soft gripper, which autonomously grasps objects upon contact; (ii) in a soft earthworm, which advances autonomously using a source of constant pressure.

Finally, actuators are developed, which use the hygro-expansion of paper to generate bending motion. The actuators consist of paper with an integrated electrically conducting channel of PEDTO:PSS, which is bonded to an adhesive tape. When a voltage is applied to the conductive channel, resistive heating causes evaporation of water from the paper. The paper contracts, and the actuator bends. The behavior of these actuators is investigated experimentally and theoretically. The use of the actuators is demonstrated in an optical shutter, and in the manipulation of liquids.
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Chapter 1

Introduction

This dissertation investigates the mechanics of soft actuators. Soft actuators have enabled the design of soft robots with functions that are difficult to achieve with hard robots. Whereas hard robots—robots, which consist of stiff, typically metallic, structures (Young’s modulus $E \sim 100$ GPa)—are successfully used for well-defined tasks, requiring high forces and speeds, such as manufacturing, they require sensory feedback, and complicated control in unstructured environments.$^{1,2}$ They are without special provisions also non-collaborative, and dangerous to people.$^3$ Soft robots—robots, which consist either of materials that are intrinsically soft ($E < 1$ GPa), or which consist of stiff components that give the robot due to their shape, or interconnection an extrinsic compliance—embody some of this feedback directly into their structure (“material intelligence”).$^{1,2}$ Handling of an object of unknown shape, for example, requires feedback control in hard actuators, whereas soft actuators automatically conform to the shape of the object, and limit the force exerted.$^{2,4}$ This compliance makes soft robots also safe in the interaction with humans ($i.e.$, collaborative).$^{2,4}$

Numerous different types of soft actuators have been developed, often inspired by nature, which provides countless examples of soft organisms, and actuation schemes ($e.g.$, octopus arm, elephant trunk, and inchworm body).$^1$ Even though many designs of soft actuators mimic functions of biological organisms, they do not necessarily try to copy their mechanisms. Instead, they draw from a large pool of physical mechanisms, and from phenomena occurring in soft materials. Among these mechanisms are mass transport ($e.g.$, swelling, diffusion),$^{5-7}$ instabilities ($e.g.$, buckling, snap through),$^{8-10}$ and responses to various physical stimuli ($e.g.$, pressure, light,
temperature, pH, electric, and magnetic fields).\textsuperscript{6,11–15} It is beyond the scope of this chapter to discuss all the known actuation mechanisms in detail. Instead, the remainder of this chapter provides an overview of the three classes of actuators, which are investigated in this thesis: (i) dielectric elastomer transducers (Chapters 2 - 4); (ii) fluid-driven actuators (Chapter 5); (iii) paper actuators (Chapter 6).

\subsection{Dielectric Elastomer Transducers}

Dielectric elastomer transducers consist of electrically insulating, stretchable, elastomeric membranes that are coated on both sides with compliant electrodes (Figure 1.1A).\textsuperscript{14} When a voltage ($V$) is applied between the electrodes, an electric field ($E$) arises across the membrane, which results in a Maxwell stress

$$\sigma = \varepsilon E^2$$

(1.1)

in thickness direction, where $\varepsilon$ is the dielectric constant of the elastomer.\textsuperscript{14,16} The Maxwell stress causes a reduction in thickness, and an expansion in area of the elastomer (Figure 1.1B). Soft actuators, which use this principle, have been used for a wide range of applications including walkers,\textsuperscript{17,18} grippers,\textsuperscript{19–21} deformable lenses,\textsuperscript{22} a braille display,\textsuperscript{23} and loudspeakers.\textsuperscript{24–30}

Dielectric elastomer transducers also behave like stretchable capacitors with capacitance

$$C = \frac{\varepsilon A}{H},$$

(1.2)

where $A$ is the area of overlapping stretchable electrodes, and $H$ the thickness of the dielectric membrane (Figure 1.1A). When the membrane deforms, $A$, $H$, and consequently $C$ change. Dielectric elastomer transducers can therefore also be used to measure deformation, and force as changes in capacitance,\textsuperscript{31–36} and to generate electrical energy from the mechanical deformation of the dielectric membrane.\textsuperscript{37–41}
Figure 1.1 Structure of a dielectric elastomer actuator. (A) A dielectric elastomer actuator consists of an insulating elastomeric membrane (area $A$, thickness $H$), coated on both sides with stretchable electrodes. (B) When a voltage $V$ is applied between the electrodes, the membrane becomes thinner, and expands in area.
The most common electrodes for dielectric elastomer transducers are made of electronic conductors.\textsuperscript{42,43} These conductors struggle to achieve high optical transparency.\textsuperscript{42,43} Chapter 2 reports a stretchable, transparent ionic conductor based on a hydrogel.\textsuperscript{24} This conductor has a higher optical transmittance to visible light (> 99 %) than previously reported stretchable, electronic electrodes with comparable sheet resistance. We use this conductor to fabricate a transparent dielectric elastomer loudspeaker, that generates sound across the entire audible spectrum (20 Hz to 20 kHz). Chapter 3 investigates the acoustic properties of a transparent membrane, based on the design of the transparent loudspeaker presented in Chapter 2, and demonstrates its use for active noise cancellation. Chapter 4 investigates the durability of dielectric elastomer transducers under electromechanical excitation, when ionic liquids instead of hydrogels are used as electrodes.

1.2 Fluid-Driven Soft Actuators

Fluid-driven (pneumatic, or hydraulic) soft actuators are in principle inflatable balloons, whose shape, and material give their deformation directionality, and enable function. Various designs of fluid-driven actuators exist, of which pneu-nets, and fiber-reinforced elastomeric actuators are the most common.\textsuperscript{2,4,44–46} Pneu-nets are networks of inflatable channels integrated into an elastomeric actuator. When the actuator is connected to a pressure source, the channels distribute air inside the actuator, and inflate (Figure 1.2A).\textsuperscript{11,47} The geometry of the channels determine the deformation of the actuator.\textsuperscript{47} Pneu-nets are often combined with strain limiting layers (e.g., paper infused with elastomer) to enhance, and direct their deformation.\textsuperscript{11} Fiber-reinforced elastomeric actuators consist, usually, of tubular, elastomeric balloons with embedded inextensible fibers (Figure 1.2B).\textsuperscript{48} The orientation of the fibers determines the mode of actuation.
Figure 1.2 Two types of fluid-driven actuators. (A) Pneu-nets consist of channels embedded in an elastomer, and a strain limiting layer, which govern the deformation, when the actuator is pressurized. (B) A linear fiber reinforced elastomeric actuator. The actuator consists of an elastomeric tube with integrated inextensible fibers. The fibers constrain radial expansion, but allow axial expansion.
(e.g., rotation, extension, contraction, and bending). Fluid-driven actuators can also work with negative pressure (vacuum).  

Fluid-driven soft actuators have been used in the design of robots, which walk,\(^4\) jump,\(^5\) swim,\(^6\) and grasp.\(^7\) The size of fluid-driven actuators can be scaled, which enables micrometer size pumps,\(^8\) and millimeter size grippers.\(^9\) Because fluid-driven soft actuators have elastic moduli similar to biological tissue, they are safe in their interaction with humans.\(^1\) This property has led to the development of wearable robots for rehabilitation,\(^10\)\(^11\) and even a robotic sleeve to support the beating of a heart.\(^12\)

In many of the listed applications, fluid-driven soft actuators work between two pressure-limit set-points, which reduces control to a simple “on/off”-valve.\(^13,14\) Even though the actuators are soft, they still rely mainly on hard valves for control. Such valves are usually placed outside of the soft actuator.\(^2\)\(^4\) Chapter 5 describes a soft valve that uses a bistable membrane to switch between two pressure levels. This new type of valve can be integrated directly into soft actuators, and enables automated feedforward and feedback control. Its characteristics are demonstrated with a gripper, that automatically grasps objects upon contact, and a soft earthworm, which advances using a source of constant pressure.

1.3 Paper Actuators

Paper is an abundantly available material, which consists of a network of cellulose fibers that give paper its mechanical properties.\(^15\) Paper is stiff \((E \sim 1\) GPA\()\), but when processed into thin sheets (usually < 1mm), bendable, and foldable.\(^16\) Felton \textit{et al.} designed a paper-based robot, that autonomously unfolded itself from a flat, two-dimensional state to its final three-dimensional geometry.\(^17\) Paper acts as the strain-limiting layer in many pneu-net actuators, and can, when
folded with origami techniques, enable different modes of deformation (e.g., extension, bending, contraction, rotation).  

Even though paper is mainly used as a structural element in soft actuators, actuation mechanisms have also been directly integrated into paper. A design for a paper actuator uses paper strips soaked with a ferromagnetic fluid, which bend, when exposed to a magnetic field.\textsuperscript{15} Chen \textit{et al.} made paper electrically conductive by imbibing it with carbon nanotubes.\textsuperscript{60} Two of these conductive paper layers, which were separated by a dielectric layer, “zipped up” due to electrostatic forces, when a voltage was applied between them. When paper is coated on both sides with conductive electrodes, and a voltage is applied between the electrodes, ions dissolved in the paper diffuse to the electrodes, which leads to an asymmetric volume change, and thus bending of the actuator.\textsuperscript{7,61,62} When the cellulose fibers are aligned, paper also exhibits piezoelectric behavior.\textsuperscript{61}

Paper absorbs water from the environment, which leads to swelling of the cellulose fibers, and to an increase of the dimensions of the paper (strains on the order of $\sim 0.1\%$). This hygroscopic expansion is reversible (\textit{i.e.}, when the water content in the paper decreases, it contracts). Chapter 6 describes an electrically activated paper actuator that uses hygroscopic expansion to generate bending motion.\textsuperscript{63} The actuator consists of paper with an integrated electrically conducting channel bonded to a strain limiting layer. When a voltage is applied to the conducting channel, resistive heating evaporates water in the paper layer. The evaporation of water leads to contraction of the paper layer, and consequently to bending of the actuator.
Chapter 2

Stretchable, Transparent, Ionic Conductors*

2.1 Introduction

The emergence of the field of stretchable electronics, along with its biomedical applications, has highlighted a challenge: Electronic devices are usually made of hard materials, whereas tissues and cells are soft. Stretchable conductors are needed to enable electronics to meet skin, heart, and brain. Stretchable conductors are also needed in applications, such as electro-mechanical transduction and solar energy conversion. Existing stretchable conductors are mostly electronic conductors, including carbon grease, microcracked gold films, serpentine-shaped metallic wires, carbon nanotubes, graphene sheets, and silver nanowires. Stretchable devices have also been made by using corona discharge, liquid metals, and saline solutions; these conductors, however, are not solid, and their uses are limited. Attributes other than conductivity and stretchability are also important in specific applications. Conductors may need to operate at high frequencies and high voltages, remain conductive while undergoing areal expansions of 1000 % or more, be biocompatible, and be transparent.

Whereas electronic conductors struggle to meet these demands, ionic conductors meet most of them readily. Many ionic conductors, such as hydrogels and gels swollen with ionic liquids, take a solid form and are stretchable and transparent. Many hydrogels are biocompatible and conformal to tissues and cells down to the molecular scale. In this chapter we demonstrate that

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*This Chapter is a modified version of the publication Keplinger, C.; Sun, J.-Y.; Foo, C. C.; Rothemund, P.; Whitesides, G. M.; Suo, Z.: Stretchable, Transparent, Ionic Conductors, Science, 2013, 341, 984-987. The text of the publication was modified to fit within the format of the thesis, and a part of the supplementary information of the publication integrated into the main text. The remaining supplementary information of the publication is appended to this dissertation as Appendix A.
ionic conductors can even be used in devices requiring voltages and frequencies much higher than those commonly associated with devices using ionic conductors.

2.2 Experimental Details

2.2.1 Materials

Unless otherwise specified, experiments were carried out using copper as the electrode, VHB 4910 (3M) as the dielectric, and NaCl-containing polyacrylamide hydrogel as the electrolyte.

The hydrogel was synthesized using acrylamide (AAm; Sigma, A8887) as monomers, N,N-methylenebisacrylamide (MBAA; Sigma, M7279) as crosslinkers, ammonium persulfate (AP; Sigma, A9164) as photo initiator, and N,N,N’,N’-tetramethylethylenediamine (TEMED; Sigma, T7024) as crosslinking accelerator. AAm and NaCl were dissolved in deionized water, with AAm fixed at 2.2 M, but NaCl varied from 1.37 to 5.48 M. Also added were MBAA 0.0006 the weight of AAm, and AP 0.0017 the weight of AAm. After degassing in a vacuum chamber, TEMED 0.0025 the weight of AAm was added. The solutions were poured into a 100.0 x 100.0 x 0.1 mm³ glass mold, and covered with a 3-mm thick glass plate. The gels were cured using an ultraviolet light crosslinker (UVC 500, Hoefer) for 20 min with power 8 W and 254 nm wavelength. The gels were cut into the designed shape by using a laser cutter (VersaLaser VLS3.50, Universal Laser Systems) with a power of 50 W and a speed of 14 cm/sec.

Dielectric elastomer actuators were also fabricated using two other types of conductors: carbon grease (MG Chemicals Carbon Conductive Grease: 846-80G), and an ionic liquid (1-Decyl-3-methylimidazolium chloride, [C₁₀MIM][Cl]; Sigma, 690597).
2.2.2 Transparent Actuator

VHB has been marketed as adhesive tapes; this property played a significant role in the fabrication of the devices. The actuator involved layered electrolytic and dielectric elastomers (Figure 2.1). We used fairly thick (100 μm) hydrogel to slow drying. To minimize the elastic constraint of hydrogels, we stacked three layers of the VHB. The layers of VHB were stretched radially to three times the initial radius, and fixed to a circular rigid frame made of an acrylic, inner radius 4 cm for the circular hydrogel and 8 cm for the heart-shaped hydrogel. Before the hydrogel was stacked onto the dielectric, the surface of the hydrogel was dried with nitrogen gas. The removal of water on the surface of the hydrogel enhanced the adhesion between the hydrogel and the dielectric.

We characterized actuators with hydrogels of circular shape. The radius of the hydrogels was 1/4 the radius of the rigid frame, and hydrogels containing 2.74 M NaCl were used. Voltage was generated using a high-voltage amplifier (Model 50/12, TREK) and a function generator. The movements of the hydrogels were recorded using a high-speed camera (Vision Research, Phantom V310).

When the voltage was suddenly applied and was held constant subsequently, the actuator deformed as a function of time, and the rate of expansion became small after about 20 s (Figure 2.2). The expansion in area at 20 s was recorded as a function of the applied voltage. The voltage-induced strain was limited by electromechanical instability, and the elasticity of the hydrogels did not appreciably constrain the dielectric; see the theoretical analysis in Appendix A.

When the voltage was cycled at a certain frequency, the actuator oscillated. The initial cycles were not steady; the amplitude of the oscillating actuator changed from cycle to cycle. After some cycles the actuator oscillated steadily, and the relative change between the maximum and
Figure 2.1 Layered electrolytic and dielectric elastomer, a design of stretchable ionics that enables electromechanical functions without electrochemical reaction. (A) A layer of a dielectric elastomer is sandwiched between two layers of an electrolytic elastomer. The electrodes are placed outside the active region of the device, so that the device is stretchable and transparent. (B) Equivalent circuit of the device. (C) A specific design of an actuator with the electrolyte in the shape of a heart. (D) The heart expands when the voltage is applied.
Figure 2.2 Actuator using hydrogels of circular shape. (A)-(D) When a voltage was suddenly applied and subsequently held constant, the actuator expanded in area over time. (E) At time zero, a voltage of 18 kV is rapidly applied, and is then held constant. (F) The area strain is measured as a function of time, and the experimental data are compared with the theoretical prediction. (G) The area strain measured at 20 s after the voltage is applied. (H) The viscoelasticity of the dielectric elastomer is represented by a rheological model of two parallel units: the top unit consists of spring $\alpha$, and the bottom unit consists of spring $\beta$ in series with a dashpot.
minimum areas defined the area strain. We determined the area strain as the voltage cycled between steps of 0 and 18 kV, at frequency ranging from 0.05 to 1024 Hz (Figure 2.6F).

Movies were recorded with frame rates from 1000 up to 3200 frames per second. To obtain the electrode area single frames of the videos were analyzed with ImageJ. A LED connected to the sync output of the function generator was placed at the corner of the recorded scene to indicate voltage on and off in the video.

In these demonstrations, we used relatively thick layers to make material handling easy, and to demonstrate the high-voltage capability of the design. In many practical devices, one may wish to scale down the thickness and voltage.

Actuators were also made using carbon grease as conductors. The actuators were subject to cyclic voltage, and the area strains were recorded. This response of the actuators using carbon grease as the conductor is nearly indistinguishable from that of actuators using the hydrogel as the conductor (Figure 2.3).

2.2.3 Transparent Loudspeaker

The fabrication process was similar to that of the transparent actuator, but with different dimensions. Three layers of the VHB 4910 were stacked together, stretched to three times the initial radius, and fixed to a circular acrylic frame of radius 10 cm. Hydrogels of radius 5 cm were attached to the surfaces of the dielectric.

The transparent loudspeaker was placed in front of a laptop, which was playing a music video (Figure 2.4). The sound of the music was fed to the loudspeaker as analog electrical signal, from the audio output of the laptop, through a high-voltage amplifier (TREK, model 30/20A; fixed 3000 V/V gain; slew rate larger than 750 V/μs). The electric field deforms the dielectric elastomer by the Maxwell stress, which is quadratic in voltage. To preserve the polarity of the analog
Figure 2.3 An actuator using carbon grease as the conductor is compared with an actuator using the hydrogel as the conductor. The area-frequency curves of the two actuators are nearly indistinguishable.
Figure 2.4 The experimental setup for the transparent loudspeaker. The loudspeaker is placed in front of a laptop playing a music video. The video is clearly visible through the loudspeaker. The sound track of the video is fed to the loudspeaker as a high-voltage signal, from the audio output of the laptop, through a high voltage amplifier. A microphone records the sound produced by the loudspeaker.
electrical signal and prevent frequency doubling, the high voltage amplifier was also programmed to add a 12.5 kV DC offset. The offset also shifted the time-varying signal up, to the region just below the dielectric strength of the elastomer, where the electromechanical coupling was strongest. This high voltage signal was applied to the hydrogel layers. A webcam with a microphone (electromagnetically shielded with a grounded metallic mesh; webcam model: Logitech, QuickCam® Pro 9000) recorded the scene from 15 cm distance. The sound produced from the transparent loudspeaker was clearly audible from 5 m distance.

We study the fidelity of the sound reproduction by feeding the loudspeaker with a 20 s test signal. The signal was of constant amplitude and a linear sine sweep from 20 Hz to 20 kHz. The audio recordings of the video files were extracted in form of WAV (Waveform Audio File Format) files. Spectrograms of the WAV files were used to analyze the sound. The colors show the intensity distribution of frequencies as a function of time (Figure 2.7). The frequency spectrum at a given time was obtained by short-time Fourier transform of the WAV data.

Our transparent, stretchable loudspeaker was intended to illustrate the combination of three attributes of the layered electrolytic and dielectric elastomers: high voltage, a frequency bandwidth large enough to cover the entire audible range, and ultrahigh transparency. For commercial applications, such as transparent loudspeakers placed in front of screens or on windows, the resonance behavior has to be optimized by special frame constructions or alternative geometries. Lowering the membrane thicknesses and using multilayered electrolytic and dielectric elastomers will bring the required driving voltage down to practical levels.

2.2.4 Transmittance of Elastomeric Ionic Hydrogels.

Optical transmission spectra of the hydrogels were measured using a spectrophotometer (DU530, Beckman) with quartz cuvettes for the whole range of visible light. A quartz cuvette with
pure water (≈ 99.99 % transmittance at 550 nm) was used as a reference to reduce the reflection from index mismatch. Since the transmittance of the hydrogels is very high, a normal spectrophotometer cannot measure the transmittance of layers of 100-μm thick hydrogels used in our devices. Therefore, a layer of 11 mm thick hydrogel containing 5.48 M of NaCl was used to measure the transmittance. The transmittance is defined as \( T = I / I_0 \), where \( I \) is the intensity of the transmitted light, and \( I_0 \) is the intensity of the incident light. The transmittance was measured over the visible range of wavelengths (Figure 2.8C).

The transmittance decreases as the thickness of the sample \( l \) increases, and is assumed to obey the Beer-Lambert law:

\[
T = e^{-\alpha l}
\]

where \( \alpha \) is the absorption coefficient of the hydrogel. This expression was used to convert the transmittance measured from the 11-mm thick hydrogel to the transmittance of the 100-μm thick hydrogel.

### 2.2.5 Resistance of Elastomeric Ionic Hydrogels under Stretch

The resistivity of the hydrogels was measured as the hydrogels were pulled by a uniaxial force. The resistance was measured by using four-point probes. To minimize the effect of ions built up on the surface of the probes, the resistance was measured with three relatively large voltages (20 ~ 50 V; electrochemical potentials are much smaller) and the corresponding currents after saturation. Hydrogels containing 1.37, 2.74 and 5.48 M of NaCl were used. When the hydrogels were not stretched, the measured molar conductivity was 120.19 Scm²/mol, which was close to a reported value of 118.5 Scm²/mol for aqueous solutions. The resistivity increased somewhat when the hydrogels were pulled (Figure 2.8A).
To a first approximation, the resistivity of a hydrogel, $\rho$, may be taken to be a constant independent of the stretch $\lambda$ under uniaxial force. We use this approximation to estimate the resistance as a function of the stretch. The hydrogel has length $L$ and cross-sectional area $A$ in the undeformed state, and has length $\lambda L$ and cross-sectional area $A / \lambda$ in the deformed state. Here the hydrogel is taken to be incompressible. The resistance of the hydrogel is $R_0 = \rho L / A$ in the undeformed state, and is $R = \rho \lambda L / (A / \lambda)$ in the deformed state. Thus, the ratio of these two values of resistance is $R / R_0 = \lambda^2$. This expression closely approximates the measured resistance of the hydrogel (Figure 2.8B).

2.3 Results and Discussion

2.3.1 Basic Design of the Stretchable, Ionic Conductor

One basic design places two electrodes (electronic conductors), an electrolyte (ionic conductor), and a dielectric (insulator) in series (Figure 2.5A). The electrode/electrolyte interface forms an electrical double layer (Figure 2.5B). For some combinations of the electrode and electrolyte, if the voltage across the interface is within a certain range ($\sim 1$ V), the interface is ideally polarizable—that is, electrons and ions do not cross the interface, no electrochemical reaction occurs, and the electrical double layer behaves like a capacitor (Figure 2.5C). A circuit with electronic and ionic conductors in series cannot carry sustained electrical current in the absence of electrochemical reaction. The electrical double layer and the dielectric are two capacitors in series. They capacitively couple the electrical signals carried by the two electrodes and transport power with alternating current.

The two capacitors have vastly different capacitances. For the electrical double layer, charges in the electrode and in the electrolyte are separated over nanometers; this small separation leads to a large capacitance, on the order of $c_{EDL} \sim 10^{-1} \text{ F/m}^2$. For the dielectric, charges on its
Figure 2.5 A basic design of stretchable ionics. (A) An electrolyte and a dielectric are in series. When they are elastomeric, the device is solid and stretchable. When a voltage $V$ is applied between the two electrodes, the voltage across the electrode/electrolyte interface is much smaller than the voltage across the dielectric, $V_{EDL} \ll V_D$. (B) The electrode/electrolyte interface forms an electrical double layer. (C) When the voltage across the interface is within a certain range, electrons and ions do not cross the interface, no electrochemical reaction occurs, and the electrical double layer behaves like a capacitor.
two faces are separated over the thickness of the dielectric (millimeters in our experiments); this large separation leads to a small capacitance, on the order of $c_D \sim 10^{-8} \text{ F/m}^2$. At equilibrium, the voltage $V$ applied between the two electrodes is carried entirely by the electrical double layer and the dielectric, $V = V_{\text{EDL}} + V_D$ (Figure 2.5A). In response to the applied voltage, the two capacitors add the same amount of charge, $c_D A_D V_D = c_{\text{EDL}} A_{\text{EDL}} V_{\text{EDL}}$, where $A_{\text{EDL}}$ is the area of the electrical double layer and $A_D$ is the area of the dielectric. In our experiments, $A_{\text{EDL}} / A_D \sim 0.01$; the large ratio $c_{\text{EDL}} / c_D \sim 10^7$ ensures that the voltage across the electrical double layer $V_{\text{EDL}}$ is well below 1 V when the voltage across the dielectric is on the order of $V_D \sim 10 \text{ kV}$. A small $V_{\text{EDL}}$ prevents electrochemical reaction, whereas a large $V_D$ enables electromechanical transduction.

2.3.2 Transparent Actuator

We built a transparent, high-speed, large-strain actuator. A membrane of a dielectric elastomer was sandwiched between two membranes of an electrolytic elastomer (Figure 2.1 and Figure 2.6). The dielectric and the electrolyte are stretchable and transparent, but the electrodes need not be. The electrode/electrolyte interfaces can be much smaller than the area of the dielectric, allowing us to place the electrodes outside the active region. Consequently, the active region of the actuator consisted of only electrolytic and dielectric elastomers and was highly stretchable and transparent. When a voltage was applied between the electrodes, ions of different charge polarities collected on the two faces of the dielectric; the oppositely charged interfaces attracted each other and caused the sandwich to reduce its thickness and enlarge its area (Figure 2.6B).

We demonstrated this design by using 1-mm-thick VHB 4910 tape (3M) as the dielectric and 100-µm-thick polyacrylamide hydrogel containing NaCl as the electrolyte. To compare the performance of the ionic conductor to an electronic conductor, we used the hydrogel to replace the carbon grease in the actuator used by Pelrine et al.. We stacked three layers of VHB together,
Figure 2.6 Transparent actuator capable of fast voltage-induced deformation. (A) A dielectric elastomer is sandwiched between two layers of an electrolytic elastomer. Both the dielectric and the electrolyte are transparent and stretchable, and the device is transparent if the electrodes are placed outside the active area of the device. (B) Subject to voltage, the two layers of the electrolyte spread ions of opposite signs on the two sides of the dielectric, causing the sandwich to reduce thickness and expand area. (C and D) The actuator is transparent to all colors. The area strain is measured as a function of voltage (E) and as a function of the frequency (F) by using an actuator with electrolytes of circular shape.
stretched them radially to three times their initial radius, and fixed them to a circular acrylic frame (Figure 2.1). We then sandwiched the dielectric stack between two layers of heart-shaped hydrogels, which are linked, through thin hydrogel lines, to copper electrodes placed on the frame. When a voltage is applied and removed, the heart expands and contracts. The beating heart is transparent to all colors (Figure 2.6C and D).

We also fabricated an actuator with the dielectric sandwiched between layers of the hydrogels of circular shape, and compared the experimental data with theory (Figure 2.2 and Appendix A). An area strain of up to 167 % was achieved (Figure 2.6E and F). This voltage-induced strain was limited by electromechanical instability (Figure A.1). Furthermore, the soft hydrogels did not appreciably constrain the dielectric. The area strain of our actuator reduced as the frequency of applied voltage increased and became vanishingly small at a frequency on the order of $10^3$ Hz (Figure 2.6F). These characteristics of the actuator using the hydrogel are comparable to those of an actuator using carbon grease (Figure 2.3), but the carbon grease is an opaque electronic conductor.

The frequency of actuation is not limited by electrical resistance but by mechanical inertia. The time to charge the device, the RC delay, scales as $\tau_{\text{RC}} \sim c_D A_D R$, where $R$ is the sheet resistance of the ionic conductor (see Appendix A for detailed calculation). For representative values $c_D = 10^{-8}$ F/m$^2$, $A_D = 10^{-2}$ m$^2$, and $R = 102$ ohm/square, we estimate that $\tau_{\text{RC}} \sim 10^{-8}$ s, corresponding to a frequency much higher than the observed limiting frequency of actuation. The fundamental resonance sets a time $\tau_{\text{inertia}} \sim (A_D \rho / Y)^{0.5}$, where $\rho$ is the mass density and $Y$ the elastic modulus (see Appendix A for detailed calculation). For representative values $\rho = 103$ kg/m$^3$ and $Y = 10^5$ N/m$^2$, we estimate that $\tau_{\text{inertia}} \sim 10^{-3}$ s; this value is consistent with the observed limiting frequency of actuation.
2.3.3 Transparent Loudspeaker

To demonstrate that the ionic conductors enable electromechanical transduction much beyond the fundamental resonance, we built a transparent loudspeaker that produces sound across the entire audible range, from 20 Hz to 20 kHz. The loudspeaker was placed in front of a laptop, which played music videos (Figure 2.4, Figure 2.7A and B). The screen of the laptop was visible through the loudspeaker. The sound tracks of the videos were fed to the loudspeaker as analog voltage signals, from the audio output of the laptop, through a high-voltage amplifier. A video of the transparent loudspeaker and the sound produced were recorded with a Web camera (webcam) at a distance of 15 cm.\(^{24}\)

We studied the fidelity of the sound reproduction by feeding the loudspeaker with a 20-s test signal of constant amplitude and a linear sine sweep from 20 Hz to 20 kHz (Figure 2.7C and D). The sound generated by the loudspeaker was recorded by the webcam.\(^{24}\) In the first few seconds, the amplitude of the recorded sound was large (Figure 2.7E). This interval reflects the resonance of the frame of the loudspeaker, which was not optimized to suppress vibration. The amplitude varied only slightly over the remainder of the recording, where variations in amplitude might be overshadowed by background noise from the high-voltage amplifier. The spectrogram of the recorded sound displayed the successful reproduction of the main signal of the original test sound throughout the audible frequency range (Figure 2.7F). In the lower part of the tested frequency range, vibrations of the frame and the membrane were visible.\(^{24}\)

Loudspeakers have been made by using electronic conductors, such as graphene sheets,\(^{84}\) carbon nanotubes,\(^{85}\) carbon grease,\(^{25}\) and conducting polymers.\(^{86}\) Some loudspeakers use piezoelectric polymers,\(^{84,85}\) and others use dielectric elastomers.\(^{25,86}\) Pairing the ionic conductors with transparent piezoelectric polymers will take advantage of the high transparency of the ionic
Figure 2.7 Transparent loudspeaker capable of producing sound across the entire audible range. (A) The voltage signal from the audio output of a laptop was fed through a high-voltage amplifier to the loudspeaker. The loudspeaker transformed the voltage signal into sound, which was recorded with a microphone. (B) The screen of the laptop was visible through the loudspeaker. Amplitude (C) and spectrogram (D) of a test signal with constant amplitude and a linear sine sweep of frequency from 20 Hz to 20 kHz. The colors correspond to the intensities of frequency, with warmer colors indicating higher intensity. Amplitude (E) and spectrogram (F) of the sound generated by the loudspeaker.
conductors; piezoelectric polymers can be operated with lower electric fields compared with dielectric elastomers and have a linear relation between electric field and strain.\textsuperscript{84,85}

2.3.4 Comparison of the Performance of Ionic and Electronic Conductors

The ionic conductors have higher resistivity than many electronic conductors; however, when high stretchability and transmittance are required, the ionic conductors have lower sheet resistance than existing electronic conductors, such as silver nanowires (AgNWs), single-wall carbon nanotubes (SWNTs), graphene, and indium tin oxide (ITO). The resistivity of the hydrogel is almost identical to that of water containing the same concentration of NaCl when the hydrogel is not stretched and increases when the hydrogel is stretched (Figure 2.8A). For a conductor whose resistivity is independent of deformation, the resistance of the conductor increases with stretch as \( R / R_0 = \lambda^2 \), where \( R_0 \) is the resistance before the conductor is stretched and \( R \) is the resistance after the conductor is stretched \( \lambda \) times its initial length. This prediction closely approximates the measured resistance-stretch curve for the ionic hydrogel (Figure 2.8B). When SWNTs on VHB are stretched, the resistance increases far faster than the prediction of the square law, indicating that the resistivity of the SWNTs is increased greatly by the stretch. An 11-mm-thick hydrogel containing 5.48 M NaCl shows 98.9 % average transmittance in the visible range (Figure 2.8C), corresponding to a transmittance of 99.99 % for a 100-mm-thick hydrogel used in constructing actuators and loudspeakers. Among all conductors of electricity, these hydrogels show the highest transmittance (Figure 2.8D).

2.4 Summary

The diversity of ionic conductors creates a large pool of candidates for applications. Hydrogels are easy to make and inexpensive, ideal for demonstrating conceptual designs and for fabricating devices that require biocompatibility. In higher organisms, it is ionic conductors that
Figure 2.8 Performance of ionic and electronic conductors. (A) Electrical resistivities of hydrogels of several concentrations of NaCl were measured as functions of stretch and compared with the resistivities of water containing the same concentrations of NaCl. Error bars show standard deviation; sample size N = 3. (B) Stretch was plotted against normalized resistance for ITO, AgNWs, graphene, SWNTs, and hydrogel (this work). (Inset) Details in the small stretch region. (C) An 11-mm-thick polyacrylamide hydrogel containing 5.48 M NaCl shows 98.9 % average transmittance in the visible range. (D) Transmittance (at 550 nm) is plotted against sheet resistance for ITO, AgNWs, SWNTs, graphene, and hydrogels (100-mm thickness, this work).
transmit in vivo information. This characteristic offers the potential to build nondamaging interfaces between ionic/biological and ionic/electronic signals. Although the loss of water from hydrogels can be an issue in some applications, the rate of evaporation may be reduced by encapsulation. Also, ionic liquids are nonvolatile electrolytes and can be used as conductors for large-strain, high-speed dielectric elastomer actuators (Figure A.2). Furthermore, it will be interesting to explore polyelectrolytes, in which charged polymers covalently cross-link into networks, and counterions are mobile. Hydrogels can be an issue in some applications, the rate of evaporation may be reduced by encapsulation. Also, ionic liquids are nonvolatile electrolytes and can be used as conductors for large-strain, high-speed dielectric elastomer actuators (Figure A.2). Furthermore, it will be interesting to explore polyelectrolytes, in which charged polymers covalently cross-link into networks, and counterions are mobile.

Replacing electronic conductors with ionic conductors raises further scientific questions. For example, various types of conductors may lead to different characteristics of charge transport through the dielectric. We have measured charge leakage through VHB when three types of conductors are used: carbon grease, a hydrogel, and an ionic liquid. The three types of conductors give a comparable time scale for leakage for the chosen experimental conditions (Figure A.3). As a second example, we used thin lines of ionic conductors, hydrogels and ionic liquids, to link active regions of devices to the copper electrodes. Our calculations indicate that ionic conductors can serve as high-speed, long-distance interconnects (Figure A.4). As a third example, the lifetime of a device depends on the electrolyte, the dielectric, and the electrode. However, finding ideal combinations of materials for long lifetimes is a large undertaking beyond the scope of this chapter.

Our design of layered electrolytic and dielectric elastomers differs from existing transducers in which ionic conduction plays essential roles, such as actuators made of carbon
nanotubes and conducting polymers in electrolytes,\textsuperscript{87,88} actuators made of ionic polymer–metal composites,\textsuperscript{6} and resistive strain sensors made of elastomeric ionic hydrogels.\textsuperscript{89} Our design introduces a dielectric in series with an electrolyte, and the small capacitance of the dielectric enables high-speed and large-strain actuation. Layered electrolytes and dielectrics also appear in one set of electrowetting devices, but the electrolytes are liquids, and the dielectrics do not deform.\textsuperscript{90}

Stretchable ionics offer new opportunities for designers of soft machines. Dielectric elastomers have been paired with electronic conductors to make transducers in robotics, bioelectronics, and energy harvesting.\textsuperscript{91} Pairing dielectric elastomers with ionic conductors will substantially expand the scope of applications, leading to devices such as tunable optics and localized haptics with transparent ionic conductors placed in the path of the light. Transparent loudspeakers might be attached to windows to achieve active noise cancellation.\textsuperscript{85} The ultrahigh transparency and compliance of the ionic conductors will enable transparent devices of multilayered electrolytic and dielectric elastomers. To scale up the active area of a device, one can enlarge the electrode/electrolyte interface by using porous electrodes, such as those used in supercapacitors. This paper focuses on the use of ionic conductors in devices operating at high speed and under high voltage, but the layered electrolytic and dielectric elastomer also works for applications that require low voltage or low frequency. When stretched mechanically, the layered material increases area and reduces thickness, so that its capacitance increases. This characteristic will enable transparent sensors operating at low voltage, capable of measuring strains over a large range, and conformal to soft tissues.
Chapter 3

A Transparent Membrane for Active Noise Cancellation*

3.1 Introduction

Long term exposure to loud noise—in urban life, or at work—is unpleasant, and can have detrimental health effects beyond damaging hearing.92 Passive sound insulating materials effectively attenuate noise at high frequencies, but become increasingly inefficient at lower frequencies.93,94 At frequencies below 500 Hz, active noise cancellation systems can be more cost-effective than passive sound insulating materials, and save space.93

Such an active noise cancellation system records incoming noise and generates sound that interferes destructively with that noise. Because destructive interference only cancels noise locally in space, active noise cancellation is difficult to implement in an entire three-dimensional space (e.g., a room inside a building).93,95 An alternative approach is to inhibit the transmission of noise into a room by exciting vibrations in walls and windows.95 The development of optically transparent, active noise cancellation windows has been a challenge.96,97

Here, we characterize the acoustic behavior of a transparent membrane for active noise cancellation. The membrane is of the same design as the transparent loudspeaker in Chapter 2. It consists of a layer of a dielectric elastomer, radially stretched and clamped between two rigid dielectric rings (Figure 3.1A). At the center, on both faces of the dielectric two layers of ionic conductors are attached, which are connected via lines of ionic conductors to metallic leads fixed on the opposite sides of the rings. Because we use transparent materials for the dielectric elastomer

* This chapter is based on unpublished work that was performed under my lead in collaboration with Xavier Morelle, Kun Jia, and Zhigang Suo. Supplementary information for this chapter is appended to this dissertation as Appendix B.
Figure 3.1 A transparent membrane for active noise cancellation. (A) The membrane consists of a dielectric membrane clamped between two rigid circular rings. Circular, ionically conducting electrodes are attached to the center of the elastomeric membrane on both faces. Thin ionic conductors connect the electrodes to metallic leads on opposite sides of the ring. An impinging acoustic sound wave ($F_R$) is partially reflected ($F_L$), and transmitted ($B_R$). Applying a time-dependent voltage signal ($V$) to the metallic conductors causes rearrangement of ions in the electrodes and leads to an electric field across the thickness of the elastomeric membrane. The resulting Maxwell stress causes oscillations of the membrane, which influence the transmitted, and reflected soundwaves. (B) A photograph of the transparent membrane made with an acrylic elastomer (VHB 4910 by 3M), frames of acrylic plastic, polyacrylamide hydrogels swollen with an aqueous solution of sodium chloride as ionic conductors, and conductive copper tape as metallic leads.
(VHB 4910, 3M) and the ionic conductors (a polyacrylamide hydrogel swollen with an aqueous solution of NaCl), the entire membrane is transparent (Figure 3.1B). This level of transparency is difficult to achieve with membranes sandwiched between electronic conductors.\textsuperscript{24,85} 

Upon application of a voltage $V$ to the metallic leads, ions inside the ionic conductor rearrange such that an electric field $E$ arises across the dielectric in the region of the overlapping ionic conductors (Figure 3.1A). The resulting Maxwell stress in the dielectric, 

$$
\sigma = \varepsilon E^2,
$$

where $\varepsilon$ is the dielectric constant of the elastomer, causes a reduction in thickness and an increase in area in the region of the dielectric, which is covered by the circular ionic conductors.\textsuperscript{14} In Chapter 2 we applied a sinusoidal voltage to the metallic leads and the membrane generated sound over the entire audible range (20 Hz to 20 kHz), but we did not characterize its acoustic properties in detail.\textsuperscript{24} In this chapter we characterize the transparent loudspeaker for frequencies $150 \text{ Hz} < f < 1000 \text{ Hz}$.

When an incident wave (amplitude $|F_R|$) reaches the membrane, it partially reflects (amplitude $|F_L|$), and partially transmits (amplitude $|B_R|$). The sound transmission loss (STL) across the membrane is defined by\textsuperscript{98}

$$
STL = 20 \log \left( \frac{|F_R|}{|B_R|} \right).
$$

The larger the $STL$, the less sound is transmitted through the membrane. A goal of this chapter is to investigate how the membrane must be excited to increase the $STL$.

We propose a linear model to characterize the reflection, transmission, and generation of sound by the membrane. We experimentally measure the acoustic properties of the membrane in the frequency range from 150 Hz to 1000 Hz inside an impedance tube, and use the linear model to calculate the theoretical limits of active noise cancellation with the membrane. We demonstrate
active noise cancellation with the membrane for an incident acoustic noise with frequencies over the range from 150 Hz to 1000 Hz at an amplitude of 80 dB in a feedforward control experiment. We calculate the necessary excitation signal for the membrane with the linear model. Without an active control signal (i.e., passive sound attenuation due to the presence of the membrane inside the impedance tube), we find that the STL of the membrane is on average 7 dB (with a peak of 19 dB) in the frequency range from 150 Hz to 1000 Hz. By applying an active control signal, we improve the STL in that frequency range to an average of 16 dB (with a peak of 37 dB).

In general, the vibration amplitude of the membrane is a nonlinear function of the excitation amplitude because of (i) the quadratic dependence of the Maxwell stress on the electric field (equation (3.1)), (ii) the nonlinear stress-strain relationship of the dielectric membrane, and (iii) the possible large deformations. The ability to use a linear model to describe sound generation, reflection, and transmission by the membrane for small excitation voltages simplifies its characterization and the control of a noise cancellation device based on this technology.

This chapter demonstrates the potential of the transparent membrane in applications of active noise cancellation. The high transparency of the membrane makes it an excellent candidate for applications in which transparency is important (e.g., reduction of sound transmission through a window). Because the transparent membrane is only ~ 2.2-mm thick, it will not significantly increase the volume of a window, and its planar design reduces unfavorable distortion of the view through it.

3.2 Background

Several attempts have been made to cancel noise through windows. Yu et al. used a thin film PVDF with transparent carbon-nanotube electrodes for active noise cancellation. The optical transmittance of the PVDF loudspeaker was 72 % - 90 % in the visible range, which can
be noticed with the eye. They actively reduced noise signals containing mixtures of single frequencies up to 15 dB, and they cancelled random noise by ~ 6 dB between 250 Hz and 800 Hz. Lane\textsuperscript{100} excited oscillations directly in the window glass of an anechoic room with four transducers located at the edges of the window glass (which were therefore visible). He actively reduced the transmission of sound energy into the room coming from a noise source with frequencies between 50 Hz and 1000 Hz in front of the window on average by ~ 3 dB. Kaiser \textit{et al.},\textsuperscript{97} and Jakob and Moser\textsuperscript{101} placed loudspeakers between two glass panes, which made their system bulkier than the above examples. They actively reduced the noise transmission close to the resonance frequency of the double pane window by up to 18 dB.\textsuperscript{97}

Even though dielectric elastomers have been used as loudspeakers,\textsuperscript{24–30} and proposed for active vibration suppression,\textsuperscript{102–104} studies of noise reduction with dielectric elastomers have so far only been concerned with the influence of static voltages on the passive sound attenuation performance. Lu \textit{et al.} investigated the sound absorption behavior of dielectric elastomer actuators attached to an air-filled cavity that acted as a resonator.\textsuperscript{105,106} Attached to the side of an impedance tube the elastomer-cavity system reduced the sound transmission along the tube by up to 43 dB, but only at the resonance frequencies of the system (The \textit{STL} decreased to below 5 dB within about 25 Hz from the resonance frequencies).\textsuperscript{106} By applying a constant voltage to the actuator Lu \textit{et al.} shifted the resonance frequencies and thus the absorption spectrum actively by up to 60 Hz.\textsuperscript{105,106} Broad band noise reduction could be achieved by combining multiple actuators with neighboring resonance frequencies.\textsuperscript{106}

Jia \textit{et al.} measured the \textit{STL} of a flat dielectric membrane installed at the center of an impedance tube at different static voltages.\textsuperscript{107} They observed equally spaced \textit{STL} peaks corresponding to the anti-resonance frequencies of the membrane \textit{(i.e.,} frequencies at which the
membrane does not oscillate), with a maximum of 15 dB at the first resonance frequency. The $STL$ decreased to below 5 dB within 25 Hz from these anti-resonance frequencies. By applying a static voltage to the actuator, they achieved frequency shifts of the absorption maxima by up to 65 Hz.

3. 3 Experimental Details

3. 3. 1 Fabrication of the Ionic Conductor

To synthesize the hydrogel, we used the following substances purchased from Sigma Aldrich: Acrylamide (AAm, monomer), ammonium persulfate (APS, initiator), $N,N,N’,N’$-tetramethylenediamine (TEMED, accelerator), $N,N’$-methylenebis(acrylamide) (MBAAm, crosslinker), and sodium chloride (NaCl). The polyacrylamide (PAAm) hydrogel was synthetized using the following protocol: We dissolved 47.3 g of AAm in 300 mL of de-ionized water to form a 2.22 M aqueous monomer solution, to which we added MBAAm, TEMED and APS in quantities of 0.00062, 0.00343 and 0.00183 times the weight of AAm. After degassing the prepared solution, we poured it into a 120 x 80 x 1 mm$^3$ acrylic mold and covered with a glass plate. We kept the sample at room temperature for 24 h to allow the polyacrylamide network to form, and then soaked it for at least 2 days in a 2.75 M NaCl solution.

We cut the ionic conductor with a laser cutter (VersaLaser VLS 3.5) into a paddle like shape (Figure 3.1) The diameter of the circular center was 51 mm, the width of the lead 7.5 mm, and the length of the lead 25 mm.

3. 3. 2 Fabrication of the Transparent Membrane

To fabricate the transparent membrane (Figure 3.1), we stuck two layers of VHB 4910 (3M) together, stretched them by hand equibiaxially to three times their initial dimension, and sandwiched them between two acrylic rings (inner diameter 4 in, outer diameter 6 in). On opposing
sides of the frame, we attached copper tapes (0.5-cm wide). On each face of the dielectric membrane we attached a hydrogel electrode, so that their circular regions overlapped in the center, and the thin ionic leads connected to the copper tapes. To improve adhesion between the hydrogel electrodes and the dielectric membrane, we dried them with a stream of N\textsubscript{2} before attaching them.

3.3.3 The Impedance Tube

We performed all experiments inside of a custom-made impedance tube (Figure 3.2). The impedance tube consisted of two 5 ft-long section of PVC tubing (inner diameter 4 in, outer diameter 5 in). We glued 1-in thick flanges to the ends of the tubes to attach the coil loudspeaker (GUI Inc., GF1004), the membrane and a termination (2-in thick polyurethane foam). The transparent membrane was positioned in the center of the impedance tube. We measured the sound waves at the centers of the front and the back sections of the impedance tube with a pair of microphones (Brüel & Kjaer, 4192-L-001) with preamplifiers (Listen Inc., SoundConnect), which were spaced 8 cm apart. We excited the transparent membrane through a high-voltage amplifier (Trek 30/20A), and the coil loudspeaker with a commercial audio-amplifier (AudioSource, AMP100VS). Signal generation and recording was done with Matlab through a DAQ-card (National Instruments, USB-6218).

We performed every experiment twice. In the first experiment, we recorded the acoustic pressure in the front section. For the second experiment, we moved the microphones to the back section of the tube. We covered the openings of the section without microphones with plugs. From the frequency components of the recorded signals (obtained by short time Fourier transformation) we calculated the amplitudes and phases of the acoustic waves in the two sections of the impedance tube (Appendix B.1).
Figure 3.2 Schematic of the experimental apparatus. (A) The transparent membrane is positioned in the center of an impedance tube, and excited through a high voltage amplifier with a sinusoidal voltage signal of bias $V_b$, amplitude $V_a$, and radial frequency $\omega$. A coil loudspeaker (excited with a sinusoidal voltage signal $V_L$ through an audio amplifier) on the left end of the impedance tube acts as a noise source, whereas a foam termination at the other end reduces reflections. Sound waves travel in the front and the back sections in positive ($F_R$, $B_R$), and negative ($F_L$, $B_L$) $x$-direction. A pair of microphones measures sound waves in both sections. The high voltage amplifier and the coil loudspeaker are controlled from a computer with Matlab through a data acquisition card, which also records the amplified signals from the microphones. (B) Photograph of impedance tube.
3. 4  Results and Discussion

3. 4. 1  Linear Model of the Acoustic Behavior of the Transparent Membrane

We studied the acoustic properties of the transparent membrane placed in the center of the impedance tube (Figure 3.2).\textsuperscript{98} The impedance tube simplifies the analysis of the experimental data, because acoustic waves travel as plane waves in the front section ($F_R, F_L$) and the back section ($B_R, B_L$) along the axis of the tube (\textit{i.e.}, only one spatial dimension has to be considered). To determine the sound waves travelling in a section we measured the sound pressure at two locations. The recorded signals can be decomposed into their frequency components with fast Fourier transformation (FFT), and with the known distance between the microphones we can calculate the frequency spectrum of the right and left travelling waves (Appendix B. 1).\textsuperscript{98}

We excited the membrane with a sinusoidal voltage signal of the form

$$V = V_b + V_a \text{e}^{i\omega t},$$

where $V_b$ is a constant bias voltage, $V_a$ the amplitude, and $\omega = 2\pi f$ the radial frequency of the signal. In this notation, $V_a$ is a complex number with the amplitude $|V_a| = (\Re(V_a)^2 + \Im(V_a)^2)^{0.5}$, and the phase angle $\varphi(V_a) = \arctan(\Im(V_a) / \Re(V_a))$. Due to the quadratic dependence of the Maxwell stress on the electric field (equation (3.1)), the response of the membrane to the applied voltage is nonlinear. However, when $V_b >> |V_a|$, the acoustic response of the loudspeaker to the applied voltage signal is linear, as we will demonstrate here.

When the transparent membrane is not excited, the sound waves in the front section are related to the sound waves in the back section with the transfer matrix $T_{ij}$.\textsuperscript{98} Under the assumption that the membrane responds linearly to small excitation amplitudes, we extended the concept of the transfer matrix by adding a term that is linearly proportional $V_a$:
\[
\begin{pmatrix}
F_R \\
F_L
\end{pmatrix} = 
\begin{pmatrix}
T_{11} & T_{12} \\
T_{21} & T_{22}
\end{pmatrix}
\begin{pmatrix}
B_R \\
B_L
\end{pmatrix} + 
\begin{pmatrix}
S_1 \\
S_2
\end{pmatrix} V_a
\] (3.4)

The components of the matrix \( T_{ij} \) (i, j = 1, or 2) are dimensionless and equivalent to the classical transfer matrix for passive sound attenuation (see Appendix B. 2 for details). The excitation column \( S_i \) describes the sound generation by the membrane. All variables in equation (3.4) are complex (i.e., they contain amplitude and phase information). The matrix \( T_{ij} \) has only two independent components, because the membrane is symmetric, and we assume reciprocity.\(^9\) These two requirements lead to \( T_{11}T_{22} - T_{12}T_{21} = 1 \) and \( T_{12} = - T_{21} \) (see Appendix B. 2 for details). The parameters \( T_{ij} \) and \( S_i \) are functions of the excitation frequency \( f \) and the applied bias voltage \( V_b \), but are independent of the lengths of the two segments and the terminations of the impedance tube.

We validated the linear model experimentally at a bias voltage of \( V_b = 9.0 \text{ kV} \) and sinusoidal excitation signals with a (randomly chosen) frequency \( f = 280 \text{ Hz} \). We applied the bias voltage for 120 seconds before the beginning of the experiment to reduce creep effects due to viscoelasticity of the membrane.\(^{10}\) We excited the coil loudspeaker alone with a linear amplitude sweep from \( V_L = 0 \text{ mV} \) to \( V_L = 150 \text{ mV} \) in 60 seconds. We then excited the membrane alone with a 60-seconds long linear amplitude sweep from \( V_a = 0 \text{ kV} \) to \( V_a = 1.5 \text{ kV} \). Then, we excited both loudspeakers simultaneously for 60 seconds with sinusoidal signals at constant amplitudes \((V_L = 75 \text{ mV}, V_a = 750 \text{ V})\), during which we shifted the phase of the excitation signal of the membrane with respect to the excitation signal of the membrane from \( \phi(V_a) = 0^\circ \) to \( \phi(V_a) = 360^\circ \) linearly over time.

When only the coil loudspeaker was excited, the amplitudes of the frequency component at \( f = 280 \text{ Hz} \) of the four waves (obtained by short-time Fourier transformation) increased linearly with the amplitude of the voltage \( V_L \) applied to the coil loudspeaker (Figure 3.3A). To obtain the
Figure 3.3 Comparison between theoretical model (circles), and experimental data (lines) at $f = 280$ Hz, and $V_b = 9.0$ kV. (A) Fundamental components of the spectra of the sound waves for linear amplitude sweeps on the coil loudspeaker. (B) Fundamental components of the spectra of the sound waves for linear amplitude sweeps on the transparent membrane. The decrease of amplitudes indicates the end of the linear regime. (C) Fundamental components of the spectra of the sound waves for a linear phase sweep on the transparent membrane at constant excitations amplitudes plotted as a function of the difference between the phase angles of $V_a$, and $F_R$. 
components of $T_{ij}$ we took $F_R$ and $B_L$ as experimentally prescribed, and fit equation (3.4) to the experimental data, which fits (with $T_{11} = 1.4 + 0.32i$, $T_{12} = -0.21 - 0.41i$, $T_{21} = 0.21 + 0.41i$, and $T_{22} = 0.76 - 0.31i$) the measured $F_{L1}$ and $B_R$ well (Figure 3.3A).

When we excited only the membrane, the amplitudes of the sound waves in both sections of the tube increased linearly for $V_a < 1.1$ kV (Figure 3.3B). At $V_a \approx 1.1$ kV, the amplitudes dropped between $\sim 0.05$ Pa and $\sim 0.1$ Pa. Before this drop, the linear model agrees well with the experimental data (with $T_{ij}$ as before, $S_1 = -0.64 + 0.38i$ Pa/kV, and $S_2 = -0.61 + 0.29i$ Pa/kV). Additionally, a subharmonic appeared in the measured acoustic spectrum at this excitation voltage (Figure 3.4). We therefore consider the appearance of the subharmonic as an indication of the end of the frequency regime in which the linear model is valid.

When both, the coil loudspeaker and the membrane were excited, the amplitudes of the acoustic waves changed with the phase difference between $V_a$ and $F_R$ (Figure 3.3C). Whereas $|F_L| / |F_R| \sim 0.5$, in the first part of the experiment (Figure 3.3A), the ratio varied now between $0.79 < |F_R| / |F_L| < 94$. For $|F_R| / |B_R|$ we observed similar (although not as large) changes. With the previously used values for $T_{ij}$, and $S_i$, equation (3.4) predicted the transmission and reflection at the membrane very well. These results demonstrate that by exciting the membrane with an appropriate amplitude, and phase angle—which can be calculated with equation (3.4)—one can influence sound reflection and transmission.

3. 4. 2 Characterization of the Acoustic Properties of the Transparent Membrane

We characterized the acoustic properties of the membrane (in terms of equation (3.4)) in the frequency range $150 \text{ Hz} < f < 1000 \text{ Hz}$ for the bias voltage $V_b = 9.0$ kV (which we applied for 120 seconds before the experiment). Even though the membrane can, in principle, generate sound at frequencies below 150 Hz, we were limited by the cut-off frequency of the coil loudspeaker.
Figure 3.4 Fundamental (solid line) and subharmonic (dotted line) component of the acoustic spectrum of $F_L$ as a function of $|V_a|$ at $V_b = 9.0 \text{kV}$, and excitation frequency $f = 280 \text{Hz}$. At $|V_a| \sim 1.1 \text{kV}$ a subharmonic appears in the acoustic spectrum, and the amplitude of the fundamental component drops.
We first applied a sinusoidal frequency sweep from 150 Hz to 1000 Hz with a sweep rate 40 Hz/s, and amplitude of 38 mV to the coil loudspeaker. Then we excited the membrane with a sinusoidal sweep from 150 Hz to 1000 Hz with a sweep rate 40 Hz/s, and amplitude $V_a = 645 \text{ V}$ (at this amplitude we did not observe a subharmonic in the acoustic spectrum, Figure B.2). From the travelling waves inside the front and back sections of the tube (Figure B.3), we determined with equation (3.4) the components of the matrix $T_{ij}$ (Figure 3.5A), and the column $S_i$ (Figure 3.5B) for all frequencies.

Because of resonances that depended on the length and the terminations of the impedance tube, it was difficult to judge the sound emission properties of the membrane from the raw data (Figure B.3). With $F_R = B_L = 0$ in equation (3.4), we obtained equations for the acoustic waves $\hat{F}_L$, and $\hat{B}_R$ that the membrane would emit in an impedance tube with anechoic terminations on both ends:

$$\hat{F}_L = \left( \frac{T_{12}S_1}{T_{11}} + S_2 \right) V_a$$

$$\hat{B}_R = - \frac{S_1}{T_{11}V_a}$$

The calculated values for $\hat{F}_L$, and $\hat{B}_R$ ranged between 69 dB and 96 dB (Figure 3.5C) (for comparison, the noise of a street traffic at 100 ft distance is ~ 70 dB, of a subway at 6 ft distance is ~ 90 dB)$^{109}$. This variation in amplitude is large compared to commercial loudspeakers (e.g., the manufacturer of the coil loudspeaker used in this work reports that it lies within 10 dB in the frequency range 145 Hz – 17 kHz). Reducing the variation in amplitude would require engineering of the membrane, and the frame. For all frequencies the amplitudes of $\hat{F}_L$ and $\hat{B}_R$ were approximately equal. This result demonstrates that the membrane emits sound equally in both directions. This conclusion is expected from the symmetric construction of the membrane, but
Figure 3.5 Characterization of the acoustic properties of the transparent membrane at bias Voltage $V_b = 9.0 \text{ kV}$ for $150 \text{ Hz} < f < 1000 \text{ Hz}$. (A) Real and complex components of $T_{11}$, and $T_{12}$. $T_{21}$, and $T_{22}$ are dependent on $T_{11}$, and $T_{12}$ and not shown for clarity. (B) Real, and complex components of $S_1$, and $S_2$. (C) Calculated amplitudes of the sound waves $\hat{F}_L$, and $\hat{B}_R$ that the membrane would emit in an impedance tube with anechoic terminations are equal because of the symmetry of the loudspeaker. The amplitude of the incident sound wave $\bar{F}_R$ that would be completely canceled by the membrane for $|V_a| = 645 \text{ V}$. (D) Because sound is generated by out of plane deformations of the membrane the phase difference between $\hat{F}_L$, and $\hat{B}_R$ is $\approx 180^\circ$. 
cannot be deduced from the raw data because the terminations of the tube were asymmetric (one end was terminated by the coil loudspeaker, and the other end by the foam).

The Maxwell stress (equation (3.1)) causes the reduction in the thickness and the expansion in the area of the membrane (the in-plane mode), but the membrane can also oscillate—like a drumhead—out of plane. In our previous work we were not able to determine which mode of oscillation is predominantly responsible for sound generation. The in-plane mode generates sound waves \( \vec{F}_L \), and \( \vec{B}_R \) in phase (Appendix B. 4). In the case of the out-of-plane mode, the phase difference would be 180°. The calculated phase difference of \( \vec{F}_L \), and \( \vec{B}_R \) is close to 180° (Figure 3.5D), which leads to the conclusion that the out-of-plane oscillation of the membrane is dominant for sound generation.

By setting \( B_R = B_L = 0 \) in equation (3.4), we calculated the maximum amplitude of the acoustic noise that the membrane could (in an ideal case) completely cancel actively:

\[
\vec{F}_R = S_1 V_a
\]  

(3.7)

For an upper limit of the excitation voltage of \( V_a = 645 \) V the amplitude varied from \( |\vec{F}_R| = 76 \) dB (~ sound of an automobile at 20 ft distance)\(^{109}\) to 107 dB (~ sound of motorcycle at 20 ft distance)\(^{109}\) (Figure 3.5C).

### 3.4.3 Passive Attenuation and Active Cancellation of Noise with the Transparent Membrane

By setting \( B_L = V_a = 0 \) in equation (3.4) we obtained an equation for the sound transmission loss due to passive attenuation \( STL_p^a \) by the membrane for an impedance tube with anechoic termination:\(^{98}\)

\[
STL_p^a = 20 \log(|T_{11}|)
\]

(3.8)
We observed strong resonance peaks in $STL_p^a$ for frequencies between 150 Hz and 250 Hz with amplitudes up to 19 dB (Figure 3.6). $STL_p^a$ was 3 dB – 12 dB without clear resonance peaks in the range of frequencies from 250 Hz to 1000 Hz. Jia et al. reported the same behavior for a dielectric elastomer actuator with electrodes made of carbon grease.$^{107}$

To illustrate active noise cancellation (at $V_b = 9.0$ kV) based on the linear model, we actively cancelled an acoustic noise wave that consisted of a sinusoidal frequency sweep from 150 Hz to 1000 Hz and amplitude $|F_R| \sim 80$ dB (sound of light trucks at 20 ft distance)$^{109}$ with feedforward control. We calculated the necessary excitation signal (Figure B.4) with equation (3.8), using the previously determined value for $S_1$ (Figure 3.5B). As mentioned above, the incident sound wave $F_R$ is composed of the sound wave generated by the coil loudspeaker and of the reflection of $F_L$ at the coil loudspeaker, and was therefore also influenced by the excitation signal of the membrane. Exploiting linearity of the coil loudspeaker, we adjusted its excitation voltage during the sweep to keep the amplitude and phase of $F_R$ approximately constant (Figure B.4).

Analogously to equation (3.8), we derived an equation, which corrects the sound transmission loss for resonance effects of the tube by subtracting the influence of the reflected wave $B_L$:

$$STL_{a}^a = 20 \log \left( \frac{|F_R|}{|B_L|} \right) = 20 \log \left( \frac{|F_R|}{|B_R + \frac{T_{12}}{T_{11}} B_L|} \right)$$  \hspace{1cm} (3.9)

We used the recorded pressure waves (Figure B.5), the previously determined values of $T_{ij}$ (Figure 3.5A), and equation (3.9) to calculate the sound transmission loss $STL_a^a$ for an anechoic tube during active noise cancellation (Figure 3.6) (see Figure B.6 for a comparison of $STL_a^a$ to the uncorrected sound transmission loss based on equation (3.2)). Except for the region close to
Figure 3.6 Comparison of sound transmission loss of passive sound attenuation and active noise cancellation for an incident sound wave $|F_R| = 80$ dB in an impedance tube with anechoic termination calculated using the linear model and the experimental results. Except for close to 200 Hz, the active sound cancellation outperforms passive sound attenuation.
200 Hz, active noise cancellation outperformed passive attenuation. The sound transmission loss increased from an average of 7 dB with a peak of 19 dB for passive sound attenuation, to an average of 16 dB with a peak of 37 dB for active noise cancellation.

### 3.5 Summary

This chapter establishes that sound transmission and reflection at the membrane can be actively controlled. Its linear response to sinusoidal excitation voltages at small amplitudes allows relatively simple control. With simple active feedforward control based on the linear model developed here we could improve the average STL through the membrane compared to passive sound attenuation by \( \sim 100\% \) \((i.e.,\ by\ a\ factor\ of\ three\ in\ amplitude)\).

Compared to the work by Yu et al., the transparent membrane shown in this work has higher optical transmittance \( (> 99\%,\ as\ opposed\ to\ 72\%\ -\ 90\%)\),\textsuperscript{24,85,96} which makes it better for applications in which transmittance is of importance. The improvement of the sound transmission loss obtained with active control in this chapter exceeds the one reported by Lane.\textsuperscript{100} It is comparable to the one achieved by Kaiser et al.,\textsuperscript{97} but the membrane used here requires less space.

Testing the acoustic behavior of the membrane in an impedance tube simplified the sound field to plane waves. Further work is required to test the performance of the membrane under more realistic conditions. This work would include not only more complex acoustic waves \( (e.g.,\ non\)-planar acoustic field, moving source of noise), but also interactions of the membrane with close structures \( (e.g.,\ the\ influence\ of\ the\ interspace,\ when\ attached\ to\ a\ window)\).

The membrane provides a new method for active noise cancellation for applications in which space is limited and high optical transparency is required. Examples include windows, wall panels, and protective gear in loud workplaces. The design of the membrane is not limited to a
planar, circular shape with a rigid frame. Non-planar, deformable membranes may enable unusual, adaptable acoustic properties, such as directionality of the emitted sound.
Chapter 4

Durability of Dielectric Elastomer Transducers Made with Ionic Liquids*

4.1 Introduction

In this chapter, we investigate the lifetime of dielectric elastomer transducers, under electromechanical excitation when ionic liquids are used as conductors. The lifetime of transducers is important for their commercial application, because the devices are expected to last for years, and to be able to sustain millions of actuation cycles. In Chapter 2, and Chapter 3 we used hydrogels swollen with an aqueous solution of NaCl, as ionic conductors. Because ionic conductors require mobile ions, the devices lose function when the water evaporates, and the hydrogels dry out. Hygroscopic salts (e.g., LiCl) can prevent complete evaporation of water, but the water content in the hydrogel varies with the relative humidity of the environment. Coating the hydrogels with butyl rubber would reduce changes in the water content due to fluctuation in relative humidity, but would also constrain the actuation of the transducers mechanically. To circumvent this issue, we used room temperature ionic liquids as conductors.

Room temperature ionic liquids are salts that are liquid at room temperature. They are, due to their low (often unmeasurable) vapor pressures, considered to be nonvolatile, and can be used as solid state conductors in ionic gels. Ionic liquids are a large class of material (theoretically, more than a million different combinations of anions and cations exist that are liquid at room temperature), which allowed us to test different combinations of ions.

* This chapter is based on unpublished work that was performed under my lead in collaboration with Christoph Keplinger, and Zhigang Suo.
In this study, we fabricated the dielectric membrane from the acrylic elastomer VHB 4910 (3M)—one of the most commonly used materials for dielectric elastomer transducers—and used 20 different ionic liquids as ionic conductors. We performed a compatibility test to identify which ionic liquids swell the elastomer. To investigate the difference in behavior, when swelling, or non-swelling ionic liquids are used, we measured the change in capacitance over time of a dielectric elastomer transducer made with a non-swelling ionic liquid (1-Ethyl-3-methylimidazolium tetrafluoroborate, [EMIM][BF$_4$]), and with a swelling ionic liquid (1-Decyl-3-methylimidazolium chloride, [DMIM][Cl]). The capacitance of the transducer made with [EMIM][BF$_4$] increased with time but reached an equilibrium after ~50 h (~40 % above the initial value). A one-dimensional diffusion model agreed well with the measured results. The capacitance of the transducer made with [DMIM][Cl] increased 20-fold, until the dielectric membrane became ion-conducting after 9 h.

Under repeated electromechanical excitation, the transducers failed by electric breakdown. We tested the influence of the operating conditions on the lifetime of the transducers by varying the amplitude, and the frequency of the excitation signal. The number of actuation cycles before failure decreased exponentially with the applied voltage, and increased linearly with the applied oscillation frequency. We compared the increase in capacitance with time, as well as the lifetime under electromechanical actuation, for transducers made with 10 different ionic liquids. The lifetime decreased exponentially with the increase in capacitance, and varied by three orders of magnitude.

4.2 Background

The failure of dielectric elastomer transducers as a result of rupture, electrical breakdown, and electromechanical instability are well understood.$^{83,115–117}$ Even though the ability to withstand
repeated actuation cycles is important for practical applications, the lifetime of dielectric elastomer transducers is the focus of only a handful of studies, and a standardized test method does not exist.\textsuperscript{118–121} These studies focus on dielectric elastomer transducers made with electronic conductors. When excited with an AC-voltage, which is large enough for electromechanical transduction, the transducers usually fail after a certain number of cycles by sudden electric breakdown (one study reported mechanical failure of the connection between the electrodes and the conductor).\textsuperscript{118–120} Increasing the relative humidity in the environment also reduces the number of cycles to failure.\textsuperscript{118,119} This effect is stronger for polar elastomers (\textit{e.g.}, acrylic elastomers) than for non-polar elastomers (\textit{e.g.}, silicone elastomers).\textsuperscript{119} Stoyanov \textit{et al.} developed a carbon nanotube-based electrode that burns at the location of an electric breakdown event, insulating the damaged region, thus allowing continued use of the transducer even after multiple breakdown events.\textsuperscript{120}

Unlike electronic conductors, which conduct electricity using electrons, ionic conductors conduct through the movement of mobile ions.\textsuperscript{24} These ions, or their solvent (water in the case of hydrogels) may diffuse into the dielectric membrane, and affect the durability of the transducers.\textsuperscript{111,118,119,122,123} The electric field across the dielectric membrane during electromechanical actuation (~10 MV/m) may act as an additional driving force for diffusion.\textsuperscript{124,125} Water trees—tree-shaped networks of defects caused by local breakdown events, and subsequent diffusion of water into the defect—may grow inside the dielectric membrane under alternating electric fields, and ultimately lead to failure, as has been observed in high voltage insulations.\textsuperscript{126–128}
4. 3  Experimental Details

4. 3. 1 Materials

As the dielectric material we used VHB 4910 (3M), and commercially available ionic liquids (Table 4.1). Unless stated otherwise, we fabricated ionic conductors from an ionic grease made by stirring a powder of PVDF (average molecular weight 534’000, Aldrich 182702) into the ionic liquids (3:5 by weight) until we obtained a homogenous grease. As electronic conductor, we used conductive copper tapes. We also fabricated one transducer with pure [DMIM][Cl] as the conductor (this ionic liquid was viscous enough not to flow when painted onto the dielectric membrane).

4. 3. 2 Fabrication of the Dielectric Elastomer Transducers

We stretched the VHB 4910 membranes radially to three times their initial dimensions, and attached them to rigid acrylic frames with 8 cm inner diameter and 10 cm outer diameter. On opposite sides of the frame we attached conductive copper tapes as the interface to the external electronics. For the active region of the transducer, we painted a circle of 2 cm diameter with the ionic grease, in the center of the membrane, on both sides. We connected each circle to one of the copper tapes with painted lines of ionic grease. Unless stated otherwise, we stored all actuators for 12 hours before use at the same relative humidity that we used during the experiment (100 %, or < 15 % relative humidity).

4. 3. 3 Measurement of the Capacitance of the Dielectric Elastomer Transducers

We measured the capacitance of the dielectric elastomer transducers with a LCR-meter (PK Precision, Model 885). The measurements of the changes of the capacitance with time were performed at ambient humidity (40 % - 50 % relative humidity).
Table 4.1 Ionic liquids used in this chapter

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<th>Nr.</th>
<th>Abbreviation</th>
<th>Name</th>
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<tr>
<td>1</td>
<td>[THTDP][DCA]</td>
<td>Trihexyltetradecylphosphonium dicyanamide</td>
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<td>[EMIM][BF₄]</td>
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<td>TCI</td>
<td>E0496</td>
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<td>3</td>
<td>[MMIM][DMP]</td>
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<td>00796</td>
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<td>89744</td>
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<td>TCI H1099</td>
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<td>Alrich 51682</td>
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<td>Aldrich 00797</td>
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<td>Aldrich 690597</td>
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<td>1-Decyl-3-methylimidazolium tetrafluoroborate</td>
<td>Aldrich 690708</td>
</tr>
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<td>1-Ethyl-3-methylimidazolium methyl sulfate</td>
<td>Aldrich 18086</td>
</tr>
<tr>
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<td>1-Butyl-3-methylimidazolium tetrafluoroborate</td>
<td>Aldrich 91508</td>
</tr>
</tbody>
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4. 3. 4 Measurement of the Actuation Strain

We recorded the deformation of the dielectric transducer under electromechanical actuation with a high-speed camera (Vision Research, Phantom v310). We extracted the frames where the active area was the smallest, and the largest, during one actuation cycle using MATLAB. From these frames we measured the size of the active area with ImageJ. We define the actuation strain $\varepsilon_A$ as the ratio between the change in size of the active area, and the minimum size of the area, during one cycle.

4. 3. 5 Lifetime Test of the Dielectric Elastomer Transducers under Electromechanical Actuation

We performed lifetime tests at 100 % humidity in a sealed fish-tank. A humidifier was used to keep the humidity constant. We excited the seven actuators, in parallel, with a square wave signal with a high voltage amplifier (Trek, 50/12). We generated the excitation signal with a function generator (Keithley 3390), and recorded it using MATLAB, through a DAQ-Card (National Instruments, USB-6218). We ran the experiments until all samples failed, alternating between 12 hours of excitation, and 12 hours without any excitation. Then we stopped the experiment for 12 hours and restarted the experiment. To determine the influence of relative humidity on the number of actuation cycles, we performed one test while continuously flushing the fish-tank with dry air (Airgas, AI UZ300). During this experiment, the relative humidity remained below 15 %.

4. 4 Results and Discussion

4. 4. 1 Compatibility Test

Ionic liquids can swell elastomeric membranes. To identify these ionic liquids, we performed a compatibility test, in which we deposited droplets (0.5 - 1 ml) of 20 different ionic
Figure 4.1 Photographs of a membrane of VHB 4910 on a gray background. (A) Drops of 20 different ionic liquids (Table 4.1) are deposited onto the membrane. The numbers correspond to the numbers of Table 4.1. The scalebar corresponds to 1 cm. (B) Photograph of the cleaned membrane after removal of the drops after 20 hours. The ionic liquids with the numbers 1, 5, 7, 11, 16, and 17 (Table 4.1) swelled the membrane, resulting in visible thickening of the membrane at the location of the drops.
liquids (Table 4.1) onto a membrane of VHB 4910 (Figure 4.1A). After 20 hours we gently wiped the membrane with a paper towel to remove the ionic liquids. We observed clear signs of swelling (thickening of the membrane at the location of the drops, Figure 4.1B) for six ionic liquids ([THTDP][DCA], [THTDP][Cl], [THTDP][TMPP], [THTDP][Br], [N(1)888][NTF2], [DMIM][Cl]). Other ionic liquids may also have diffused into the membrane, but at concentrations too small to visibly change the surface of the membrane.

Anions, and cations must diffuse in pairs to preserve charge neutrality (i.e., if only one ion diffuses into the membrane, a potential difference would develop along the thickness of the membrane). A simple consequence of this principle could be, that if an ion only diffuses in low concentrations into the membrane, all ionic liquids containing this ion can only diffuse in low concentrations (or not at all, if one of the ions does not diffuse into the membrane). The compatibility test showed that this is not the case. The ionic liquid [THTDP][NTF2] did not swell the elastomeric membrane during the test (i.e., at least one of these two ions diffuses only at low concentrations), while [THTDP][DCA], and [N(1)888][NTF2] did swell the membrane (i.e., both ions of [THTDP][NTF2] diffused at high concentrations).

4. 4. 2 Change of Capacitance with Time

The compatibility test performed in the previous section provides qualitative information about the compatibility of an ionic liquid with the dielectric membrane (i.e., swelling of the membrane), but does not give quantitative information about diffusion. When ions diffuse into the dielectric membrane of a dielectric elastomer transducer, its permittivity, and consequently its capacitance ($C$), changes over time. We used this effect study to the difference in behavior of an ionic liquid that does not swell ([EMIM][BF4]), and an ionic liquid that does swell ([DMIM][Cl]) VHB 4910.
We fabricated two transducers using VHB 4910, stretched three times biaxially, as the
dielectric (Figure 4.2A). For one transducer, we used [EMIM][BF₄] mixed (5:3 by weight) with a
powder of polyvinylidene fluoride (PVDF) to increase its viscosity. For the other transducer we
used pure [DMIM][Cl], because it was viscous enough not to flow. We measured the capacitance
of the devices at ambient relative humidity (40 % - 50 %) as a function of time, starting within
~ 1 min after painting the active area.

The capacitance of the device with [EMIM][BF₄] as the ionic conductor increased with
time, until it reached a plateau after ~ 50 h at 42 % above its initial capacitance (Figure 4.2B). We
stopped the measurement after 72 hours, because C had not changed significantly for 22 hours.
The experiment demonstrates, that even though this ionic liquid does not swell the dielectric
membrane, the ions diffuse into it. The device with [DMIM][Cl] as the ionic conductor showed an
entirely different behavior (Figure 4.2C). Instead of reaching a plateau, the capacitance continued
to increase, at a rate which accelerated over time, and the dielectric membrane became ion-
conductive (Resistance 2 MΩ) after nine hours. At this point, electrochemical reactions could
occur at the copper-ionic liquid interface, and the transducer loses its functionality, so the
experiment was stopped.

To explain the experimental results, we developed a model based on one-dimensional
diffusion. This model relates the change in capacitance to diffusion of ions into the dielectric
membrane. Initially, there is no ionic liquid in the dielectric membrane (Figure 4.2D). With time,
ions diffuse from the surface into the membrane leading to a time-, and location-dependent
concentration distribution \( \hat{c}(t, x) \), where \( t \) is the time, and \( x \) is the location measured from the
surface of the membrane. After a certain time, the entire elastomer is saturated with the ionic liquid
\( (\hat{c}(t \to \infty, x) = \hat{c}_s) \). The diffusion process can be described by a linear boundary-value problem.\(^{130}\)
Figure 4.2 Change in capacitance of dielectric elastomer transducer with time (A) Photograph of a dielectric elastomer transducer made with a grease of PVDF, and an ionic liquid as conductor. The scalebar corresponds to 2 cm. (B) Change in capacitance as a function of time for a dielectric elastomer transducer made with [EMIM][BF₄] (squares), and fit of the theoretical model to the data (solid line). (C) Change in capacitance as a function of time for a dielectric elastomer transducer made with [DMIM][Cl] (squares), and fit of the theoretical model to the data (solid line). (D) Diffusion of ions changes the local dielectric constant of the dielectric membrane.
\[
\frac{\partial \hat{c}}{\partial t} = D \frac{\partial^2 \hat{c}}{\partial x^2} \quad (4.1)
\]

Here, \(D\) is the diffusivity of the ionic liquid inside the elastomer. Due to symmetry we only consider half of the membrane. The initial condition is given by

\[
\hat{c} = 0, \quad t = 0, \ 0 \leq x \leq H/2. \quad (4.2)
\]

where \(H\) is the thickness of the dielectric membrane. The ionic conductor acts as an infinite source of ions, so that the elastomer is always saturated at the surface. Because of the symmetry of the problem, no flux occurs across the center of the membrane. This provides the boundary conditions:

\[
\hat{c} = \hat{c}_s \quad t > 0, \ x = 0
\]

\[
\frac{\partial \hat{c}}{\partial x} = 0 \quad t > 0, \ x = H/2 \quad (4.3)
\]

The solution of equation (4.1) with the initial, and boundary conditions (4.2)-(4.3) is

\[
\frac{\hat{c}(x, t)}{\hat{c}_s} = 1 - \frac{2}{\pi} \sum_{n=0}^{\infty} (-1)^n \frac{\pi(n+1/2)}{H} e^{-\pi^2(n+1/2)^2 4Dt/H^2} \cos \left( \pi \left(2n + \frac{1}{2}\right) \frac{x}{H} \right). \quad (4.4)
\]

We approximate the dependence of the local dielectric constant \(\varepsilon\) on \(\hat{c}\) with the linear relationship

\[
\varepsilon = \varepsilon_D + \frac{\hat{c}}{\hat{c}_s} (\varepsilon_s - \varepsilon_D), \quad (4.5)
\]

where \(\varepsilon_D\) is the dielectric constant of the pure elastomer, and \(\varepsilon_s\) is the dielectric constant, when the elastomer is saturated with the ionic liquid.

The dielectric elastomer transducer can be described as a series of infinitesimally thin capacitors with a dielectric of permittivity \(\varepsilon\), and area \(A\) (the size of the active area). Its total capacitance, \(C\), can thus be calculated by

\[
\frac{1}{C(t)} = \frac{2}{A} \int_0^{H/2} \frac{1}{\varepsilon_D + \frac{\hat{c}}{\hat{c}_s} (\varepsilon_s - \varepsilon_D)} \, dx. \quad (4.6)
\]
Using the initial value of the capacitance \( C_0 \) and the value when the dielectric membrane is saturated \( C_s \), we rewrite equation (4.6) to express \( C(t) \) as

\[
\frac{C(t)}{C_0} = \frac{H}{2} \left( \int_0^{H/2} \frac{1}{1 + \frac{\dot{c}}{C_s} \left( \frac{C_s}{C_0} - 1 \right)} \, dx \right)^{-1}
\]

The integral in equation (4.7) can be solved in combination with equation (4.4), numerically.

Using the thickness of the dielectric membrane \( H = 100 \mu\text{m} \), and the change in capacitance in the plateau region \( C_s / C_0 = 1.42 \), we fit the theoretical model to the experimental data obtained with [EMIM][BF₄] (Figure 4.2B). With \( D = 2.1 \cdot 10^{-14} \text{ m}^2/\text{s} \) the theoretical model agrees remarkably well with the experimental data. The theoretical model is likely not accurate for the dielectric elastomer transducer made with [DMIM][Cl], because it does not consider changes in thickness due to swelling, or that the swollen elastomer can become conductive. With large ratios of \( C_s / C_0 \), the model can still be brought into good agreement with the experimental data. For example, with \( C_s / C_0 = 10^4 \), and \( D = 1.9 \cdot 10^{-15} \text{ m}^2/\text{s} \), the model fits the data well (Figure 4.2C). A physical interpretation of these values, however, is difficult. An estimation of the diffusivity based on the time in which the dielectric membrane becomes conductive \( t \approx 9 \text{ h} \), and using the scaling formula \( D = H^2 / t \), results in a value of \( D \approx 10^{-13} \text{ m}^2/\text{s} \), which is two orders of magnitude smaller than that predicted by the fit of the model to the experimental data.

**4.4.3 Actuation Cycles at Failure under Electromechanical Actuation**

We studied the dependence of the lifetime (the number of actuation cycles until failure, \( N \)) of the dielectric elastomer transducers under electromechanical excitation on the operating conditions. We used VHB 4910 as the dielectric, stretched biaxially by a factor of three, and a mixture of the ionic liquid [EMIM][EtSO₄] and PVDF (5:3 by weight) as the ionic conductor. We used a square-wave function as the excitation signal (Figure 4.3A) with a 50 % duty cycle \( (\text{i.e.}, \) the...
Figure 4.3 Influence of the operating conditions on the lifetime. (A) Schematic of the experimental apparatus. Seven samples are tested in parallel in a controlled environment of 100% relative humidity. The samples are excited with a square wave signal. (B) The number of actuation cycles at failure, $N$, and the area strain, $\varepsilon_A$, as a function of $f$ at $V_a = 3.5$ kV. The solid line corresponds to 8.3 h activated voltage. (C) The number of actuation cycles at failure, $N$, and area strain, $\varepsilon_A$, as function of $V_a$ at $f = 10$ Hz. The solid line is an exponential fit to the data. The errorbars in (B), and (C) show the standard deviation of the mean for 7 samples.
duration of voltage “on” and voltage “off” during one cycle was equal). We systematically varied the frequency $f_f$ and the excitation amplitude $V_a$. At each operating condition we also measured the area strain $\varepsilon_A$ (defined as the change in size of the active area divided by its minimum size during one cycle) of one transducer with a high-speed camera.

Previous work has shown that the lifetime of dielectric elastomer transducers made with VHB 4910 are sensitive to the relative humidity of the environment.\textsuperscript{118,119} Larger humidity leads to earlier failure. To reduce the duration of the experiments, we placed the samples for the test in a sealed box at 100 % relative humidity (Figure 4.3A). We performed each test with seven samples. All samples failed by electric breakdown, which triggered the current limiter of the high voltage amplifier, and interrupted the experiment. After removing the failed sample, we continued the experiment with the remaining samples until all samples failed. From the recorded excitation signal we determined $N$ for each transducer.

We studied the effect of the excitation signal on the lifetime by varying the frequency from 1 Hz to 20 Hz, at a constant amplitude of $V_a = 3.5$ kV. The average number of actuation cycles at failure increased with increasing frequency from $4.7 \cdot 10^4$ to $1.3 \cdot 10^6$, and the actuation strain decreased from 6.7 % to 1.1 % (Figure 4.3B). We then varied the amplitude of the excitation signal from 3.5 kV to 6.5 kV, at a constant frequency of $f_f = 10$ Hz. In these experiments, the number of cycles at failure decreased with increasing amplitude from $6.9 \cdot 10^5$ to $1.5 \cdot 10^3$ (Figure 4.3C), whereas the actuation strain increased from $\varepsilon_A = 1.7$ % to $\varepsilon_A = 16$ % (Figure 4.3C).

Given that certain polymers have shown a frequency dependent fatigue behavior at constant stretch amplitude, the dependence of the lifetime of the actuators on the excitation signal could be explained by mechanical fatigue of the dielectric membrane.\textsuperscript{131–133} We show below that the lifetime varies by three orders of magnitude for different ionic liquids at the same operating
conditions, for which reason we exclude mechanical fatigue of the membrane as the dominant reason for the fatigue behavior. We can also exclude the growth of water trees as a reason for failure, because experimental studies on high-voltage insulation has demonstrated that the growth rate of water trees increases with the frequency (i.e., increasing $f$ would lead to a decrease in $N$), which contradicts our experimental results.\textsuperscript{126–128}

When $V_a$ was constant, $N$ increased approximately linearly with $f$. The time of activated voltage at failure ($t_A \approx 8.3$ h) was therefore approximately constant for all frequencies (solid line in Figure 4.3B). This result indicates that cycling of the voltage is not the cause of failure, but rather the duration of the application of the voltage. At constant $f$, $N$ decreased approximately exponentially with $V_a$ (solid line in Figure 4.3C). As we demonstrated in section 4.4.2, ions diffuse into the dielectric membrane. An applied electric field imposes an additional driving force that increases diffusion.\textsuperscript{125} We hypothesize that failure occurred when the ions had diffused far enough or at high enough concentration into the membrane to cause electric breakdown. At large electric fields (here the electric field was $\sim 10^7$ V/m), the speed of diffusion can increase exponentially with the electric field.\textsuperscript{125} This observation is compatible with our hypothesis.

We performed a control experiment at relative humidity below 15\% ($f = 20$ Hz, $V_a = 3.5$ kV). The lifetime of the transducers was $N = 3.3 \cdot 10^6 \pm 5.5 \cdot 10^5$, which is approximately three times larger than at 100\% relative humidity. Kornbluh \textit{et al.},\textsuperscript{118} and Biggs \textit{et al.}\textsuperscript{119} reported an increase in $N$ of a factor of 20 - 100 for dielectric elastomer transducers with carbon black as conductors when decreasing the relative humidity from 100\% to below 30\%. The lifetime of the transducers with ionic liquids is less sensitive to relative humidity, but even at low relative humidity they survive fewer actuation cycles than transducers with carbon black at high relative humidity under comparable operating conditions (we estimate a factor of $\sim 10$).
4.4.4 Correlation of the Lifetime with the Increase in Capacitance

To determine whether diffusion of ions into the membrane has an influence on the cycle lifetime of the transducers, we investigated if their lifetime correlates with the increase in capacitance over time. We fabricated the membranes with VHB 4910, biaxially stretched by a factor of three, and used a mixture of ionic liquid with PVDF (5:3 by weight) as the ionic conductor. We tested 10 ionic liquids, that did not swell VHB 4910 during the compatibility test (\([\text{EMIM}][\text{EtSO}_4], [\text{EMIM}][\text{BF}_4], [\text{BMIM}][\text{NTf}_2], [\text{BMIM}][\text{BF}_4], [\text{BMIM}][\text{PF}_6], [\text{HMIM}][\text{BF}_4], [\text{HMIM}][\text{PF}_6], [\text{OMIM}][\text{BF}_4], [\text{OMIM}][\text{PF}_6], \text{and } [\text{DMIM}][\text{BF}_4]\)). With each ionic liquid, we fabricated seven transducers. After fabrication we measured their capacitance, and stored them for 12 hours at 100 % relative humidity. We then measured the capacitance of each transducer again, and tested their lifetime at 100 % relative humidity with a square-wave signal between 0 kV, and \(V_a = 4.5\) kV with \(f = 10\) Hz.

The average change in capacitance ranged between 7.8 % for \([\text{BMIM}][\text{BF}_4]\) and 19.9 % for \([\text{EMIM}][\text{BF}_4]\) (Figure 4.4). Similar to the results of the compatibility test, there was no clear dependence of the change in capacitance on the nature of the ions (e.g., \([\text{BMIM}][\text{BF}_4]\) showed the lowest increase in capacitance, however its components appeared in combination with different counterions on the other end of the measured range). The average lifetime of the transducers ranged between \(N = 170\) for \([\text{BMIM}][\text{NTf}_2]\) and \(N = 170 \cdot 10^3\) for \([\text{EMIM}][\text{EtSO}_4]\) (Figure 4.4). \(N\) decreased exponentially with the increase in capacitance, indicating that diffusion of ions into the dielectric membrane and failure are correlated. Because we measured the increase in capacitance only at one point in time (after 12 hours), we do not know whether ionic liquids with low change in capacitance, diffuse more slowly (lower diffusivity \(D\)), or their saturated concentration
Figure 4.4 The lifetime under electromechanical actuation ($V_a = 4.5$ kV, $f = 10$ Hz) plotted against the change in capacitance over time within 12 hours of fabrication. Ionic liquids with larger increase in capacitance showed a shorter cycle lifetime. The errorbars show the standard deviation of the mean for 7 samples.
of ions in the membrane is lower (expressed by a smaller change in capacitance at saturation) compared to ionic liquids with large changes in capacitance.

4.5 Summary

This chapter establishes that, when ionic liquids are used as conductors for dielectric elastomer transducers, their lifetime under electrochemical actuation is related to the diffusion of the ionic liquid into the dielectric membrane. The change in capacitance of the transducers can be used as a measure for the diffusion of ions into the membrane. Transducers made with ionic liquids that provide a larger increase in capacitance, fail more rapidly than when an ionic liquid that provides a smaller increase in capacitance is used. The lifetime of the actuators under an applied electric field decreases exponentially with the strength of the electric field, and is not significantly affected by fatigue from repeated excitation.

Every ionic liquid tested diffused into the dielectric membrane. Some of the ionic liquids diffused into the membrane at concentrations sufficiently high to make it conductive, whereas others reached an equilibrium concentration at which the membrane was not conductive. This result is relevant for dielectric elastomer transducers, which are used as sensors. Because these sensors rely on the measurement of their capacitance, one must wait until the concentration of ionic liquids equilibrates inside the dielectric membrane to prevent drift of the measured results with time.

None of the ionic liquids used in this chapter lead to lifetimes that are large enough for a commercial application. The largest average number of actuation cycles measured was $3.3 \cdot 10^6$ (at $f = 20$ Hz, and $V_a = 3.5$ kV), which corresponds to $\sim 46$ hours of continuous use. Future investigations would have to include two targets: (i) determining why certain ionic liquids diffuse faster or at higher concentrations than others, and (ii) the mechanism due to which they cause
failure of the transducers under electromechanical excitation. With that knowledge one can determine the optimal combination of ionic liquid and dielectric membrane to build long lasting dielectric elastomer transducers.
Chapter 5

A Soft, Bistable Valve*

5.1 Introduction

Pneumatically actuated soft robots function by networks of channels that inflate upon pressurization, or buckle upon evacuation. Soft devices, and their actuators, are intrinsically compliant, and can move in ways that are difficult or impossible to achieve using ‘hard’ components. Other useful characteristics of soft actuators and devices include: (i) collaborative behavior: that is, intrinsic safety in operating closely with humans; (ii) the ability to adapt autonomously to different shapes; (iii) relatively low cost; (iv) ease of sterilization; (v) the ability to manipulate delicate objects; (vi) high cycle lifetime. One characteristic (and deficiency, in a sense of applications) of most current soft, pneumatic actuators is that they still rely on hard valves (typically solenoid valves) and electronic components for control, whether situated externally or integrated internally into the device itself. There is no simple, universal means of incorporating a soft controller (i.e., a ‘switch’, or other logic element) into a soft actuator.

Precisely controlling the motion of soft, pneumatic actuators can be difficult when they undergo large deformations, because the properties of the elastomeric materials are often intrinsically nonlinear, viscoelastic, and hysteretic in their behavior. Feedback control is further complicated by the need for sensors that can sustain the same strain as the actuators. The compliance of the elastomers, from which devices are fabricated,

* This chapter is based on unpublished work that was performed under my lead in collaboration with Alar Ainla, Lee Belding, Sarah Kurihara, Zhigang Suo, and George M Whiteside. Supplementary information for this chapter can be found in Appendix C.
allow them to conform to different shapes, and automatically limits the force exerted by them (a form of “material intelligence”), which makes them able to operate in many applications between limit set points. These set points allow soft actuators to be controlled with the simple on/off of a pressure supply. Grippers and walkers are two examples of successful applications operating between limit set points.

The most common methods of controlling pressure in soft robots involve “hard” valves (e.g., solenoid valves) that open or close in response to a pneumatic or electronic signal. These valves often must be located externally, and thus require tethering the robot to them (one tube/tether per actuator). Although pressure controls can also be integrated into the body of soft robots, they still require a signal (through a tether, or on-board electronic components). Solenoid valves are hard, heavy, and non-elastic and therefore sometimes cumbersome to integrate into soft robots. Existing control elements that have been developed for soft robots include: (i) a braille display, which allows the control of 32 pneumatic actuators simultaneously; (ii) band-pass valves, which can address multiple actuators individually, using a single, modulated source of pressure; (iii) unidirectional, soft check-valves, which have been integrated directly into soft robots for directional air flow into or out of soft actuators (e.g., to contain gas generated in an explosion to power the soft robot); (iv) microfluidic oscillators.

Many designs exist for microfluidic valves, logic circuits, oscillators, and even fluidic processors. In these designs, elastomeric membranes block or permit flow through channels depending on an applied input pressure (positive pressure, or vacuum). A microfluidic oscillator relies on a network of fluidic components, which include at least two valves (switches), channels (resistors), and chambers (capacitors), and a pressure source. The dimensions of the components must be balanced to achieve oscillatory behavior of the circuit. Wehner et al.
controlled a 3D printed robot with such a microfluidic oscillator. The small scale of microfluidic valves permits a high density of logic elements, but limits the flow rate, and thus the size of the actuator that can be controlled. This problem can, to some extent, be avoided by operating the microfluidic circuit with a liquid that produces a gas inside of the soft actuator to increase the volume (e.g., the platinum-catalyzed decomposition of H$_2$O$_2$ to O$_2$ and H$_2$O). The small feature sizes of microfluidic channels require the use of a different fabrication technique for the controller (soft lithography) than for the robot (3D printing or molding), and leads to difficulties when interfacing the microfluidic channels with the channels of the robot (clogging of the channels).

We have developed a soft, pneumatic, valve, which uses the snap-through instability of an approximately hemispherical membrane to switch between different pressures of air (or other gases) to control the air flow through pneumatic channels. This instability provides the valve with two properties: (i) the state of the valve is binary (open/closed). The snap-forward and the snap-backwards pressures of the bistable membrane determine the pressures at which the valve switches between its two states. We measured these pressures as a function of the thickness of the membrane, the central angle of the membrane, and the shear modulus of the material used to make the membrane. (ii) the snap-through instability is hysteretic. Connected to a source of constant pressure, the valve can generate periodic pressure oscillations (between the snap-forward, and the snap-back pressures) in a volume of air (e.g., a chamber, or an actuator), when employed in a feedback control scheme (i.e., when the output pressure of the valve acts as its control pressure). We characterized the oscillations in pressure for different air volumes and input pressures.

Snap-through instabilities have previously been used in the design of soft actuators, to achieve different types of motions. The valve presented in this work is different from these examples because it operates as a control element, which can be integrated into already existing
designs for soft, pneumatic actuators. The snap-through instability enables the valve to be designed with the needed switching pressures. We demonstrated the ease of implementation and utility of the valve in two applications: (i) *As a soft gripper, which automatically grasps objects upon contact:* We fabricated a “starfish” gripper that automatically grasps a tennis ball. When the “palm” of the gripper contacts the ball, the valve is triggered, causing the gripper to close around the ball. An externally applied pressure signal resets the valve, which re-opens the gripper. (ii) *As a soft earthworm, which advances using a constant source of pressure:* We integrated the valve into a linear bellows actuator. Connected to a source of constant pressure, the valve periodically inflates and deflates the actuator, which advances because of friction acting asymmetrically on its feet.

The valve can act as a switch for automated functions in soft devices, enabling autonomous feedback and feedforward control in soft actuators. The pressures at which the valve switches can be controlled by changing the shape of the membrane and its material. The design of the valve is simple, modular, and scalable. The ability to generate oscillations inside a robot makes it possible to construct a fully soft, untethered soft robot that can react to stimuli from its environment.

5.2 Experimental Details

5.2.1 3D-Printing of the Molds

We 3D-printed the molds for the walls of the transparent valve in Figure 5.1 on a Stratasys Objet30 photopolymer-based 3D printer from VeroBlue (RGD840). We 3D-printed all remaining molds using a Stratasys Dimension Elite 3D-printer, with a layer resolution of 0.178 mm. For exact dimensions of the molds we refer to Appendix C.
5.2.2 Preparation of the Pre-Polymer Solutions

In this study, we used five different elastomers (Ecoflex 30, Dragon Skin 10NV, Dragon Skin 30, Smooth-Sil 950, all from Smooth-On, and Sylgard 184 from Dow Corning). We prepared the pre-polymer mixtures of Ecoflex 30 and Dragon Skin 30 by mixing their components, A and B, in a 1:1 ratio. We stirred the mixture manually for ~ 5 min, followed by degassing for ~ 10 min. We prepared the pre-polymer mixture of Smooth-Sil 950 by mixing its components, A and B, in a 10:1 ratio, stirring the mixture manually for ~ 5 min, and degassing for ~ 10 min. We prepared the pre-polymer mixture of Dragon Skin 10NV by stirring its components, A and B, in a 1:1 ratio, stirring the mixture manually for ~ 2 min, and degassing for ~ 5 min. We prepared the pre-polymer mixture of Sylgard 184 by mixing its components, A and B, in a 10:1 ratio, stirring the mixture manually for ~5 min, and degassing for ~30 min.

5.2.3 Fabrication of the Devices for the Characterization of $\Delta P_1$ and $\Delta P_2$ as a Function of the Geometry

We filled the molds with the pre-polymer mixture (Dragon Skin 10NV) (Figure C.8) and waited until all the air bubbles in the mixture disappeared. The molds for the caps were covered with microscope cover slips to ensure homogenous thickness. The molds for the center of the devices have a lid, containing ventilation holes, so that excess material is squeezed out when the lid is placed on the mold. We cured the elastomers in the molds for two hours at room temperature before demolding. After demolding, we cut off the excess material on the center-piece, present due to the ventilation holes, with scissors. At each end of the center-piece of the device, we punched out two holes of 3 mm diameter (on opposite sides, ~ 5 mm from the end) (Figure C.9) to connect tubing to the valve. We used Dragon Skin 10 NV to glue the pieces together (Figure C.9), and cured the assembly in an oven at 60 °C for 10 min.
5. 2. 4 Fabrication of the Tubing Inside of the Valves

To fabricate the tubing inside the valve, we filled a syringe with the pre-polymer mixture (Smooth-Sil 950) and degassed it inside the syringe for 10 min. We injected the content of the syringe into the assembled mold (Figure C.10) through an opening at the bottom. We fabricated the conical tip (Dragon Skin 10NV) and the component used to connect the tubing to the conical tip (Smooth-Sil 950) with separate molds (Figure C.10). These two components were fabricated by pouring the pre-polymer mixture into their molds, allowing the air bubbles to dissipate, and covering the molds with a microscope cover slip to ensure homogenous thickness. We cured the pieces for 24 hours at room temperature before demolding. After demolding, the tubes were cut to the desired length (which varied depending on the demonstration). We glued the pieces together (Figure C.11) using Dragon Skin 10 NV as glue, and cured the assembly at 60 °C for 10 min.

5. 2. 5 Fabrication of the Transparent Valve

We filled the molds (Figure C.12) for the walls of the valve, and the end pieces with the pre-polymer mixture of Sylgard 184. For the horizontal channels in the end pieces we stuck 16-gauge needles into the molds through openings in their sides. We filled the mold for the membrane with the pre-polymer mixture of Dragon Skin 10NV. After waiting for the air-bubbles to disappear, we closed the molds of the walls, and the end membrane with vented lids, and covered the molds for the end pieces with glass slides. We cured the elastomers for 48 hours before demolding. We glued tubing into the top end piece (11-mm long, fabricated as above), and bottom end piece (20-mm long) (Figure C.13). We glued the bottom and top wall onto the membrane. Then we glued the tip of the bottom tubing onto the bottom of the membrane. Finally, we glued the end pieces to the walls of the valve. We used Dragon Skin 10NV as glue, and cured the glue after each step for 10 min at 60°.
5.2.6 Fabrication of the Automatic Gripper

We filled the molds with the pre-polymer mixture of Dragon Skin 30 (Figure C.14) and waited for the air bubbles to disappear. We covered the molds of the valve and the contact sensor with vented lids, and covered the mold for the top of the valve with a microscope coverslip. For the vertical channels in the valve, and the gripper, we stuck 16-gauge needles through openings in the lids into a notch in the bottom of the mold. To fabricate the strain limiting layer, we put a sheet of paper (VWR 21914-758) on an acrylic plate, and spread pre-polymer of Dragon Skin 30 on the paper with fingers. We cured for 16 hours before demolding.

We glued 13-mm long tubing (fabricated as above) into the base of the gripper, and 12-mm long tubing into the top cap of the valve, using the prepolymer of Dragon-Skin 30 as glue (Figure C.15). We then glued the conical tip of the top tubing to the top side of the membrane, and top end onto the walls of the valve. We inserted a syringe needle (16 gauge) in the vertical channel of the wall to prevent clogging. In the next step, we glued the conical tip of the bottom tubing to the bottom side of the membrane. Afterwards we glued the valve onto the gripper. A syringe needle (16 gauge) in the vertical channel of gripper prevented clogging. After each step we cured the glue for 10 min at 60 °C.

We punched a hole (2 mm diameter) into the strain limiting layer for the connection to the contact sensor, and glued the assembled gripper onto to the strain limiting layer. We used a 16-gauge needle in the opening of the bottom chamber of the valve (for the connection to the contact sensor), to prevent clogging. After curing at room temperature for 16 hours we cut out the shape of the gripper. We glued a single tube (6-mm long, fabricated as above) (Figure C.15) into the contact sensor. We then glued the other end of the tube to the opening in the palm of the gripper, and the rim of the contact sensor to the gripper.
We connected the gripper to the pressure sources, by puncturing the walls of the top chamber, the bottom chamber, and the channel in the base with hypodermic needles (16 gauge). We glued the needles to the gripper with the adhesive RTV 159 (Momentive).

5.2.7 Fabrication of the Oscillator

We filled the molds of the oscillator with the pre-polymer mixture of Dragon Skin 10 NV (Figure C.16). The channels in the end pieces were formed by placing needles (16 gauge) into their molds. We waited until the air bubbles in the mixture disappeared. We covered the mold for the end pieces of the valve with microscope cover slips. We closed the mold of the center part of the valve with its lid, and stuck a syringe needle (16 gauge) through the opening in the lid into a notch in the bottom of the mold. We cured the pieces for 2 h at room temperature before demolding. After demolding we glued tubing (20-mm long, fabricated as above) to the bottom and top end pieces (11-mm long) with Dragon Skin 10 NV as glue (Figure C.17). We then glued the top end piece on the valve with a syringe needle (16 gauge) in the vertical channel to prevent clogging. In a next step we glued the conical tip of the bottom tubing to the bottom side of the membrane. Then we glued the bottom end piece to the wall. After each step we cured the glue at 60 °C for 10 min.

5.2.8 Fabrication of the Earthworm

To fabricate the bellows actuator, we poured Dragon Skin 30 pre-polymer on the outer pieces of the mold (Figure C.18), and closed the mold around the core, squeezing out excess polymer. We filled the mold for the rear end piece of the actuator with Dragon-Skin 30 pre-polymer (Figure C.18), closed the mold after the bubbles disappeared, and stuck 16-gauge needles through the lid of the mold, to form the channels. We filled the mold for the front end piece with Dragon-Skin 30 pre-polymer, and covered it with microscope cover slips, after the air bubbles disappeared. We filled the mold for the retaining spring with Ecoflex 30. We molded the pieces of
the valve with Dragon Skin 10NV with 16-gauge needles stuck into the molds, to form the channels, analogously to section 5.2.7. We cured the pieces for 24 hours before assembling the earthworm in 3 steps (Figure C.19): (i) we assembled the rear end piece and the valve (length of tubing in bottom chamber 20 mm; for the top tubing we cut one tube 11 mm long, as before and left the other tube longer to connect to the external pressure source); (ii) we glued the end pieces on the bellows actuator; (iii) we glued the retaining spring on the earthworm. We used Dragon Skin 10NV as glue.

5.2.9 Characterization of $\Delta P_1$ and $\Delta P_2$ as a Function of the Geometry

To measure $\Delta P_1$ and $\Delta P_2$, we connected one opening in the bottom chamber of the valve with a syringe pump (Harvard PhD Ultra), and the other opening with a pressure sensor (Lex 1, Keller). For the connection we used silicone tubing. We pressurized the bottom chamber at a flow rate of 15 ml/min until the membrane snapped to the other side. At that speed, the measured pressure was independent of the flow rate (i.e., the flow resistance due to the tubing was negligible). Then we depressurized the bottom chamber at the same speed. For geometries that did not snap back at positive pressures, we carefully removed the tubing of the bottom chamber, so the membrane did not snap back during the process, and connected it to the top chamber. We then pressurized the top chamber at a flow rate of 15 ml/min. We recorded the pressure as a function of time with the software of the pressure sensor (Control Center Series 30).

5.2.10 Characterization of the Oscillator

We connected the input channel of the oscillator to house nitrogen, and the output channel to a glass jar (VWR 89000-236) through 14-gauge dispenser needles (McMaster Carr, 75165A117), and silicone tubing (2.5 mm inner diameter). We adjusted the air volume in the jar by filling it with water. We measured the pressure as a function of time, using a pressure sensor
(U5244-000002-002BA, TE Connectivity), which was recorded with MATLAB. Each data point in Figure 5.4D and E corresponds to the average over 60 s.

5. 2. 11 Characterization of the earthworm

We connected the input tube of the earthworm through silicone tubing (1.5 mm inner diameter) to house nitrogen, and put it on a clean, smooth acrylic plate, for increased friction. To measure the pressure inside the earthworm, we connected a pressure sensor (Lex 1, Keller) to the earthworm with a hypodermic needle pushed through the front end of the oscillator (not shown in Figure 5.5). We used the image analysis tools of MATLAB, to determine the positions of the front, and rear ends of the earthworm from a video recording of the earthworm as a function of time. We determined the position of the center by averaging the positions of the front, and the rear end.

5. 3 Results and Discussion

5. 3. 1 The Soft, Bistable Valve

Figure 5.1 shows the design of a valve that uses two instabilities: snap-through instability of a membrane, and kinking of a tube. The two instabilities act cooperatively to control airflow through the valve. In this design, a bistable, hemispherical membrane separates two chambers (Figure 5.1A, B). Elastomeric tubing leads through each chamber. When the membrane is curved downward (state 1, Figure 5.1A, B), the tubing in the bottom chamber kinks, and blocks air flow through it, while air flows freely through the tubing in the top chamber. When the membrane is curved upwards (state 2, Figure 5.1A, B) the opposite is true; the tubing in the top chamber kinks, blocking air flow through it, while the bottom chamber is open and allows air to flow through freely. Whether the membrane is curved upward or downward depends on the pressure differential between the bottom ($P_+$) and top ($P_-$) chambers ($\Delta P = P_+ - P_-)$.
Figure 5.1 Details of the soft, bistable valve. (A) A schematic design showing the components of the valve. The valve consists of a hemispherical, elastomeric membrane, separating two chambers. Control pressures in the bottom ($P_+$), and top ($P_-$) chambers deform the membrane. When the membrane is in the downward position (state 1), it blocks air flow through a tube leading through the bottom chamber, by kinking the tube. When the membrane is in the upward position (state 2), it blocks air flow through the top tube. (B) Photographs of the valve in both states (C) The pressure-displacement curve of the membrane. When the pressure difference, $\Delta P$, between the two chambers reaches a critical value, $\Delta P_1$, the membrane snaps to the upward position. When the pressure difference decreases below $\Delta P_2$ the membrane snaps back to the downward position. (D) We chose the length of the tubes such that they kink (and un-kink) during the snapping process. The states of the bottom tubing ($Q$), and the top tubing ($\bar{Q}$) are binary (i.e., open or closed), and hysteretic.
Initially, when $\Delta P = 0$, the membrane is curved down towards the bottom chamber (state 1). As $\Delta P$ increases (i.e., as the bottom chamber is pressurized) the membrane bends towards the top chamber, and because it is constrained by the walls of the valve, it compresses in area. When a critical pressure $\Delta P_1$ is reached, the membrane passes through the center of the valve, and expands again in the top chamber (state 2). This behavior can—depending on the geometry and material of the valve—lead to a negative tangential stiffness (Figure 5.1C). When an incompressible fluid (e.g., water) pressurizes the bottom chamber, the pressure decreases upon further deformation (dashed line, Figure 5.1C). When a compressible gas (e.g., air) is used, the energy stored during compression of the membrane releases, in a dynamic “snapping” motion of the membrane, to the top chamber. When $\Delta P$ decreases, the membrane again has to overcome the constraint of the walls of the valve, to return to state 1. To overcome this constraint, and snap back to the bottom chamber, the pressure must drop below a critical pressure, $\Delta P_2$. This type of snap-through instability is mechanically well understood, and is the basis for toy “poppers”.\textsuperscript{151,152}

While the membrane is being deformed, it compresses the tubing. At a critical compression—which depends on the length, diameter, and wall-thickness of the tubing—the walls of the tubing collapse, leading to a kink that blocks air-flow. The length of the tubing can be chosen such that the collapse starts and finishes within the snapping motion of the membrane. This coupling of the two instabilities leads to binary, opposite states of air flow (“on/off”) through the bottom tubing ($\mathcal{Q}$), and the top tubing ($\overline{\mathcal{Q}}$), with hysteretic switching behavior (Figure 5.1D).

In practice, when the bistable membrane is integrated into a soft robot, the interior of an actuator can act as one of the ‘chambers’ of the valve. Depending on the application, one of the chambers and/or one of the tubes can be omitted. Because we fabricate the parts of the valve by
molding, they can be directly incorporated into the mold for a soft actuator. This integration eliminates additional fabrication techniques.

5. 3. 2 Dependence of $\Delta P_1$ and $\Delta P_2$ on the Geometry of the Membrane

The critical pressures $\Delta P_1$ and $\Delta P_2$—the pressures at which the membrane switches from one state to the other—depend on the exact geometry of the membrane, and the walls. We studied the dependence of $\Delta P_1$ and $\Delta P_2$ on the thickness $H$ and the central angle $\theta$ of the membrane, keeping all other dimensions fixed (Figure 5.2A). We used a syringe pump to pressurize and depressurize the bottom chamber with air while the top chamber was kept at atmospheric pressure, and recorded the pressure in the bottom chamber as a function of time $t$. From the measured minima and maxima of the pressure-time curves, we determined $\Delta P_1$ and $\Delta P_2$. For some geometries, the membrane did not snap back, even when the pressure in the bottom chamber was reduced to atmospheric pressure (i.e., $\Delta P_2 < 0$). In these cases, we disconnected the syringe pump after the membrane snapped upwards, and pressurized the top chamber, keeping the bottom chamber in contact with atmosphere.

We studied the dependence of $\Delta P_1$ and $\Delta P_2$ on the thickness of the membrane by varying $H$ from 0.5 mm to 4.25 mm, using membranes with diameter $D = 20$ mm fabricated from Dragon Skin 10NV elastomer (Smooth-On) (see Figure C.1 for detailed dimensions, and Chapter 5. 2 for fabrication). We kept $\theta = 180^\circ$. The critical pressure required to snap the membrane upwards ($\Delta P_1$) increased approximately linearly with increasing thickness (Figure 5.2B). For the critical pressures at which the membrane snapped back ($\Delta P_2$), however, we observed three distinct behaviors: (i) for $H < 3.00$ mm the membrane did not snap back on its own, but had to be pushed back to the original position by pressurizing the other chamber (i.e., $\Delta P_2 < 0$); (ii) for $3.00$ mm $\leq H \leq 4.00$ mm the membrane snapped back when the pressure decreased below a critical value (i.e., $\Delta P_2 > 0$), which
Figure 5.2 Measurements of the critical pressures $\Delta P_1$ and $\Delta P_2$. (A) A design schematic of the apparatus used to measure $\Delta P_1$ and $\Delta P_2$, for different membrane thicknesses ($H$), and central angles ($\theta$) (The syringe pump, and the pressure sensor are not drawn to scale). We determined $\Delta P_1$ and $\Delta P_2$ by pressurizing and depressurizing the bottom chamber of the valve with a syringe pump, and recording the extrema of the measured pressure-time curve. (B) The critical pressures, $\Delta P_1$ and $\Delta P_2$, as a function of $H$. (C) The critical pressures, $\Delta P_1$ and $\Delta P_2$, as a function of $\theta$. (D) $\Delta P_2$ plotted against $\Delta P_1$ for valves with different $H$ and $\theta$. The boundary of accessible critical pressures is defined by the diagonal line, where $\Delta P_2 = \Delta P_1$ (because by definition, $\Delta P_2 < \Delta P_1$), and the values of $\Delta P$ for a valve with $\theta = 180^\circ$, and various $H$. Valves with critical switching pressures within this boundary are obtained when $\theta < 180$. For valves where $\Delta P_2 > 0$ kPa, the membrane has only one stable state (downwards), when no pressure is applied. For valves where $\Delta P_2 < 0$, the membrane has one stable state (downwards), and one metastable state (upwards), when no pressure is applied.
increased with $H$; (iii) for $H > 4.0$ mm we did not observe the snap through instability (i.e. the measured pressure-time curve was monotonous). We note that membranes with $H \leq 1$ mm were so thin, that they did not snap quickly to the other side, but transitioned between the states in a slow process during which the pressure did not change.

The behavior of the membrane is a result of two concurrent modes of deformation: (i) compression of the membrane, and (ii) bending of the membrane. As the pressure in the bottom chamber increases, the membrane compresses in area, because it is constrained by the walls of the valve. Once the membrane passes the center of the valve, it can expand again, in area. The walls of the valve, therefore, impose a barrier that must be overcome for the membrane to transition to the opposite chamber. This barrier is the origin of the snap-through instability. The membrane also bends as the pressure increases. The bending stiffness of the membrane provides a restoring force for the membrane to return to its original position. This restoring force is the origin of the difference between the pressures, $\Delta P_1$, and $\Delta P_2$.

The bending and compressional stiffness of the membrane both increase with $H$, and therefore, $\Delta P_1$ increased as $H$ increased (the bending stiffness increases approximately proportionally to $H^3$, whereas the compressional stiffness increases approximately proportionally to $H$). For $H < 3.0$ mm, the restoring force (due to bending) was too small to overcome the constraints of the wall. The membrane, thus, needed an additional force from the top chamber to be pushed back to its original position (i.e., $\Delta P_2 < 0$). For $H > 3.0$ mm the membrane spontaneously snapped back during depressurization of the bottom chamber at $\Delta P_2 > 0$, because the restoring force due to bending was large enough to overcome the constraint of the walls. When $H$ approached 4.25 mm, the bending stiffness dominated the stretching stiffness, so that $\Delta P_2$ converged with $\Delta P_1$, and the instability disappeared.
We also measured the values of $\Delta P_1$ and $\Delta P_2$ for membranes with $\theta$ ranging from 140º to 180º, while maintaining $H = 3.0$ mm (Figure 5.2C). The central angle, $\theta$, determines how much the membrane must be compressed, in hoop direction, to pass through the center of the valve. We, therefore, observed that lower values of $\theta$ led both to smaller $\Delta P_1$, and to smaller differences in the critical switching pressures ($\Delta P_1 - \Delta P_2$). For $\theta < 140^\circ$, we did not observe snap-through. When $\theta = 140^\circ$, we encountered a special case, where the membrane did not snap when pressurizing the bottom chamber, but only when depressurizing it.

Figure 5.2D illustrates the behavior of the valve with different geometries of the membrane. The range of achievable switching pressures is defined by the diagonal $\Delta P_2 = \Delta P_1$ (because for the snap-through instability $\Delta P_1 > \Delta P_2$) and the data measured for $\theta = 180^\circ$. Points within this region can be obtained by reducing $\theta$. It is possible to increase the size of the area by using a stiffer elastomer (or to decrease it by using a softer elastomer), because the critical pressures scale linearly with the shear modulus of the material (see Figure C.2 for comparison of $\Delta P_1$, and $\Delta P_2$ for Ecoflex 30 (shear Modulus $\mu = 26.2$ kPa, Figure C.3), Dragon Skin 10NV ($\mu = 72.0$ kPa, Figure C.3), and Dragon Skin 30 ($\mu = 261$ kPa, Figure C.3)). The curve $\Delta P_2 = 0$ splits the $\Delta P_2 - \Delta P_1$ plane into two regions, with distinctly different behaviors. These behaviors can be exploited to fabricate valves with different functionalities. In the region where $\Delta P_2 < 0$, the membrane has one stable state (downwards), and one metastable state (upwards) when $\Delta P = 0$. These membranes can be used to fabricate latching pneumatic switches that require pressure signals only for switching between the two states, and hold that state after the pressure signal is removed. In the region where $\Delta P_2 > 0$, the membrane only has one stable state (downwards) when $\Delta P = 0$, so it snaps back on its own when $\Delta P$ drops below $\Delta P_2$. These membranes can be used to fabricate non-latching pneumatic switches.
5.3.3 A Pneumatic Gripper for Automatic Grasping

We designed a soft gripper that automatically closes when it contacts an object, and can be re-opened with an external pressure signal. The gripper consists of five fast pneu-net bending actuators arranged circularly around a soft valve, with a contact sensor integrated in the palm of the gripper (Figure 5.3A). The contact sensor consists of an elastomeric cap, which surrounds a tube that connects the bottom chamber of the valve to the atmosphere. An air supply (pressure $P_V$) is connected to the bottom chamber of the valve. When an object compresses the cap, the venting tube kinks and blocks the flow of air. A second air supply (pressure $P_S$, kPa) is connected through the bottom tubing of the valve to a ring channel that distributes air to the five bending actuators (Figure 5.3A). The ring channel is also connected to the atmosphere, through the tubing that passes through the top chamber of the valve. When the membrane is in the downward position, it blocks air flow from the pressure supply to the ring channel, and allows air to flow from the ring channel to the atmosphere, thus leaving the actuators dormant. When the membrane is in the upward position, air flows from the pressure supply to the actuators, and air-flow leading from the actuators to the atmosphere is blocked, thus pressurizing the actuators. The second pressure supply ($P_S$) is also connected to the top chamber, through an external valve, so that pressure in the top chamber can be switched from atmospheric to that of the pressure supply, $P_S$.

We can explain the function of the pneumatic circuit with an analogous electric circuit (Figure 5.3B), in which the actuators act as a pneumatic capacitor, the tubing and the channels act as resistors, and the contact sensor and the external valve act as switches. The electronic equivalent to the bistable valve is a Schmitt trigger. A Schmitt trigger is a hysteretic switch with a continuous input (here, the input is the pressure difference between the bottom and top chambers of the valve) and a binary output (here, the output is the bending actuators connected to atmosphere
Figure 5.3 Details of a gripper that uses the soft, bistable valve to function automatically. (A) Design schematic of a gripper that automatically grasps objects upon contact. The gripper consists of five fast pneu-net bending actuators and a soft, bistable valve. The bending actuators are connected to a ring-shaped channel around the valve. When the membrane in the valve is in its downward position, the pressure supply for the bending actuators ($P_S$) is blocked, and the ring channel is connected to the atmosphere. A second pressure supply ($P_V$) leads to the bottom chamber of the valve and out through the contact sensor at the palm of the hand. The top chamber can be connected through an external valve to the atmosphere, or the pressure supply $P_s$. (B) An equivalent electrical circuit that represents the pneumatic control in the automatic gripper. (C) - (E) Photographs of the gripper, automatically closing around a tennis ball. When the gripper contacts the ball, air flow through the contact sensor is blocked (because the contact sensor compresses, and the tubing leading through it kinks). The bottom chamber pressurizes, the membrane snaps upwards, and the gripper closes around the tennis ball. (F) - (H) Photographs of the gripper holding, and releasing the tennis ball. When the top chamber is connected to the pressure source $P_S$ the membrane snaps downwards, the bending actuators deflate, and the tennis ball is released.
or to $P_s$). When the tubing in the contact sensor is open (i.e., the electronic switch is closed), air flows from the pressure source $P_V$, through the bottom chamber of the valve to the atmosphere. The flow resistance of the tubing into the bottom chamber, and through the contact sensor acts as a “voltage”-divider, so that the pressure in the bottom chamber (positive input of the Schmidt trigger) lies below the switching pressure $\Delta P_1$ of the Schmidt trigger. When an object kinks the tubing through the contact sensor, the switch in the contact sensor opens, and the pressure inside the bottom chamber increases to $P_V$. The Schmidt trigger switches, and air flows into the capacitor (the fingers of the gripper, which actuate). When we switch the top chamber of the valve (negative input of the Schmidt trigger) to the pressure source $P_s$, the Schmidt trigger switches back, and the capacitor empties to the environment (the fingers of the gripper vent, and the gripper opens).

We fabricated a gripper, with an integrated valve, using Dragon Skin 30 (see Chapter 5.2.6 for fabrication). We used Smooth-Sil 950 to make the tubing inside the chambers of the valve and the contact sensor, and Dragon Skin 10NV for the contact sensor. For the air supplies we used $P_V = 55$ kPa (for the valve), and $P_s = 69$ kPa (for the bending actuators). We used the gripper to pick up a tennis ball. Before the gripper contacted the ball, air vented through the contact sensor to the environment (Figure 5.3C). When the contact sensor, located at the palm of the gripper, touched the ball, the contact force due to the weight of the gripper kinked the tube leading through it. The bottom chamber of the valve pressurized, and the membrane snapped upwards (Figure C.4 shows a gripper where the top chamber of the valve was removed to show snapping of the membrane). Air flowed through the bottom tubing into the bending actuators (Figure 5.3E). From a video recording of the gripping process, we determined that the gripper began closing $\sim 100$ ms after contact with the ball, and was completely closed around the ball after $\sim 350$ ms. After the gripper was closed (Figure 5.3F), we could lift the ball (Figure 5.3G).
Because of the bistability of the membrane, the gripper stayed closed even in cases when the ball moved and did not close the contact sensor anymore after picking it up. To reset the valve, and vent the gripper, we connected the top chamber to the pressure source $P_s$ (Figure 5.3H). After switching the top chamber of the valve back to atmosphere (Figure 5.3I), we could reuse the gripper. The gripper opened in less than 1 s.

5.3.4 Feedback Control for Oscillatory Motion

Figure 5.4A shows a soft, bistable valve, designed to use an air supply of constant pressure to generate periodic pressure oscillations in a pneumatic capacitor (e.g., a soft actuator). In this design, the top tubing of the valve is connected to an air supply of pressure $P_s$, and the bottom tubing is connected to the atmosphere. Feedback is established by connecting the bottom tubing and the bottom chamber of the valve (i.e., the pressure of the bottom chamber is the same as the output pressure of the valve). A vertical channel within the wall of the valve connects the top tubing to the bottom chamber of the valve. To characterize the valve, we connected it to a glass jar. Figure 5.4B shows the electrical analog of the pneumatic circuit.

When the output pressure ($P$) of the valve is smaller than $\Delta P_1$, the membrane bends downward (state 1, Figure 5.4A), and air flows from the pressure supply, through the tubing in the top chamber, to the glass jar. Because of the feedback (Figure 5.4B), the membrane snaps upwards (state 2, Figure 5.4A) when the pressure in the glass jar exceeds the critical pressure $\Delta P_1$. The glass jar vents through the tubing in the bottom chamber to the atmosphere until the pressure drops below $\Delta P_2$, at which point the valve snaps back to state 1. This behavior leads to periodic oscillation of the output pressure between $\Delta P_2$, and $\Delta P_1$. Without the snapping motion of the membrane (i.e., if the transitions between the two states were continuous) the valve would
Figure 5.4 A pneumatic oscillator made from a soft, bistable valve. (A) Schematic of the pneumatic oscillator in two states. When the membrane is in its downward position, air flows from pressure source, $P_S$, through the bottom chamber of the valve into a jar of volume $V$, while air flow from the jar to the atmosphere is blocked. Because jar is connected to the bottom chamber of the valve (feedback), the pressure, $P$, in the jar and the bottom chamber is the same. When $P$ exceeds $\Delta P_1$, the membrane snaps upwards. When the membrane is in its upward position, air flow from the pressure source, $P_S$, is blocked, and the jar vents to the environment. When $P$ decreases below $\Delta P_2$ the membrane snaps back, and the jar pressurizes again. (B) An equivalent electrical circuit that represents the design for pneumatic feedback control used to generate oscillations. (C)–(D) Experimental data measured with a valve with $H = 3$ mm, and $\theta = 175^\circ$. (C) The measured pressure oscillates periodically between $P = 2.4$ kPa, and $P = 9.8$ kPa. (D) The rise time, $t_R$, decreases with $P_S$ and increases with $V$. (E) The fall time, $t_F$, increases with $V$, and is not significantly affected by $P_S$. 
equilibrate in a state in which the tubing through both chambers is partially open so that the air-flow into the jar equals the air-flow out of the jar, and oscillations would not occur.

Based on the design of the valve in Figure 5.4A, we fabricated a soft oscillator, using Dragon Skin 10NV for the valve ($D = 20$ mm, $H = 3$ mm, $\theta = 175^\circ$), and Smooth-Sil 950 for the tubing (see Chapter 5. 2. 7 for fabrication). We connected the soft oscillator to a glass jar, with a volume of $V = 300$ mL (we adjusted the volume of the jar by filling it with water). Using a pressure input of $P_S = 16$ kPa, we recorded the pressure inside the jar as a function of time (Figure 5.4C). The valve periodically, and autonomously, pressurized (rise time $t_R = 0.3$ s) and depressurized (fall time $t_F = 0.65$ s) the jar, which oscillated between $P = 0.24$ kPa and $P = 0.98$ kPa.

Figure 5.4D and Figure 5.4E show the dependence of $t_R$ and $t_F$ on the supply pressure $P_S$, for capacitors with volumes ($V$) ranging between 100 mL and 300 mL. The times $t_R$ and $t_F$ scaled with the volume of the capacitor, because less air is required to change the pressure in a smaller volume. Increasing $P_S$ led to faster pressurization of the jar, but did not significantly affect the fall time $t_F$. We observed the fastest oscillations (frequency 2.0 Hz) for $V = 100$ mL and $P_S = 11$ kPa, and the slowest oscillations (frequency 0.7 Hz) for $V = 300$ mL and $P_S = 10$ kPa.

The lower limit for $P_S$ to generate oscillations is determined by the critical pressure, $\Delta P_1$ (here 0.98 kPa), so we observed no oscillations at $P_S = 9$ kPa. Experimentally, we observed an upper limit for $P_S$, which depended on the volume of the jar (the last data-point of each measured curve), when $t_R$ ranged between 0.23 s and 0.28 s. Beyond this upper limit, the valve stopped oscillating after a few cycles, because the membrane equilibrated in a transition state (between state 1, and state 2), in which both channels of the valve were not completely kinked. For volumes $V = 50$ mL, we did not observe stable oscillations, possibly because $t_R$ was too short, even for $P_S = 10$ kPa. To test whether the upper limit of supply pressure ($P_S$) is dictated by the duration of
we introduced a 2-cm long tube, with an inner diameter of 0.79 mm, between the pressure supply and the valve to increase the flow resistance. We obtained stable oscillations even at $P_s = 50$ kPa, and $V = 50$ mL (Figure C.5), suggesting that $t_R$ is the limiting factor, and not $P_s$, or $V$. To determine if the behavior of the valve changes over time, we recorded the oscillations of a valve, using a constant pressure input of $P_s = 1.1$ kPa, connected to a glass jar ($V = 150$ mL). After $10^5$ cycles, we measured a 5% decrease of $\Delta P_1$, and a 3% decrease of the oscillation frequency (Figure C.6). The critical pressure $\Delta P_2$ did not change significantly.

5.3.5 Autonomous Earthworm-like Walker

We demonstrate that the valve can be used as a feedback controller for soft robots. Using the valve to generate oscillatory motion using air from a source of constant pressure ($P_s$), we designed a soft robot with earthworm-like motion (Figure 5.5A). The worm (see Chapter 5.2.8 for materials, and fabrication) consists of a bellows actuator (which expands in the axial direction), surrounded by a cylindrical sleeve (which acts as a restoring spring). One end of the bellows actuator contains the soft, bistable valve, while the other end is capped with a circular elastomeric disc. Both ends of the robot possess elastomeric feet, angled at 10°, to create asymmetric friction during expansion and contraction.

Figure 5.5B shows snapshots of the earthworm moving on a smooth surface, connected to an air supply of pressure $P_s = 17$ kPa. When the bellows actuator inflated, frictional forces at the feet caused the earthworm to bend upwards. This bending caused the front foot to contact the ground with its leading edge only, while the back foot touched the ground with its entire surface (Figure 5.5A). Thus, the front foot slid forward and the back foot stuck. During deflation, the bellows actuator bent downward, so that the front foot stuck and the back foot moved forward.
A

State 1: Inflation

- Constant pressure supply $P_1$
- Restoring spring
- Bellows actuator
- Front end moves
- Sliding contact
- Undeformed foot
- Sticking contact

State 2: Deflation

- Rear end moves
- Exhaust atmospheric pressure
- Tubing closed

B

$\text{t} = 0.0 \text{ s}$

$t = 20.7 \text{ s}$

$t = 49.4 \text{ s}$

C

Pressure (kPa)

Front end

Center

Rear end

Position (cm) vs. Time (s)
Figure 5.5 An autonomous soft robot, with earthwork-like locomotion. (A) Schematic diagram of an earthworm that uses a soft, bistable valve to generate periodic expansion, and contraction, from a source of constant pressure ($P_s$) for locomotion. The earthworm consists of a linear bellows actuator and a soft, bistable valve, integrated into one of the ends of the actuator. The design of the valve is the same as that for the pneumatic oscillator (Figure 5.4), with the bottom chamber of the valve connected to the bellows actuator. A cylindrical sleeve connects the two ends of the actuator, and acts as a restoring spring. When the bellows actuator inflates, frictional forces cause it to bend upward, and when it deflates, it bends downward. This bending causes asymmetric contact between the front and back feet of the actuator and the ground, and consequently, leads to asymmetric friction and directional movement. During inflation, the front feet slide and the back feet stick, and during deflation, the back feet slide and the front feet stick. (B) Photographs of the moving earthworm at three points in time. (C) Pressure inside the actuator throughout the movement of the earthworm, when $P_s = 17$ kPa. The pressure inside the actuator oscillates between 4.0 kPa, and 10 kPa, and the center of the worm travels at 8.4 cm/min. The red dots indicate the time at which the photographs in (B) were taken. Oscillations in the position of the front and back of the actuator are caused, predominantly, by rotation of the ends during bending.
The worm stretched and compressed each cycle by 12%, and the worm advanced at a rate of 8.4 cm/min (Figure 5.5C). The oscillation period was 1.8 s.

5.4 Summary

This article describes a new design concept for a pneumatic valve that consists entirely of soft components. The valve functions based on a snap-through instability, and uses pneumatic signals for control, which can be external signals, or internal feedback, depending on the pneumatic circuit required for a given application. It demonstrates that a feedback loop can be integrated with the soft valve to inflate and deflate a soft actuator—autonomously, and periodically—using a constant pressure input. This system is fundamentally different from the microfluidic oscillators reported previously; whereas microfluidic oscillators rely on the interplay of pneumatic capacitors, resistors, and valves, this valve oscillates due to two instabilities: snap-through instability of a hemispherical membrane, and kinking of a tube.

Here, we used a hemispherical membrane as the control element of the valve, but there is a multitude of other structures that show reversible snap-through behavior, and may be equally suitable for autonomous actuation of soft devices (e.g., the snap through instability of arched structures, or the instability during inflation of spherical or cylindrical balloons). We used pneumatic channels that ran parallel to the bistable membrane, although other designs are possible (e.g., feeding the tubing directly through the membrane, to fabricate pressure-release valves, or pressure-limiting valves (Figure C.7).

To fabricate autonomous, untethered robots, the valve may also be used in combination with energy sources, which are directly integrated into a soft device. The two chambers of the valve can be parts of two different actuators, to switch the valve depending on their differential pressure, to obtain coordinated motion. If the surrounding walls are designed to maintain structural
integrity under negative pressure, the valve may also be used with vacuum. The valve can be used to regulate the flow of fluids through the tubing of the valve. If, however, a fluid is used to control the membrane within the valve, the incompressibility of the fluid would prevent the membrane from snapping, and for that reason the oscillator would not work with a fluid-filled valve with a mechanism analogous to that which we describe (feedforward control is still possible).

Although the valves can be directly integrated into the mold of the actuators they control, they still require additional bonding steps during assembly. We envision that by using a 3D-printer that prints elastomeric materials, an entirely soft actuator, including the control elements, could be printed as one monolithic piece. Another limitation of the bistable valve is that the switching pressures ($\Delta P_1$, and $\Delta P_2$) do not depend solely on the geometry and material of the membrane, but also on the surrounding structure. To obtain the desired switching behavior, one has to design the membrane together with the soft actuators. The mechanics of the snap-through instability is well understood, so that computational models (e.g., a finite element simulation) can aid the design and optimization of the geometry of the membrane. The characterization performed in this work (Figure 5.2) gives general guidelines for how changes in geometry influence the switching pressures.

Soft materials allow structures to undergo deformation—caused by inherent instabilities—repeatedly, and this dynamic capability provides the basis for this work. Through our demonstration of the automatic gripper and the autonomous “earthworm”, we demonstrate that simple logic and control elements can be directly integrated into soft robots; this integration decreases their dependence on hard control elements, and is a step toward the design and fabrication of entirely soft, complex, autonomous robots.
Chapter 6

Electrically Activated Paper Actuators*

6.1 Introduction

Paper is an abundant, lightweight, and biodegradable material that has been used for centuries for printing, packaging, and absorbing liquids. Paper is a porous hydrophilic network of intertwined cellulose fibers. This network defines the mechanical and fluidic properties of the material. In recent years, paper has become increasingly interesting as a material in new applications. For example, we and others have used it for microfluidic and electroanalytical devices as the basis for low-cost diagnostics, as 3D scaffolds for cell growth, as a substrate for printed electronics, and in microelectromechanical systems (MEMS). A missing component for paper-based devices is an electrically controlled actuator that is embedded within the paper, can be fabricated by printing, and continues to operate when the paper that supports it is creased and/or folded. Paper actuators that fulfill these requirements have the potential to allow control of liquid transport in paper-based microfluidic devices, to enable assembly of micromachines through self-folding, and to serve as microactuators for paper devices. If mechanical work could be performed with paper actuators, we could build new kinds of “paper-based machines.”

Given the attractive properties of paper, there has been surprisingly little research done on the fabrication of paper actuators. Three examples are: (i) paper-based actuators made by

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* This Chapter is a modified version of the publication Hamedi, M. M.; Campbell, V. E.; Rothemund, P.; Güder, F.; Christodoulas, D. C.; Bloch, J.-F.; Whitesides, G. M.: Electrically activated Paper actuators, Advanced Functional Materials, 2016, 26, 2446-2458. The text of the publication was modified to fit within the format of the thesis, and the supplementary information of the publication integrated into the main text.
integrating magnetic particles in paper. The actuation in these systems is achieved by (and requires) an external magnetic field.\textsuperscript{15,186} This approach produces tunable composite materials, but requires large quantities of magnetic additives that are controlled by the application of a localized external magnetic field; this control is difficult and/or inconvenient to accomplish. (ii) electrostatic zippers. These actuators consist of two sheets of paper that are coated with carbon nanotubes and are separated by a dielectric layer. They actuate using electrostatic fields.\textsuperscript{60} The paper zippers are simple in design, and low in cost, but have the disadvantage that they require several kilovolts to actuate, and deliver only small movement. (iii) Electroactive papers.\textsuperscript{7,61,62} These structures are fabricated by printing electrodes (usually made from gold) on both sides of paper or of cellulose films. They are lightweight, and flexible, but have the drawback that they require metallic electrodes. This requirement adds to both their cost and complexity. Their performance also degrades over time, and they do not function when creased. Because paper is hydrophilic, most systems based on it are affected by the relative humidity (RH) of the environment.

This chapter describes the design and fabrication of electrically controlled paper actuators that exploit the hygroexpansive properties of paper; they expand or contract based on changes of moisture within the paper. We call these actuators “Hygroexpansive Electrothermal Paper Actuators” (HEPAs). Figure 6.1 shows a schematic representation of the four different types of HEPAs (straight, pre-curved, creased-curved, and creased-sawtooth) that we fabricated and characterized. These simple devices are made from paper, conducting polymer, and adhesive tape. They are lightweight and inexpensive, and they continue to operate when bent and scratched. We describe a theoretical model for their behavior, show that the actuation of the HEPAs is reversible over a large range of relative humidity, and demonstrate that they can be designed to move along
Figure 6.1 (A) Schematic representation of the fabrication of the HEPAs: i) Wax is printed and melted into the paper to form the fluidic channels; ii) a suspension of PEDOT:PSS is added to the channels; iii) the water evaporates and deposits the conducting film of PEDOT:PSS in the channel; iv) a piece of tape is attached onto the paper/PEDOT:PSS composite to act as a strain-limiting layer. (B) Photograph of the wax patterned channel. (C) Photograph of the paper/PEDOT:PSS composite. (D) Photograph of the cross section showing that the PEDOT:PSS is embedded in the paper channel. (E) Schematic representation of the four HEPAs (straight, precurved, creased-curved, and creased-sawtooth) and of their motion of actuation; \( L \), length; \( W \), width.
either a straight or curved path. We show that these actuators can be used for the manipulation of liquids and for the fabrication of an optical shutter.

6. 2 Experimental Details

6. 2. 1 Materials

Poly(3,4-ethylenedioxythiophene):poly(styrenesulfonic acid) (PEDOT:PSS solution 10 % in water) Clevios PH1000 was purchased from Heraeus and mixed with 5 wt% dimethylsulfoxide (DMSO). PEDOT:PSS was stable in ambient conditions, and upon drying, it formed thin films that were highly conducting (> 1000 Scm\(^{-1}\)),\(^{187}\) and resilient to bending and stretching.\(^{188}\) Whatman Chromatography Paper Grade A was purchased from Sigma-Aldrich. Scotch tape (40-\(\mu\)m thick) was used as the strain-limiting layer to direct the movement of actuation. This tape was unaffected by moisture and heat (even up to 100 °C).

6. 2. 2 Fabrication of HEPAs

The devices were fabricated by printing wax patterns with a Xerox ColorQube and melting the wax into the paper by placing the sheet of paper in an oven at 140 °C for 2 min. Then, the PEDOT:PSS solution was added with a pipette to the channels that were defined by the wax. The patterned actuators were cut, and a layer of Scotch tape was attached to obtain the HEPA.

6. 2. 3 Electrical Operation

A Keithley 2410 source-meter was used as a digital multimeter, and as a power source to deliver the power for resistive heating. The HEPAs were connected to the source-meter with copper alligator clips.

6. 2. 4 Thermal Characterization

Thermal images were acquired using a FLIR T600-Series infrared imaging camera.
6.2.5 Control of Relative Humidity

RH was controlled with two mass flow controllers [MKS 1179 A—1000 and 10 000 sccm], using LabView, to regulate the flow of nitrogen (to dry) and water vapor (to humidify) at different ratios into a glass chamber.

6.2.6 Calculation of Optical Shutter Intensity

MATLAB was used for calculating the light intensity from each frame of a video from the optical shutter. The intensity was calculated as the average gray-scale intensity of pixels corresponding to the circular optical window.

6.3 Results and Discussion

6.3.1 Design and Fabrication

Figure 6.1 sketches the fabrication of the HEPAs. The general fabrication process is as follows. We wax printed a fluidic U-shaped channel on paper. Using a pipette, we added to the paper a suspension of the conducting polymer (PEDOT:PSS) in water. The PEDOT:PSS suspension wicks into the predefined channel, and upon drying coats the cellulose fibers throughout the thickness of the paper sheet (rather than localizing on the surface, as seen from optical, and scanning electron microscope images; Figure 6.1B–D, and Figure 6.2). When dry it forms a conducting paper/PEDOT:PSS composite, which we use to provide electrothermal heating. We note that without the patterned wax barrier (which also spans the width of the paper), the PEDOT:PSS suspension would spread unevenly across the paper by capillary wicking, and would not form a well-defined conducting path. To direct actuation, we attached pieces of Scotch tape (40 µm thick), either on one side of the paper—yielding the straight, pre-curved or creased-curved HEPAs—or on alternating sides—yielding the creased-sawtooth HEPA—(Figure 6.1E). The final step consists in designing the form of the HEPA. We fabricated two shapes: (i) the
Figure 6.2 Scanning electron microscopic (SEM) images of (A) the wax/paper/PEDOT:PSS composite interface, (B) the paper, and (C and D) the paper/PEDOT:PSS composite at different magnifications.
pre-curved HEPA fabricated by manually curling one side of the straight HEPA, using a dull blade (similar to curling a ribbon);¹⁹⁰ (ii) the creased-curved HEPA fabricated by manually folding the straight HEPA.

The HEPAs operate because of the moisture-induced dimensional changes of the paper/PEDOT:PSS composite, and the absence of moisture-induced dimensional changes of the tape. When the resistive heating element is turned on, the composite heats, the moisture content of the paper is reduced, and the paper component of the composite contracts. When the resistive heating element is turned off, the composite absorbs moisture from the environment, the moisture content of the paper increases, and the composite returns to approximately its original shape. Since the tape is unaffected by moisture and heat (even up to 100 °C), it acts as the strain-limiting layer, thus the dimensional changes of the paper/PEDOT:PSS composite are translated into a bending motion.

We designed four types of HEPAs (straight, pre-curved, creased-curved and creased-sawtooth) that can perform a range of motions. Figure 6.1E shows schematic representations of the four types of HEPAs, and their initial (the electrothermal element is off, OFF state) and final (the electrothermal element is on, ON state) positions. For example, the initial position (OFF state) of a straight HEPA is straight and then it bends to a certain angle to reach its final position (ON state). A pre-curved HEPA, conversely, can perform the exact opposite motion compared to a straight HEPA; its initial position is curved, and its final position is straight (Figure 6.1E). The creased configurations of the HEPAs can perform more complex motions (e.g., angular and accordion-type motions) as each crease changes its angle independently. The total motion, therefore, is the sum of the change in angular motion of each individual crease. The exact motion of each configuration of HEPA is influenced by three parameters: (i) the consumed power; (ii) the
dimensions of the actuators; (iii) the relative humidity of the environment. In the following sections we examined in detail the effect of each of these parameters (Figure 6.5, Figure 6.6, Figure 6.9, and Figure 6.10 show photographic images of the HEPAs during actuation).

6.3.2 Consumed Power, Resistance, and Driving Voltage

The conducting paper/PEDOT:PSS composite is used as a resistive element to generate the heat necessary for a change in the moisture content—and in turn for actuation—of the HEPA. The speed of actuation is proportional to the heat generated (greater heat = faster actuation), and this heat is related to the power consumed, the resistance, and the driving voltage of the paper/PEDOT:PSS composite. Using infrared (IR) thermal images, we concluded that temperatures of 80 °C are high enough to generate maximum and fastest actuation at the lowest consumed power. To study the heat distribution, we recorded the thermal profile of the paper/PEDOT:PSS composite, using an IR camera, at different applied voltages, and recorded the corresponding power ($P$). The IR images indicated that at each voltage the temperature was uniform across the surface of the resistive element, and the heat dissipated into the wax barrier (Figure 6.3). We note that we did not observe any degradation in the paper/PEDOT:PSS composite (i.e., it did not dissolve or burn) when we increased the temperature to 100 °C for 10 min (this time is much longer than the required time for our actuation experiments). We calculated the normalized power per area ($P_n$), using $P_n = P / LW$ (we used $P_n$ because it is independent of the size of the actuator and is directly related to the temperature), and we concluded that we need $P_n = 0.3$ Wcm$^{-2}$ to reach a temperature of 80 °C.

Power is related to voltage and resistance through Ohm’s law ($P = V / R^2$). We measured the sheet resistance ($R_s$) across the conductive paper/PEDOT:PSS channel at room temperature at different PEDOT:PSS loading. The sheet resistance is a parameter that is independent of the
Figure 6.3 (A) Photograph of the paper/PEDOT:PSS composite (the conducting path) embedded in a sheet of paper. The dashed lines indicate the borders of the resistive element. (B) IR images of this conducting path at different consumed powers. (C) Normalized resistance ($R / R_\text{0}$) of the conducting path as a function of temperature, measured from the IR images.
length, \( L \), and the width, \( W \), of the HEPA \( (R_s = RW/4L) \). As expected, the resistance lowered with increased PEDOT:PSS loading (the lowest sheet resistance we obtained was \( R_s = 70 \ \Omega \text{sq}^{-1} \)). For our experiments, we fabricated HEPAs with sheet resistance at room temperature of around 350 \( \Omega \text{sq}^{-1} \).

The driving voltage of the actuator can be expressed as a function of the sheet resistance and normalized power \( (V = 2L(P_nR_s)^{0.5}) \). From this relation, we note that actuators with lower sheet resistance, and/or shorter length, operate at lower driving voltage for the same power per area (note that the driving voltage is independent of the width of the HEPA). In this study, we could, for example, actuate HEPAs with different dimensions (at \( P_n = 0.3 \ \text{Wcm}^{-2} \)) at driving voltages between 20 V (for actuators with \( W = 1 \ \text{cm}, \ L = 3 \ \text{cm} \)) and 100 V (for \( W = 1 \ \text{cm}, \ L = 10 \ \text{cm} \)).

Since the resistance changes with temperature, we calculated the sheet resistance as a function of power (and thus temperature). We plotted \( R(T)/R_0 \), where \( R_0 \) is the resistance at room temperature, and noted that the resistance decreased by less than 10% at the highest temperature (Figure 6.3C). We, thus, concluded that the change in resistance does not have any significant effect on the performance of the actuator, and, that we can therefore approximate the resistance as constant for a given dimension.

**6.3.3 Theoretical Model of the Motion**

To understand the actuation behavior of HEPAs, we developed an analytical mechanical model that relates the curvature \( \kappa \) of the bi-layer actuator to the temperature-induced strain \( \varepsilon_h \) (Figure 6.4). Because the paper/PEDOT:PSS composite (layer 1), and the strain limiting layer (layer 2) are linear elastic, their stress-relationship can be written as

\[
\varepsilon = \sigma / E_1 + \varepsilon_h,
\]

(6.1)
Figure 6.4 Mechanical model of the HEPA. (A) Undeformed state; the thickness of the paper/PEDOT:PSS is $t_1$ and its Young’s modulus $E_1$. The thickness of the strain limiting layer is $t_2$, and its Young’s modulus $E_2$. The length of the HEPA is $L$ (the width $W$ is not shown here). (B) Deformed state; when water evaporates from the paper/PEDOT:PSS composite, the HEPA curves to curvature $\kappa$, and changes length (strain $\varepsilon_0$). Both are measured from the centerline of the HEPA. The model assumes that the axial strain $\varepsilon$ changes linearly with the distance $y$ from the centerline.
\[ \varepsilon = \sigma / E_2. \] (6.2)

In equations (6.1), and (6.2) \( \sigma \) is mechanical stress, \( \varepsilon \) is the axial strain, \( \varepsilon_h \) the strain caused by electrothermal heating, and \( E_1 \) and \( E_2 \) are the Young’s moduli of the two layers. The strain energy of per unit volume for the two layers \( (U_1, U_2) \) become

\[ U_1 = \frac{1}{2} \sigma (\varepsilon - \varepsilon_h) = \frac{1}{2} E_1 (\varepsilon - \varepsilon_h)^2, \]

\[ U_2 = \frac{1}{2} \sigma \varepsilon = \frac{1}{2} E_2 \varepsilon. \] (6.3)

We modeled the HEPA as an Euler-Bernoulli beam, and assumed perfect bonding between the layers, so that the strain along the thickness of the actuator could be written as:

\[ \varepsilon = \varepsilon_0 - \kappa y, \] (6.4)

where \( \kappa \), and \( y \) are measured from the centerline of the HEPA, and \( \varepsilon_0 \) is the strain of the centerline (Figure 6.4).

The curvature of the HEPA in equilibrium can be determined by minimizing the strain energy of the entire actuator:

\[ U_t = LW \int_{(t_2 - t_1) / 2}^{(t_2 + t_1) / 2} \frac{1}{2} E_1 (\varepsilon_0 - \kappa y - \varepsilon_h)^2 \, dy + LW \int_{(t_2 - t_1) / 2}^{(t_2 + t_1) / 2} \frac{1}{2} E_2 (\varepsilon_0 - \kappa y)^2 \, dy. \] (6.5)

In equation (6.5), \( t_1 \) and \( t_2 \) are the thicknesses of the two layers, and \( L \) and \( W \) the length and the width of the actuator. The resulting formula for \( \kappa \) is independent of the length and the width of the HEPA.

\[ \kappa = \frac{6 \varepsilon_h E_1 E_2 t_1 t_2 (t_1 + t_2)}{\left( E_1 t_1^2 - E_2 t_2^2 \right)^2 + 4 E_1 E_2 t_1 t_2 (t_1 + t_2)^2} \] (6.6)

This result is analogous to a bi-layered beam under thermal expansion, but instead of a thermal strain, this model uses the hygroexpansive strain \( \varepsilon_h \) defined as the ratio between the changes in length to initial length as a function of moisture content \( m_c \). In certain regions, \( \varepsilon_h \)
is a linear, and reversible function of the moisture content\textsuperscript{192} described by equation (6.7), where $\beta$ is defined as the hygroexpansion coefficient, and $\Delta m_c$ is the change in the moisture content.\textsuperscript{193}

$$\varepsilon_h = \beta \Delta m_c$$ \hspace{1cm} (6.7)

A typical value of $\beta$ for many types of paper is $\approx 0.1$.\textsuperscript{192,194} By using equation (6.6), and the largest curvature that we measured (0.12 cm\textsuperscript{-1}), we calculated $\varepsilon_h = -0.3\%$ for a HEPA with $L:W = 10:1$. In this calculation, we used the following values for Young’s modulus and thickness: $E_1 = 1.6$ GPa, $E_2 = 2.0$ GPa, $t_1 = 180$ $\mu$m, $t_2 = 51$ $\mu$m. This value is in agreement with the reported values for the hygroexpansion of paper.\textsuperscript{195} In reality, $E_1$ can also change with the moisture content (the paper becomes stiffer as it dries), but the change in $E_1$ has a small effect on $\kappa$, and we assume $E_1$ to be constant. Equation (6.6) can also be used to optimize the relation between $t_1$, $t_2$ and $E_2$, and to design a HEPA that has maximum bending curvature for a given type of paper (with fixed thickness and elastic modulus).

6.3.4 Influence of the Dimensions of the HEPAs on Actuation, and Generated Force

Equation (6.6) predicts that the curvature of actuation, at constant temperature, is independent of the length of the actuators. Our experiments, however, showed that this prediction only holds true for longer actuators ($L:W$ ratios $\geq 3$). Short actuators show a smaller, length-dependent actuation (Figure 6.5). We reasoned that the boundary effects become important for shorter actuators, to the point that equation (6.6) is no longer valid, and the actuators displace by a smaller percentage (at the same power) than do longer ones. In this work, we only focused on actuators with large $L:W$ ratios ($> 3$) at an on-state temperature of around 80°C. In addition to the movement of the HEPAs, we also measured their static force, (at the tip of the actuators) using a
Figure 6.5 Photographic images of the actuators at different operating temperature/power, with two different $L:W$ ratios: (A) $L:W = 10:1$, and (B) $L:W = 6:1$ (RH = 40%); (C) Curvature vs. temperature measured for actuators with different $L:W$ ratios. The temperature was varied by varying the driving power of the actuators, and recorded at equilibrium with an IR camera. The curvature was calculated from photos of the actuators. The actuators moved parallel to the gravitational force, so that we could neglect gravity.
straight HEPA. The largest specific force (force/total weight of the HEPA) was measured to \( \approx 0.7 \text{ Ng}^{-1} \) (see Table 6.1). In contrast with the results from the free actuation (without any loading), the specific force of the actuators increased as the \( L:W \) ratio decreased. This behavior is expected because the stiffness (spring constant) of a bending beam increases with a decrease in length, which allows the actuator to generate a larger force. A noteworthy point about these results is that HEPAs can lift weights that are larger than their own weights. For example, the shortest actuator can lift around 20 times (0.57 g / 0.027 g) its own weight. We demonstrate this structure with the fabrication and operation of an optical shutter described in below.

6.3.5 Humidity Dependence

Because HEPAs actuate due to moisture-induced dimensional changes of the paper/PEDOT:PSS composite, we studied the influence of RH on the motion of bending, on the range of actuation, and on the speed of actuation (Figure 6.6 and Figure 6.7). As shown in Figure 6.6, we operated the same HEPA in two environments of different humidity: (i) dry (RH = 15 %), and ii) humid (RH = 85 %). We started the experiment at low RH (15 %). When we turned on the electrothermal element, moisture evaporated from the paper/PEDOT:PSS composite, the HEPA moved down, and the extra paper section (attached to the end of the HEPA) contacted and absorbed a droplet of a solution of a blue dye (Figure 6.6B). When we turned off the electrothermal element, the HEPA returned to its original position (Figure 6.6C). We then increased the RH to 85 %, and repeated the experiment (Figure 6.6D-F). At this RH, the paper/PEDOT:PSS composite absorbed more ambient moisture, which resulted in an initial state (off-state) with greater curvature Figure 6.6D). Its final state (on-state), however, was not visibly affected by the increase in RH, and the actuator picked up a solution of a red dye (Figure 6.6E). We tested the speed of actuation and recovery of the same HEPA at seven different RH (15 % – 85 %) and \( P = 1 \text{ W} \) (Figure 6.7). We
Table 6.1 The maximum static force measured for actuators with different $L:W$ ratios.

<table>
<thead>
<tr>
<th>Length / Width [cm / cm]</th>
<th>Weight of the paper in the actuator [g]</th>
<th>Maximum static force / Weight [mN]</th>
<th>Maximum specific force [Ng$^{-1}$]</th>
</tr>
</thead>
<tbody>
<tr>
<td>6.5 / 0.85 = 7.4</td>
<td>0.015</td>
<td>1.86</td>
<td>0.13</td>
</tr>
<tr>
<td>5.5 / 0.85 = 6.5</td>
<td>0.013</td>
<td>3.53</td>
<td>0.27</td>
</tr>
<tr>
<td>3.5 / 0.85 = 4.0</td>
<td>0.080</td>
<td>5.58</td>
<td>0.68</td>
</tr>
</tbody>
</table>
Figure 6.6 Time-lapse photographs of the same precurved HEPA ($L:W = 5:1$ cm) operated at room temperature (at $P = 1$ W and $I = 1.5$ mA): (A) at RH = 15 % and (B) at RH = 85 %. The volumes of the solutions of the red and blue dyes are 50 µL each.
Figure 6.7 (A) Actuation and (B) recovery times at different RH ($P = 1 \, \text{W}$); in each case, the time is constant within ± 10%.
note that for the same HEPA \((L:W = 10:1)\), the speed of actuation, and of recovery, is not noticeably affected by the RH (Figure 6.7). We also tested the HEPA at RH = 35 % and \(P = 2 \, \text{W}\), and we note that the speed of actuation (18 s) and of recovery (16 s) are not influenced by this difference in power. These experiments demonstrate that (i) the HEPAs can move weights (in the form of a paper appendix, and water droplets), (ii) the HEPAs actuate over a wide range of values of RH, because electrothermal heating releases the majority of the moisture absorbed in the paper irrespective of RH, and (iii) the speed of actuation of the HEPAs is not noticeably affected by the RH or the consumed power.

6.3.6 Creased HEPAs

An attractive property of HEPAs is that they function even when folded/creased. Unlike other paper actuators (fabricated with metallic conductors on the surface of the paper), HEPAs do not break, upon repeated creasing when subject to high strain on bending. To test the influence of creasing on the paper/PEDOT:PSS composite, we fabricated a conducting path \((1 \, \text{mm} \times 18 \, \text{cm})\), folded it 11 times, and measured its resistance as a function of the fold angle (Figure 6.8). The resistance changed less than 10 % between the unfolded and folded structures, even after 1000 open/close cycles (Figure 6.8). Figure 6.9A shows time-lapse photographs of the creased-curved HEPA that operated by closing and opening the half circular shape. This HEPA closed at a greater angle than the pre-curved variations, and could lift a weight (a piece of paper, \(m = 46 \, \text{mg}\)).

Figure 6.9B shows time-lapse photographs of a creased sawtooth HEPA that was creased in an accordion-shape. The difference between these two designs was the position of the tape. When the tape is positioned on one side of the paper/PEDOT:PSS composite, the angular motion of each crease is in the same direction; this concerted motion resulted in curving (Figure 6.9A). When we positioned the tape on alternate sides of the composite, the cumulative angular motion
Figure 6.8 (A) Photographs of an accordion shaped folded paper structure at different compression lengths. (B) Normalized resistance of the paper electrode measured from one side of the folded paper to the other side, as a function of compressed length. (C) Normalized resistance ($R / R_0$) measured at the uncompressed state, as a function of number of cycles, where each cycle corresponds to a complete closing and opening of the folded structure (i.e., moving each crease from $180^\circ$ to $0^\circ$ angle).
Figure 6.9 Time-lapse photographs of: (A) creased-curved HEPA ($L:W = 7:0.8 \text{ cm}$): dashed lines highlight the attached passive paper; (B) creased-sawtooth actuator fabricated with the tape positioned on alternating side of a HEPA ($L:W = 5:1 \text{ cm}$; $P = 1 \text{ W}$; $I = 1.5 \text{ mA}$; $\text{RH} = 40 \%$).
resulted in linear actuation (Figure 6.9B), which is an essential motion in many mechanical machines.

6.3.7 Proof-of Concept of Application: A Paper Optical Shutter

Figure 6.10 shows a pre-curved HEPA that acts as an optical shutter. We designed this HEPA to have an almost semi-circular shape, with an extra paper section (shutter), and mounted it onto a backlit table (Figure 6.10). When the electrothermal element was off, the shutter covered the hole and blocked the light. When we turned the electrothermal element on, the shutter moved, uncovered the hole, and allowed the light to pass through (Figure 6.10C). The shutter opened in 8 s, and closed in 16 s (at room temperature and RH = 40 %, see time-lapse photographs in Figure 6.10C). We tested 260 open/close cycles and did not observe degradation of actuation (see Figure 6.10B).

6.4 Summary

HEPAs made using paper, conducting polymer, and tape can be actuated using the hygroexpansive properties of paper and are controlled electrically by heating. The central element is the porous conducting path (used to provide electrothermal heating). This conducting path is made by coating the fibers in the paper (across the thickness of the sheet) with a conducting polymer (PEDOT:PSS) and thus making a paper/polymer composite that retains the porous and hydrophilic properties of paper. Upon electrothermal heating, the moisture content of the paper decreases, and the paper/polymer composite contracts. When the electrothermal heating element is turned off, the paper/polymer composite absorbs moisture from the environment (moisture content increases), and expands. Using theoretical calculations, we have shown that HEPAs can operate over a range of voltages, and that a normalized power per area of 0.3 Wcm⁻² is suitable for achieving maximum actuation. We derived a mechanical model that relates the curvature of
Figure 6.10 (A) Photograph of a HEPA ($L:W = 5:1$ cm) optical shutter ($P = 1$ W; $I = 1.5$ mA; RH = 40 %). (B) Number of on/off cycles versus normalized light intensity calculated from the videos of actuation. (C) Time-lapse photographs of the HEPA optical shutter. The device is placed on a backlit table, and the light is seen only when the shutter opens. The dashed lines highlight the shutter area.
the actuator to its dimensions, material parameters, and hygroexpansion. We showed experimentally that this model could be used for actuators with a length to width ratio greater than three (actuators with $L:W < 3$ are less power-efficient). We showed that the driving voltage of the HEPA was independent of $L$, and that larger actuation required larger $L:W$ ratios. We measured a maximum specific static force of $0.7 \text{ Ng}^{-1}$, and we showed that HEPAs with smaller $L:W$ ratios have larger specific static force.

The HEPAs have four advantages: (i) they are lightweight, inexpensive, and biodegradable. (ii) they are easy to fabricate using simple printing techniques. (iii) they operate even when bent and scratched because the element that provides electromechanical work is embedded within the paper. These features offer the possibility of designing complex folded structures. (iv) they can actuate many times without showing signs of degradation of performance. HEPAs are similar to other paper-based actuators, in that they have the disadvantage that their initial state is influenced by the relative humidity of the environment. Unlike most other paper-based actuators, however, their final state is not affected by relative humidity, as it is contingent on the electrothermal element drying the paper/polymer composites. They also have obvious shortcomings: they exert only small forces, and are relatively slow.

Due to their attractive properties (i.e., cost, weight, printability, and porosity), HEPAs could be useful in applications benefitting from monolithic integration in paper-based printed microfluidic, and electronic devices, paper MEMS, printable and foldable micromachines, and robots. Their speed of actuation, and the force they produce, however, is low (by standards of more conventional electromagnetic and pneumatic/hydraulic systems), but they are also lighter, much less expensive, and much more easily integrated with paper devices (diagnostic, bioanalytical, and electromechanical systems, for example) than are the
more universal systems. The systems in Figure 6.6, for example, might have the right characteristics to be used to sample a droplet medium in bioanalysis, and the one in Figure 6.9 to reposition a filter in a low-cost environmental monitor. These systems have the functional advantage that they can be reconfigured by folding and creasing without damaging them. The ability to fabricate small (submillimeter scale) porous wires of PEDOT:PSS that extend across the full thickness of a sheet of paper by using capillary wicking of an aqueous suspension of PEDOT:PSS into a channel defined by wax-printed walls, is a new fabrication technique with the potential for wide application in paper-based devices and machines, especially when folding and creasing are an issue.\textsuperscript{58,182,183}
Chapter 7

Conclusions

This dissertation explored the mechanics of three types of soft actuators experimentally, and theoretically: (i) dielectric elastomer actuators; (ii) fluid-driven actuators; (iii) paper actuators. This chapter summarizes the main results presented in this dissertation, and presents concluding thoughts, and questions which emerged from this work, and which are interesting for future exploration.

7.1 Summary

Chapter 2 introduced a stretchable, transparent conductor, which functions using an ion-conducting hydrogel. The novelty of this conductor is the placement of an insulating layer into the path of the electric current; this layer suppresses electrochemical reactions. Because electrochemical reactions are suppressed, we could use the ionic conductor for dielectric elastomer transducers, which require electrical voltage signals on the order of kilovolts. To demonstrate that the ionic conductor is fast enough to transmit signals within the audible frequency range (20 Hz - 20 kHz), we used it in a transparent dielectric elastomer loudspeaker. Compared to transparent, stretchable, electronic conductors, the ionic conductor had a higher optical transmittance.

Next, in Chapter 3, we characterized the acoustic properties of a transparent membrane based on the design of the transparent loudspeaker of Chapter 2, and demonstrated its use in active noise cancellation. We showed that the membrane responds linearly to electromechanical excitation (for excitation voltages below a frequency dependent threshold). For the linear regime,
we developed a mechanical model to describe sound transmission, reflection, and generation by the membrane. We performed an active noise cancellation experiment with a feedforward control algorithm, which we derived from the linear model. In this experiment we reduced sound transmission through the transparent membrane by a factor of three compared to passive sound absorption.

We then investigated the durability of dielectric elastomer transducers with ionic liquids as conductors (Chapter 4). We used ionic liquids because they are, unlike hydrogels, involatile. The work showed that ionic liquids diffuse into the dielectric membrane, and cause an increase of the capacitance of the dielectric elastomer transducers. Some ionic liquids swell the dielectric membrane with high enough concentrations to make the membrane ionically conductive. In this case, the dielectric elastomer transducer loses its function. Other ionic liquids reach an equilibrium concentration in which the membrane remains insulating. Under electromechanical excitation, the transducers fail, independent of the excitation frequency, after a certain amount of time, which decreases approximately exponentially with the excitation voltage. Dielectric elastomer transducers made with different ionic liquids increased show different increases in capacitance with time. The lifetime under electromechanical actuation of these transducers decreases approximately exponentially with the increase in capacitance with time.

Chapter 5 described a soft valve that uses the snap-through instability of a hemispherical membrane to control pneumatically driven actuators. The membrane separates the valve into two chambers. Depending on the differential pressure between the chambers, the membrane is curved towards one of the chambers, and away from the other chamber. The transition between these two states occurs in a fast snapping motion. We studied the dependence of the critical pressures at which the membrane snaps between the two states on its geometry. The membrane acts as a switch
that blocks, depending on its orientation, the flow through one of two pneumatic channels that lead through the valve. We utilized the hysteresis of the snap-through instability to design a soft oscillator that uses an air source of constant pressure to generate periodic oscillations of pressure between the critical pressures, and characterized its properties. To demonstrate the utility of the soft valve, we integrated it into a soft gripper that automatically grasped objects upon contact, and a soft earthworm-like robot which automatically advanced using an air source of constant pressure.

Finally, in Chapter 6, we developed a paper actuator that uses the volume change during hygro-expansion of paper to generate bending motion. The actuator consists of a paper layer with an integrated electrically conducting channel (PEDOT:PSS), bonded to an adhesive tape. When we applied a voltage to the conductive channel, resistive heating caused the water in the paper to evaporate, and the paper layer contracted. Because the adhesive tape was unaffected by the heat, the actuator bent. We developed a mechanical model for the behavior of the actuators, and studied the dependence of the bending deformation on the geometry as a function of the temperature of the paper layer, and the relative humidity of the environment.

7.2 Outlook

Even though we demonstrated the stretchable, transparent, ionic conductor in Chapter 2 only in dielectric elastomer transducers, it can also be used for other applications. Since the publication of this work, ionic conductors have been used in various devices, which include stretchable cables,197 electroluminescent displays,198,199 liquid crystal displays,200 and washable conductive fibers.111 These examples show that ionic conductors can perform many functions of electronic conductors, and may outperform them in applications that require stretchability. The human body transmits signals based on ionic conduction, whereas most of our technology is
electronic. Ionic conductors offer a new way of interfacing the human body with electronic devices (e.g., perhaps for medical diagnosis).

By characterizing the transparent membrane, and performing the active noise cancellation experiment in an impedance tube in Chapter 3, we simplified the acoustic field to one dimension. Real applications would involve complex, directional acoustic fields, which may influence the performance of the membrane as a noise cancellation device. Future work would therefore have to include testing the membrane under more realistic conditions. We also determined the parameters of the linear model for the transparent membrane only empirically. A mechanical model, which relates the parameters of the linear model to the material, and to the geometry of the membrane, would help to develop a transparent membrane with improved acoustic properties.

Chapter 4 demonstrated that different ionic liquids diffuse into the dielectric membrane at different speeds and concentrations, and that this diffusion is correlated with the lifetime of dielectric elastomer actuators under electromechanical excitation. We could not answer two important questions: (i) why do different ionic liquids interact differently with the membrane? (ii) why is the time to failure independent of the excitation frequency, and decrease approximately exponentially with the applied voltage? To find pairs of ionic liquid and dielectric membrane, which lead to long lifetimes, with an approach that goes beyond simple trial and error, these questions must be answered.

The design of the soft valve presented in Chapter 5 is bulky, and the fabrication of the tubing, which leads through its chambers, cumbersome. Possible future research could involve finding a method to simplify the fabrication of the tubing, and to scale the valve down. In the automatic gripper, the valve acted like a (hysteretic) transistor. The valve can therefore also be
used as a building block for complex logic circuits that might be integrated directly into a soft robot.

In Chapter 6 we controlled the hygroscopic expansion of the paper actuator by heating the paper layer electrically. Other mechanisms could be exploited to change the water content the paper layer (e.g., heating by light, or change of relative humidity of the environment) to fabricate passive control elements, such as a humidity controller for a room that opens, or closes an air vent depending on the relative humidity inside the room.
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A. 1 Voltage Across the Electrode/Electrolyte Interface

The electrode/electrolyte interface forms an electrical double layer (EDL), and behaves like a capacitor when the voltage across the interface is sufficiently small (Figure 2.5). The EDL and the dielectric are in series. Consequently, when voltage \( V \) is applied between the two electrodes, the two capacitors add the same amount of charge, \( Q \). To estimate the behavior of the circuit, we assume that both capacitors are linear: \( Q = C_{\text{EDL}} V_{\text{EDL}} \) and \( Q = C_D V_D \), where \( C_{\text{EDL}} \) is the capacitance of the EDL, and \( C_D \) the capacitance of the dielectric. Thus,

\[
C_{\text{EDL}} V_{\text{EDL}} = C_D V_D. \tag{A.1}
\]

The capacitance of the EDL is \( C_{\text{EDL}} = c_{\text{EDL}} A_{\text{EDL}} \), where \( c_{\text{EDL}} \) is the capacitance per unit area of the EDL, and \( A_{\text{EDL}} \) the area of the EDL. The capacitance of the dielectric is \( C_D = \varepsilon A_D / H_D \), where \( \varepsilon \) is the permittivity, \( A_D \) is the area of the dielectric, and \( H_D \) is the thickness of the dielectric. Also note that \( V_D = H_D E_D \), where \( E_D \) is the electric field in the dielectric. Rewrite equation (A.1) as

\[
\frac{A_{\text{EDL}}}{A_D} = \frac{\varepsilon_D E_D}{c_{\text{EDL}} V_{\text{EDL}}}. \tag{A.2}
\]

The electric field in the dielectric is limited by the electrical breakdown field \( E_{\text{EB}} \), while the voltage across the EDL is limited by the range within which electrochemical reaction is averted. For representative values \( E_{\text{EB}} = 10^8 \text{ V/m} \), \( \varepsilon_D = 10^{-11} \text{ F/m} \), \( c_{\text{EDL}} = 10^{-1} \text{ F/m}^2 \) and \( V_{\text{EDL}} = 1 \text{ V} \), we find that \( A_{\text{EDL}} / A_D = 10^2 \). When the area of the electrode/electrolyte interface is sufficiently large, the device will be limited by the electrical breakdown of the dielectric, rather than by the
electrochemical reaction at the EDL. If needed, the area of the electrode/electrolyte interface can be increased, for example, by using a porous electrode, similar to that used in a supercapacitor. The electrode/electrolyte interface may form thin layers of reaction products, so long as they are stable under a small voltage.

A. 2 Equilibrium States of Layered Electrolytic and Dielectric Elastomers

This calculation assumes a square dielectric membrane, length \( L \) and thickness \( H_0 \) in the undeformed state. The elastomer is taken to be incompressible. The dielectric membrane is prestretched by an equal-biaxial force \( P \) to length \( \lambda_{\text{pre}}L \) and thickness \( H_1 = H_0 \lambda_{\text{pre}}^{-2} \), and then sandwiched between two membranes of an electrolytic elastomer, of combined thickness \( H_2 \) in the undeformed state. Subject to both the force \( P \) and the voltage \( V \), the sandwich is further stretched to length \( \lambda L \). Dielectric elastomers show pronounced strain-stiffening, and are represented by the Gent model.\(^{201}\) In equilibrium, the equation of state takes the form:\(^{202}\)

\[
\frac{P\lambda}{\lambda_{\text{pre}}^2 L (H_1 + H_2)} + \phi^\alpha \varepsilon \left( \frac{V}{H_0} \right)^2 \lambda^4 = \frac{\phi^\alpha \mu^\alpha \left( \lambda^{-2} - \lambda^{-4} \right)}{1 - \left( 2\lambda^{-2} + \lambda^{-4} - 3 \right) / J_{\text{lim}}^\alpha} + \frac{\phi^\beta \mu^\beta \left( \lambda_{\text{pre}}^{-2} \lambda_{\text{pre}}^{-2} - \lambda_{\text{pre}}^{-4} \right)}{1 - \left( 2\lambda_{\text{pre}}^{-2} + \lambda_{\text{pre}}^{-4} - 3 \right) / J_{\text{lim}}^\beta},
\]

where \( \phi^\alpha = H_1 / (H_1 + H_2) \), \( \phi^\beta = H_2 / (H_1 + H_2) \), \( \varepsilon \) is the dielectric permittivity, \( \mu^\alpha \) is the shear modulus of the dielectric elastomer, and \( \mu^\beta \) is the shear modulus of the electrolyte. Similarly, \( J_{\text{lim}}^\alpha \) and \( J_{\text{lim}}^\beta \) are the parameters related to the stiffening of the elastomer and the electrodes. Equation (A.3) defines the stretch of the membrane subject to given values of \( P \) and \( V \).

Define the area strain as \( \varepsilon_{\text{area}} = \left( \lambda / \lambda_{\text{pre}} \right)^2 - 1 \). We compare the predicted area strain as a function of the voltage to the experimental data for the actuator of a prestretch of \( \lambda_{\text{pre}} = 3 \) (Figure 2.2G). A good agreement between theory and experiments is observed, when the shear
modulus of the dielectric elastomer is chosen as $\mu^\alpha = 34$ kPa, with $J_{\text{lim}}^\alpha = 90$. The fitted shear modulus compares well with those reported elsewhere.$^{115,203}$ For the dielectric elastomer, the permittivity is $\varepsilon = 4.11 \times 10^{-11}$ F/m. The shear modulus of the hydrogel is set to the experimentally determined value of $\mu^\beta = 3$ kPa, and we assume a neo-Hookean stress-strain relation with $J_{\text{lim}}^\beta = \infty$.

A. 3 Viscoelasticity of Layered Electrolytic and Dielectric Elastomers

Viscoelastic dielectric elastomers can be simulated using rheological models of springs and dashpots.$^{108}$ Here we adopt a model of two polymer networks connected in parallel (Figure 2H). One network consists of a spring $\alpha$, and the other network consists of another spring $\beta$ connected in series with a dashpot. Subject to a force, the networks deform by a stretch $\lambda$. By geometry, the spring $\alpha$ deforms by stretch $\lambda$. For the bottom network, the spring $\beta$ deforms by stretch $\lambda^e$, and the dashpot deforms by stretch $\xi$. The total stretch in the bottom network is given by $\lambda^e \xi$ which must be equal to $\lambda$. The equation of motion may be written as:

$$\frac{P \lambda}{\lambda_{\text{pre}}^2 L (H_1 + H_2)} + \frac{\phi^\alpha \varepsilon}{H_0} \left( \frac{V}{H_0} \right)^2 \lambda^4 = \frac{\phi^\alpha \mu^\alpha (\lambda^2 - \lambda^4)}{1 - (2\lambda^2 + \lambda^4 - 3)/J_{\text{lim}}^\alpha} + \frac{\phi^\beta \mu^\beta (\lambda^2 - \lambda^4)}{1 - (2\lambda^2 + \lambda^4 - 3)/J_{\text{lim}}^\beta},$$  \hspace{1cm} (A.4)

where $\mu^\alpha$ and $\mu^\beta$ are shear moduli of the two springs, and $J_{\text{lim}}^\alpha$ and $J_{\text{lim}}^\beta$ are constants related to the limiting stretches of the two springs. The three terms on the right-hand side of equation (A.4) are the stresses carried by the two springs in the rheological model.

We next model the dashpot as a Newtonian fluid. In the rheological model, the state of stress in the dashpot is the same as that in spring $\beta$. Consequently, we define the rate of deformation in the dashpot:

$$\frac{d\xi}{\xi \, dt} = \frac{1}{6\eta} \left( \frac{\phi^\alpha \mu^\beta (\lambda^2 - \lambda^4)}{1 - (2\lambda^2 + \lambda^4 - 3)/J_{\text{lim}}^\beta} \right),$$  \hspace{1cm} (A.5)
where $\eta$ is the viscosity of the dashpot. With (A.4) and (A.5), we solve for $\lambda(t)$ and $\zeta(t)$, once $V(t)$ and $P(t)$ are prescribed. The theoretical model is compared to the experimental data (Figure 2.2F). The actuator was subject to a voltage, which was applied suddenly at time zero, and was then held constant subsequently (Figure 2.2E). The initial vertical segment and the solid curve are theoretical predictions. In short time, the elastomer behaves as a purely elastic material, and expands instantaneously in response to the sudden jump in the applied voltage. Subsequently, the elastomer expands as a function of time due to viscoelastic creep. The fitting parameters used are $\mu^a = 26$ kPa, $\mu^b = 110$ kPa, $J_{\text{lim}}^a = 89$, $J_{\text{lim}}^b = 30$, and $\eta = 0.33$ MPas.

**A. 4 Effect of the Elasticity of the Electrolyte**

We show that the voltage-induced deformation is limited by electromechanical instability, and examine the effect of the electrolyte on the instability using equation (A.3). The electromechanical response of actuators varies with the levels of prestretch $\lambda_{\text{pre}}$ (Figure A.1). Here we use representative values of shear modulus for hydrogels ($\mu^B = 3$ kPa) and ion gels ($\mu^B = 400$ kPa). For the dielectric, the electric breakdown voltage $V_{\text{EB}} = E_{\text{EB}} h$, with a constant value of the electric breakdown field set to $E_{\text{EB}} = 200$ MV/m.

The hydrogel is less stiff, resulting in a larger actuation strain at the same applied voltage. At $\lambda_{\text{pre}} = 1$, the voltage-stretch curve of the actuator using the hydrogel shows a local maximum before intersecting with the electrical breakdown curve (plotted in red), and the actuator fails by electromechanical instability once the local maximum of the curve is reached. When $\lambda_{\text{pre}} = 2$, the voltage-stretch curve for the hydrogel electrolyte becomes monotonic. Consequently, the maximum actuation strain is limited by electric breakdown. By eliminating instability, the membrane is able to achieve large actuation strain. The transition from the occurrence of electromechanical instability to stable behavior occurs at $\lambda_{\text{pre}} \approx 1.93$. The ion gel is stiffer, so that
Figure A.1 Theoretical voltage-stretch curves of actuators with different levels of pre-stretch. The red curves correspond to the electric breakdown field of 200 MV/m. (A) The electrolyte is a hydrogel with a shear modulus of 3 kPa. (B) The electrolyte is an ion gel with a shear modulus of 400 kPa.
larger pre-stretches are required to avert instability; the transition occurs at $\lambda_{\text{pre}} \approx 3.24$. Even in the stable region, the achievable strain at electrical breakdown is lower for the stiffer electrodes. Therefore, the actuator may achieve large actuation strains for a small electrolyte modulus and a $\lambda_{\text{pre}}$ large enough to avert instability. Increasing the stiffness of the electrolyte of an actuator operating in the electromechanically stable regime may render it unstable.

**A. 5 Time Delay due to Resistance and Capacitance (RC Delay)**

In layered electrolytic and dielectric elastomers (Figure 2.1), the net capacitance $C$ of the circuit is given by

$$\frac{1}{C} = \frac{2}{C_{\text{EDL}}} + \frac{1}{C_{\text{D}}}.$$  \hspace{1cm} (A.6)

The capacitance of the EDL is proportional to the area of the electrode/electrolyte interface. The electrochemistry of this interface is expected to be similar to that of an interface between the electrode and an aqueous solution. The capacitance per unit area of the EDL is on the order $c_{\text{EDL}} \sim 10^{-1} \text{ F/m}^2$.\textsuperscript{79} The capacitance of the dielectric is proportional to the area of the dielectric. The capacitance per unit area of the dielectric is

$$c_{\text{D}} = \frac{\varepsilon}{H_{\text{D}}} \sim \frac{10^{-11} \text{ F/m}}{10^{-3} \text{ m}} = 10^{-8} \text{ F/m}^2,$$  \hspace{1cm} (A.7)

where $\varepsilon$ is the permittivity of the dielectric. So long as the area of the EDL is not excessively small compared to that of the dielectric, the capacitance of the EDL is much larger than that of the dielectric:

$$C_{\text{EDL}} \gg C_{\text{D}}.$$  \hspace{1cm} (A.8)

Consequently, the net capacitance of the circuit is dominated by the contribution of the dielectric, $C \approx C_{\text{D}}$. 

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The time delay due to the capacitance of the dielectric and the resistance of the electrolyte is

\[ \tau = RC. \]  \hspace{1cm} (A.9)

Consequently, the \( RC \) time constant for the layered electrolytic and dielectric elastomer under equal-biaxial stretch \( \lambda \) may be written as:

\[ \tau = \frac{2\varepsilon A_D \rho_{\text{electrolyte}}}{H_D H_{\text{electrolyte}}} \lambda^6, \]  \hspace{1cm} (A.10)

where \( \rho_{\text{electrolyte}} \) and \( H_{\text{electrolyte}} \) are the resistivity and thickness of the electrolyte, respectively. Thus, \( \tau \sim c_D A_D R \lambda^6 \), where \( R \) is the sheet resistance of the electrolyte. For representative values \( c_D = 10^{-8} \text{ F/m}^2 \), \( A_D = 10^{-2} \text{ m}^2 \), and \( R = 10^2 \text{ \Omega/sq} \), we estimate that \( \tau \sim 10^{-8} \text{ s} \) when \( \lambda = 1 \), and \( \tau \sim 10^{-6} \text{ s} \) when \( \lambda = 2 \). This estimate does not include the electrical resistance of the lines of hydrogels between the large pad of the hydrogels and the copper electrodes. If these lines are thin and long, they will dominate the net resistance, and increase the RC delay.

### A. 6 Fundamental Resonance due to Elasticity and Inertia

We estimate the resonant frequency of the actuator by the in-plane vibration of a thin sheet, area \( A \) and thickness \( H \). The mass of the sheet is \( m = \rho A H \), where \( \rho \) is the mass density. The effective stiffness of the sheet scales as \( k \sim YH \), where \( Y \) is the elastic modulus. The frequency of the fundamental mode of resonance is \( \omega = (k / m)^{0.5} \). When the actuator is subject to a cyclic voltage of a certain frequency of excitation \( \Omega \), the amplitude of vibration of the actuator is the same as that induced by the static voltage when \( \Omega \ll \omega \), and vanishes when \( \Omega \gg \omega \).\footnote{Thus, the fundamental resonance sets a time scale, \( \tau_{\text{inertia}} = 1 / \omega \), for the amplitude of actuation to vanish. This estimate gives \( \tau_{\text{inertia}} \sim (A_D \rho / Y)^{0.5} \). This estimate is consistent with the observed limiting frequency of actuation. The estimate is also consistent with previous observations for dielectric...}
elastomer actuators made of silicone coated with carbon grease,\textsuperscript{205} where the actuation speed was limited by the resonant frequency of approximately 1 kHz, or $\tau_{\text{inertia}} \approx 10^{-3}$ s for estimated values $A_D = 10^{-2}$ m$^2$, $\rho = 10^3$ kg/m$^3$ and $Y = 10^6$ N/m$^2$.

Incidentally, a similar estimate also applies to the fundamental resonance of a dielectric elastomer actuator deflecting out of plane, like the membrane of a drum. Because the membrane is pre-stretched substantially, the tension in the membrane scales with the elastic modulus as $T \sim YH$. The frequency of fundamental resonance scales as $\omega = (T / m)^{0.5}$ The fundamental mode of resonance sets a time scale, $\tau_{\text{inertia}} = 1 / \omega$, which again gives $\tau_{\text{inertia}} \sim (A_D \rho / Y)^{0.5}$. This estimate is close to an experimental observation of miniaturized diaphragm dielectric elastomer actuators,\textsuperscript{206} where the limiting frequency of actuation was approximately 1 kHz. Using representative values $A_D \sim 10^{-5}$ m$^2$, $\rho \sim 10^3$ kg/m$^3$ and $Y \sim 10^6$ N/m$^2$, we find that $\tau_{\text{inertia}} \approx 10^{-4}$ s.

A. 7 Ionic Liquids as Conductors for High-speed, Large-Strain Dielectric Elastomer Actuators.

Ionic liquids are nonvolatile electrolytes, and are being developed for applications including lithium-ion batteries, fuel cells and dye-sensitized solar cells.\textsuperscript{82} A large number of ionic liquids exists, and may be selected to suit specific applications. Furthermore, ionic liquids can be used as solvents to form ion gels, which are stretchable, transparent, ionic conductors.\textsuperscript{79} An example of using ionic liquids as conductors for dielectric elastomer actuators was given in a previous paper.\textsuperscript{207} By a combination of experiment and theory, here we demonstrate that ionic liquids can be used as conductors for high-speed, large-strain dielectric elastomer actuators.

We built an actuator using a commercially available ionic liquid 1-Decyl-3-methylimidazolium chloride, [C\textsubscript{10}MIM][Cl]. One layer of VHB 4910 was stretched to three times the initial radius and fixed to a circular rigid acrylic frame of diameter 12 cm. The ionic liquid was
Figure A.2 Performance of a dielectric elastomer actuator using an ionic liquid as the conductor. 
(A) Area strain of an actuator using the ionic liquid [C10MIM][Cl] as conductor as a function of applied voltage. (B) Area strain measured as a function of excitation frequency at an applied voltage of 4 kV.
painted to the two faces of the dielectric elastomer within circular regions of diameter 3 cm. On each face, a line of the ionic liquid was also painted from the circular region to the acrylic frame, where the line was connected to a copper electrode.

When a step voltage was applied, the actuator expanded gradually. The area strain at 20 s after the voltage is applied is plotted as a function of the magnitude of the voltage (Figure A.2A). When a cyclic voltage was applied, the steady area strain was recorded as a function of the frequency (Figure A.2B). Both characteristics are comparable to those observed for actuators using carbon grease as conductors.

The RC delay of the actuator using the ionic liquid as the conductor remains exceedingly small, so that the frequency of actuation is not limited by the electrical resistance of the ionic liquid, but by the mechanical inertia. The RC delay can be estimated using equation (A.10) derived above, \( \tau \sim c_D A_D R \). Assuming representative values for the thickness \( H \sim 10^{-3} \text{ m} \) and resistivity \( \rho \sim 10 \ \Omega\text{m} \) of the ionic liquid, we find that the sheet resistance is \( R = 10^4 \ \Omega/\text{sq} \). For representative values \( c_D = 10^{-7} \text{ F/m}^2 \) and \( A_D = 10^{-3} \text{ m}^2 \), we estimate that \( \tau \sim 10^{-6} \text{ s} \). Even though the sheet resistance of the ionic liquid is two orders of magnitudes higher than that of the ionic hydrogels, high-speed actuation is nevertheless readily achieved.

A. 8 Charge Leakage through VHB when Three Types of Conductors are Used

Leakage of electrical charge carriers through a dielectric is an important characteristic of devices operating at large electrical fields. For example, charge leakage decreases the efficiency of a device, and may lead to premature failure. Here we quantify charge leakage through VHB when three types of conductors are used: carbon grease, a hydrogel (2.74 M NaCl-containing polyacrylamide), and an ionic liquid ([C\(_{10}\)MIM][Cl]).
A test sample was placed in a glass tank, in which air was replaced with argon to minimize the influence of ambient charged species (Figure A.3A). Temperature of the argon atmosphere was held constant at 22 °C and the oxygen content was monitored to ensure consistent experimental conditions. An electric field mill (AlphaLab, Inc.; Ultra Stable Surface DC Volt Meter) monitored the electrical potential of a metal plate without direct contact to minimize charge loss into experimental equipment. The metal plate shared the electrical potential of one side of the test sample, while the other side was grounded. A HV source (Model 50/12, TREK) was connected to the metal plate to charge the test sample up to a voltage of 1000 V. We disconnected the HV voltage source and measured the decay of the voltage down to a level of 1000 V / e (~ 368 V). The geometry of the test samples was identical to the samples used in Figure 2.6E, except that only one layer of VHB was used here. Due to unavoidable, small variations in the dimensions of the handmade samples, we quantified differences in initial conditions by measuring the capacitance ($C_0$) immediately after fabrication.

The decays of voltage were recorded for samples using the three types of conductors, and each sample was tested three times (Figure A.3B-D). Some variations in the shape of the curve and the decay time were observed, but variations between different samples with the same type of electrode were on the same order as the observed differences between samples fabricated with different types of conductors.

The sample with the hydrogel as the conductor was retested after 24 h of storage time in a high-humidity environment (Figure A.3E). Surprisingly, the leakage process was slower compared to the initial result. The reason for the observation is unclear, but we observed that some water had evaporated from the hydrogel over night and thereby increased the concentration of salt.
Figure A.3 Charge leakage through a dielectric when conductors of several types are used. (A) An electric field mill is used for contactless measurement of the electrical potential of a metal plate which is connected to a test sample. (B)-(D) Experimental results on the decrease of electrical potential over time for electrodes based on carbon grease, ionic liquid and hydrogel. (E) Test results for the same hydrogel sample used in (D) after 24 h of storage time.
A. 9 High-Speed, Long-Distance Ionic Interconnects

In the actuator and loudspeaker, we connect the active regions of the devices to the copper electrodes using thin lines of ionic conductors. This design demonstrates that ionic conductors can be used as high-speed, stretchable and transparent interconnects. The ionic conductors have much higher electrical resistance than typical electronic conductors; for example, the resistivity of our hydrogels is \( \rho \sim 10^{-2} \, \Omega m \), and the resistivity of copper is \( \rho \sim 10^{-8} \, \Omega m \). How fast and how far can a signal propagate along the ionic interconnects? The answers depend on the resistivity of the conductor and the permittivity of the surrounding insulators, as well as on the geometry of the setup. To gain some insight, here we use an idealized model for the propagation of potentials along myelinated axons.\(^{208}\)

In this model, a conducting line is inside a dielectric sheath, which is surrounded by another conductor (Figure A.4). The outside conductor is grounded. The conducting line and the dielectric sheath are a resistor and a capacitor. As an electric current propagates along the resistor, part of the electric charge is deposited to the capacitor. Let \( x \) be the distance from one end along the length of the resistor. At time \( t \), let \( I(x,t) \) be the electric current along the length of the resistor, and \( Q(x,t) \) be the electric charge per unit length of the capacitor. The conservation of electric charge requires that

\[
\frac{\partial Q}{\partial t} + \frac{\partial I}{\partial x} = 0. \tag{A.11}
\]

Let \( V(x,t) \) be the electric potential in the resistor. Ohm’s law requires that

\[
\frac{\partial V}{\partial x} = -rI. \tag{A.12}
\]

where \( r \) is the resistance per unit length of the conducting line. Because the outside conductor is grounded, \( V \) is also the voltage across the thickness of the capacitor, so that
Figure A.4 Propagation of an electrical signal along the length of an interconnect. A conducting line is inside a dielectric sheath, which is surrounded by another conductor. The outside conductor is grounded.
\[ Q = cV. \]  
(A.13)

where \( c \) is the capacitance per unit length of the dielectric.

Combining (A.11)-(A.13) gives that

\[ \frac{\partial V}{\partial t} = \frac{1}{rc} \frac{\partial^2 V}{\partial x^2}. \]  
(A.14)

This is a diffusion equation, with the effective diffusivity

\[ D = \frac{1}{rc}. \]  
(A.15)

Consider a conducting line of circular cross section of radius \( a \) inside of a dielectric sheath of external radius \( b \). The resistance per unit length of the conducting line is

\[ r = \frac{\rho}{\pi a^2}. \]  
(A.16)

where \( \rho \) is the resistivity of the conductor. The capacitance per unit length of the dielectric sheath is

\[ c = \frac{2\pi \varepsilon}{\log (b/a)}. \]  
(A.17)

where \( \varepsilon \) is the permittivity of the dielectric.

In the following estimates, we drop factors of order unity. Using (A.15)-(A.17), we find that the effective diffusivity scales as

\[ D \sim a^2/\rho \varepsilon. \]  
(A.18)

The product \( \rho \varepsilon \) is a time scale specific to the materials, and is independent of the geometry of the setup. For a conductor of \( \rho \approx 10^{-2} \, \Omega \text{m} \), and a dielectric of permittivity \( \varepsilon \approx 10^{-11} \, \text{F/m} \), we obtain that \( \rho \varepsilon \approx 10^{-13} \, \text{s} \).

When a step voltage is suddenly applied at one end of the interconnect, the signal takes some time to reach the other end. For an interconnect of length \( L \), the delay time scales as

\[ \tau \sim \frac{L^2}{D}, \]  
namely,
\[ \tau \sim \rho \varepsilon (L/a)^2. \quad (A.19) \]

For an interconnect of radius \( a = 10^{-4} \) m and length \( L = 10^{-1} \) m, the delay time is \( \tau \sim 10^{-7} \) s.

When a sinusoidal voltage of frequency \( \omega \) is applied at one end of the interconnect, the amplitude of the voltage decays as the signal propagates along the interconnect. The decay length scales as \( L \sim (D / \omega)^{0.5} \), namely,

\[ L \sim \frac{a}{\sqrt{\rho \varepsilon \omega}}. \quad (A.20) \]

For an interconnect of radius \( a = 10^{-4} \) m and a signal of frequency \( \omega = 10^5 \) Hz, the decay length is \( L \sim 1 \) m.
Appendix B

Supporting Information for Chapter 3

B. 1 Calculation of the Acoustic Waves from the Measured Pressures

In both sections of the impedance tube plane waves travel in positive and negative x-direction (Figure B.1). These waves can be decomposed into their frequency components with radial frequency \( \omega \) and wavenumber \( k \) (\( k = \omega / c \), where \( c \) is the speed of sound). At each location \( x \) the pressure is the sum of the right, and left traveling waves:

\[
p_F = F_R e^{i(\omega t - kx)} + F_L e^{i(\omega t + kx)} \tag{B.1}
\]

\[
p_B = B_R e^{i(\omega t - kx)} + B_L e^{i(\omega t + kx)} \tag{B.2}
\]

Microphones measure the pressures at four locations \((x_j, j = 1 - 4)\) as a function of time. Short term Fourier transformation can decompose the measured pressure signals of the microphones into their frequency components \((p_j, j = 1 - 4)\). With equations (B.1), and (B.2) one can calculate the (complex) amplitudes of the traveling waves in both sections of the tube:

\[
F_R = \frac{i(p_1 e^{ikx_2} - p_2 e^{ikx_1})}{2 \sin(k(x_1 - x_2))} \tag{B.3}
\]

\[
F_L = \frac{i(p_2 e^{-ikx_2} - p_1 e^{-ikx_1})}{2 \sin(k(x_1 - x_2))} \tag{B.4}
\]

\[
B_R = \frac{i(p_3 e^{ikx_4} - p_4 e^{ikx_3})}{2 \sin(k(x_3 - x_4))} \tag{B.5}
\]

\[
B_L = \frac{i(p_4 e^{ikx_4} - p_3 e^{ikx_3})}{2 \sin(k(x_3 - x_4))} \tag{B.6}
\]
Figure B.1 Acoustic waves inside the impedance tube. In the front section, the acoustic wave $F_R$ travels in positive $x$-direction and the acoustic wave $F_L$ travels in negative $x$-direction. In the back section the acoustic wave $B_R$ travels in positive $x$-direction and the acoustic wave $B_L$ travels in negative $x$-direction. Microphones record the pressures $p_1$, $p_2$, $p_3$, and $p_4$ at the four locations $x_1$, $x_2$, $x_3$, and $x_4$ as a function of time. With short time Fourier transformation, the signals can be decomposed into their frequency components (radial frequency $\omega$, wave number $k$).
B. 2 Derivation of the Transfer Matrix

The classical transfer matrix does not directly relate the pressures of the incident and reflected waves on the left \((x = 0)\) and right \((x = d)\) sides of a sample, but the total pressures \((p_0, p_d)\), and velocities \((v_0, v_d)\) at the two interfaces:\(^98\)

\[
\begin{pmatrix} p_0 \\ v_0 \end{pmatrix} = \begin{pmatrix} Q_{11} & Q_{12} \\ Q_{21} & Q_{22} \end{pmatrix} \begin{pmatrix} p_d \\ v_d \end{pmatrix}
\]

(B.7)

\[
p_0 = F_R + F_L
\]

(B.8)

\[
p_d = B_R e^{ikd} + B_L e^{ikd}
\]

(B.9)

\[
v_0 = \frac{F_R - F_L}{\rho_0 c}
\]

(B.10)

\[
v_d = \frac{B_R e^{ikd} - B_L e^{ikd}}{\rho_0 c}
\]

(B.11)

In equations (B.9), and (B.11), \(d\) is the thickness of the sample. It is not straightforward to define \(d\) for the transparent membrane, because it is thicker in the region covered by the ionic electrodes \((\sim 2\, \text{mm})\) than in the region that is not covered \((\sim 0.1\, \text{mm})\). Because the smallest wavelength in our experiments \((\lambda \sim 0.34\, \text{m at} \; f = 1000\, \text{Hz})\) is much larger than the maximum thickness of the loudspeaker \((d \approx 2\, \text{mm})\), we neglect the exponential factors in equations (B.9), and (B.11) (the error in phase angle due to this simplification is \(\sim 2^\circ\)). With this simplification we derive the relationship between, \(T_{ij}\) and \(Q_{ij}\):

\[
\begin{pmatrix} T_{11} \\ T_{21} \\ T_{22} \end{pmatrix} = \frac{1}{2} \begin{pmatrix} Q_{11} + \rho_0 c Q_{21} + \frac{Q_{12}}{\rho_0 c} + Q_{22} & Q_{11} + \rho_0 c Q_{21} - \frac{Q_{12}}{\rho_0 c} - Q_{22} \\ Q_{11} - \rho_0 c Q_{21} + \frac{Q_{12}}{\rho_0 c} - Q_{22} & Q_{11} - \rho_0 c Q_{21} - \frac{Q_{12}}{\rho_0 c} + Q_{22} \end{pmatrix}
\]

(B.12)
For a symmetric, reciprocal sample $Q_{ij}$ has only two independent components:

$$Q_{11} = Q_{22}$$  \hspace{1cm} (B.13)

$$Q_{11}Q_{22} - Q_{12}Q_{21} = 1$$  \hspace{1cm} (B.14)

By plugging equations (B.13), and (B.14) into equation (B.12) we obtained the analogous relationships for the components of $T_{ij}$:

$$T_{21} = -T_{12}$$  \hspace{1cm} (B.15)

$$T_{11}T_{22} - T_{12}T_{21} = 1$$  \hspace{1cm} (B.16)

**B. 3 Characterization of the Acoustic Properties of the Transparent Membrane**

Figure B.2 shows that the acoustic spectrum of the left traveling wave in the front section of the impedance tube ($F_L$) does not have a subharmonic during a frequency sweep on the transparent membrane from 150 Hz to 1000 Hz at amplitude $V_a = 645$ V, and bias voltage $V_b = 9.0$ kV. Figure B.3 shows the fundamental components of the right traveling, and left traveling waves in both sections of the impedance tube during the characterization of the transparent membrane.

**B. 4 Mode of Oscillation of the Transparent Membrane**

The Maxwell stress (equation (3.1)) causes a reduction in thickness of the dielectric membrane. A sinusoidal excitation voltage on the membrane causes a periodic change in thickness. In this oscillation mode, the air at the two interfaces between the membrane and the air moves in opposite directions ($v_0 = -v_d$). From equations (B.10), and (B.11) follows for an anechoic tube ($F_R = B_L = 0$) that $\hat{F}_L / \hat{B}_R = 1$ (i.e., the phase angle of $\hat{F}_L / \hat{B}_R$ is 0°). When the membrane oscillates out-of-plane like a drumhead, the velocities at the two interfaces are equal ($v_0 = v_d$). From
Figure B.2 Acoustic spectrum of $F_1$ for $V_b = 9.0$ kV during a sinusoidal frequency sweep from $f = 150$ Hz to $f = 1000$ Hz with amplitude $V_a = 645$ V. The spectrum does not contain a subharmonic.
Figure B.3 Characterization of the transparent membrane. Amplitudes of the fundamental component of the measured waves travelling in the front ($F_R, F_L$) and back ($B_R, B_L$) sections of the impedance tube during the sinusoidal frequency sweep on (A) the coil loudspeaker, and (B) the transparent membrane. The equidistant peaks indicate resonances of the impedance tube.
equations (B.10), and (B.11) follows for an anechoic tube that $\frac{F_L}{F_R} = -1$ (i.e., the phase angle of $\frac{F_L}{F_R}$ is 180$^\circ$). It is also possible, that both oscillation modes contribute equally to sound generation. In this case the phase angle of $\frac{F_L}{F_R}$ is neither 0$^\circ$ nor 180$^\circ$. In our experiments, the phase angle was approximately 180$^\circ$ (Figure 3.5), so we conclude that the dominant oscillation mode is out-of-plane.

**B. 5 Active Noise Cancellation Experiment**

The incident sound wave on the transparent membrane $F_R$ is composed of the sound generated by the coil loudspeaker, and the reflection of $F_L$ at the coil loudspeaker. The sound wave $F_L$ is therefore modified by exciting the membrane (equation (3.4)). During the noise cancellation experiment, we adjust the excitation signal on the coil loudspeaker to keep the amplitude and phase of $F_R$ constant. The amplitude of the sound generated by the coil loudspeaker is linearly proportional to the excitation amplitude, and the wave reflected from the coil loudspeaker is proportional to the incident sound wave

$$F_R = S_L V_L + T_L F_L. \quad (B.17)$$

This equation is equivalent to equation (3.4) for the transparent membrane. The parameters $S_L$, and $T_L$ can be determined from the same data that we used to determine the properties of the transparent membrane.

Assuming that the membrane cancels a prescribed incident noise $F_R$ perfectly (i.e., $B_R = B_L = 0$), we combine equations (3.4), (3.7), and (B.17) to obtain the excitation signal on the coil loudspeaker to keep $F_R$ constant:

$$V_L = \left(1 - \frac{T_L S_2}{S_1} \right) \frac{F_R}{S_L} \quad (B.18)$$
With equation (3.7), we calculated the excitation signal for the transparent membrane to cancel a noise wave, which consists of a sinusoidal frequency sweep from 150 Hz, and 1000 Hz of 80 dB amplitude. To ensure linearity of the membrane, we capped the amplitude of the excitation signal at 645 V. Using the calculated excitation signal for the transparent membrane, and equation (B.18) we calculated the excitation signal for the coil loudspeaker to generate a constant incident acoustic wave of amplitude 80 dB for all frequencies (Figure B.4A, and B). Because the membrane did not completely cancel $F_R$ (an assumption in the derivation of equation (B.18)), and because of inaccuracies in the parameters in equation (B.18), $F_R$ deviated slightly from the desired value during the experiment (Figure B.5C and D).

Figure B.5 shows the measured amplitudes of the acoustic spectrum during the noise cancellation experiment, and Figure B.6 a comparison of the sound transmission loss calculated with equation (3.2) \(i.e.,\) uncorrected), and equation (3.9) \(i.e.,\) corrected).
Figure B.4 Active noise cancellation experiment. (A) Excitation signal applied to the transparent membrane. To ensure linearity of the membrane, the amplitude of the signal was limited to 645 V. (B) Excitation signal applied to the coil loudspeaker (C) Comparison of the amplitude of the fundamental component of the measured spectrum of $F_R$ with the theoretically predicted amplitude. (D) Comparison of the phase angle of the fundamental component of the measured spectrum of $F_R$ with the theoretically predicted phase angle.
Figure B.5 Fundamental components of the acoustic waves measured during the active noise cancellation experiment.
Figure B.6 Comparison between the corrected $STL_a$ (equation (3.9)) and the experimentally obtained $STL$ (equation (3.2)).
Appendix C

Supporting Information for Chapter 5
Figure C.1 Geometry of valves used for characterization. The dimensions are in mm. We measured the switching pressures as a function of $H$ for $H = 0.5$ mm to $H = 4.50$ mm with $\theta = 180^\circ$, and as a function of $\theta$ from $\theta = 140^\circ$ to $\theta = 180^\circ$ at $H = 3$ mm. All other dimensions remained constant.
Figure C.2 The measured critical pressures as a function of the shear modulus of the material. The membranes were fabricated using E-30 (Ecoflex 30, shear modulus $\mu = 26.2$ kPa), DS-10 (Dragon Skin 10NV, $\mu = 72.0$ kPa), and DS-30 (Dragon Skin 30, $\mu = 262$ kPa), with $\theta = 180^\circ$, and $H = 2.0$ mm, 3.0 mm, 3.5 mm, and 4.0 mm. (A) Measured critical pressures on the $\Delta P_2 - \Delta P_1$ plane. Larger critical pressures are observed with stiffer material. (B) Measured critical pressures on the $\Delta P_2 - \Delta P_1$ plane, after dividing $\Delta P_1$, and $\Delta P_2$ by $\mu$. Values for the same geometry collapse to a single point after correction, indicating that $\Delta P_1$, and $\Delta P_2$ are linearly proportional to the shear modulus of the material.
Figure C.3 Nominal stress ($s$) as a function of the ratio between the extended length and the initial length (i.e., the stretch, $\lambda$), for Dragon Skin 30, Dragon Skin 10NV, and Ecoflex 30 in an uniaxial tension test. Uniaxial samples of the three materials (length 100 mm, width 5 mm, thickness 2 mm) with an Instron 5966 (500 N load cell) were stretched (50 mm/min extension rate) up to $\lambda = 1.5$. We obtained the shear moduli of the materials (Dragon Skin 30: $\mu = 261$ kPa; Dragon Skin 10: $\mu = 72$ kPa; Ecoflex 30: $\mu = 26.2$ kPa;) by fitting the incompressible Neo-Hookean model for uniaxial deformation ($s = \mu (\lambda - \lambda^{-2})$) to the experimentally obtained curves.
Figure C.4 Photographs of the starfish gripper with an integrated valve, which does not have a top chamber, (A) before contacting the ball, and (B) after contacting the ball.
Figure C.5 Measurement of pressure as a function of time, for the soft oscillator in Figure 5.4, with $P_S = 50 \text{ kPa}$, $V = 50 \text{ ml}$ with 2 cm long tubing of 0.79 mm inner diameter introduced between the pressure source, and the oscillator.
Figure C.6 Measurement of pressure as a function of time, for the soft oscillator in Figure 5.4, with $P_S = 11$ kPa, $V = 150$ ml during initial characterization, and after $10^5$ cycles.
Figure C.7 Alternative designs of the valve. (A) A pressure limiting valve. (B) A pressure release valve.
Figure C.8 Mold designs for the devices used for characterizing $\Delta P_1$, and $\Delta P_2$ (A) Assembly of mold for the center piece (the membrane and surrounding chamber wall). The mold is closed after the pe-polymer is filled in. (B) Mold for the end pieces. The dimensions of the molds can be found in Figure C.20, and Table C.1.
Figure C.9 Assembly of the device used for characterizing $\Delta P_1$ and $\Delta P_2$. (A) The gray areas mark areas where glue was applied. The dashed line indicates the orientation of the membrane. (B) Photograph of the assembled device. The scalebar corresponds to 1 cm.
Figure C.10 Molds for the tubing inside the chamber of the valves. (A) Assembly of the mold to fabricate six tubes, simultaneously. (B) Mold for the connector (between the tubing and the conical tip). (C) Mold for the conical tip. The dimensions of the molds can be found in Figure C.21.
Figure C.11 Assembly of the tubing used inside the chambers of the valve. (A) The gray areas mark the locations where glue was applied. Alignment of the tubes with the connector is facilitated by keeping syringe needles in the tubes during the assembly. (B) Photograph of the assembled tubing.
Figure C.12 Molds for the transparent valve. (A) Assembly of molds for the walls of the valve. The mold is closed after the pre-polymer is filled in. (B) Assembly of the mold for the membrane of the valve. The mold is closed after the pre-polymer is filled in. (C) Assembly of the mold for the top and bottom end pieces. The dimensions of the molds can be found in Figure C.22.
Figure C.13 Assembly of the transparent valve. The gray areas mark locations where glue was applied.
Figure C.14 Molds for the automatic gripper. (A) Assembly of the molds for the bending actuators. (B) Assembly of the mold for the valve. The mold is closed after the pre-polymer is filled in. (C) Assembly of the mold for the contact sensor. The mold is closed after the pre-polymer is filled in. (D) Assembly of the mold for the top end piece of the valve. The dimensions of the molds can be found in Figure C.23 to Figure C.25.
Figure C.15 Assembly of the starfish gripper. (A) Assembly of the gripper. (B) Gluing of the contact sensor to the gripper. The gray areas mark locations where glue was applied. Alignment of tubing, and vertical channels with the connector is facilitated by keeping syringe needles in the tubes during assembly.
Figure C.16 Molds for the oscillator. (A) Assembly of the molds for the center of the valve. The mold is closed after the pre-polymer is filled in. (B) Assembly of the mold for the top end piece of the valve. (C) Assembly of the mold for the bottom end piece of the valve. The dimensions of the molds can be found in Figure C.26.
Figure C.17 Assembly of the oscillator (A) The gray areas mark locations where glue was applied. Alignment of the vertical channel in the wall with the top end piece is facilitated by keeping a syringe needle inside the vertical channel during gluing. (B) Photograph of the assembled oscillator. The scalebar corresponds to 2cm.
Figure C.18 Molds for the earthworm (A) Assembly of the mold for the bellows actuator. The mold is closed after the pre-polymer is filled in. (B) Mold for the retaining spring (C) Assembly of the mold for rear end piece. The mold is closed after the pre-polymer is filled in. (D) Mold for the front end piece. (E) Assembly of the mold for the valve. The mold is closed after the pre-polymer is filled in. (F) Assembly of the mold for the top and bottom end pieces of valve. The dimensions of the molds can be found in Figure C.27 to Figure C.29.
Figure C.19 Assembly of the earthworm. Grey areas mark where glue was applied. (A) Assembly of the rear end piece of the valve. (B) Gluing of the bellows actuator to both end pieces. (C) Gluing of the retaining spring to the bellows actuator.
Figure C.20 Dimensions of the molds for the devices used for characterizing $\Delta P_1$, and $\Delta P_2$. All dimensions are in mm.
Table C.1 Parameters of the molds for the devices used for characterizing $\Delta P_1$, and $\Delta P_2$.

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Figure C.21 Dimensions of the molds for the tubing. All dimensions are in mm.
Figure C.22 Dimensions of the molds for the transparent valve. All dimensions are in mm.
Figure C.23 Dimensions of the molds for the gripper 1. All dimensions are in mm.
Figure C.24 Dimensions of the molds for the gripper 2. All dimensions are in mm.
Figure C.25 Dimensions of the molds for the gripper 3. All dimensions are in mm.
Figure C.26 Dimensions of the molds for the oscillator. All dimensions are in mm.
Figure C.27 Dimensions of the molds for the earthworm 1. All dimensions are in mm.
Figure C.28 Dimensions of the molds for the earthworm 2. All dimensions are in mm.
Figure C.29 Dimensions of the molds for the earthworm 3. All dimensions are in mm.