Biaxially compliant metal thin film electrodes for dielectric elastomer actuators

Low, Sze Hsien

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**NANYANG
TECHNOLOGICAL
UNIVERSITY**

**BIAXIALLY COMPLIANT
METAL THIN FILM ELECTRODES
FOR
DIELECTRIC ELASTOMER ACTUATORS**

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METAL THIN FILM ELECTRODES
FOR
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LOW SZE HSIEN

SCHOOL OF MECHANICAL AND AEROSPACE ENGINEERING

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Abstract

Dielectric elastomer actuators (DEAs) are soft actuators that are capable of large actuated strains. Being variable area capacitors, DEAs require electrodes that are stretchable enough to be able to remain conductive as they follow the area expansions of the elastomer, while being sufficiently compliant, such that they do not impede the elastomer's deformations. Metal thin films are highly conductive, but tend to be very stiff and to fracture at low strains of just a few percent. The work in this thesis shows that it is possible to make metal thin film electrodes almost as biaxially stretchable and biaxially compliant as conventional carbon-based electrodes, with the added benefits of being more conductive and having the ability to self-clear. Two mechanisms were used: an inherently brittle metal thin film that resulted in a network of zigzagging micro-cracks and a biaxially compressed thin film that buckled to form crumpled metal thin films. The metal thin films used were predominantly electrolessly deposited silver (ELD silver), as electroless deposition is a relatively simple and inexpensive method that can be carried out using relatively safe chemicals, under room temperature and pressure conditions.

The first mechanism utilises the spontaneously formed network of zigzagging micro-cracks in ELD silver thin films to improve its stretchability, such that the electrode remained conductive up to 33% uniaxial strain and 11% radial strain. In addition, the micro-cracked ELD silver thin films appear to have a lower stiffening effect on the DEA's elastomer-electrode composite, as compared to sputtered silver films of the same thickness. As a result, DEAs using ELD silver thin film electrodes managed to attain actuated area strains of up to 50%. Such actuated strains can be considered as high when compared to DEAs using sputtered gold and silver films that generally have maximum actuated area strains of less than 10%.

The second mechanism makes use of biaxially buckled or crumpled metal thin films to enhance its biaxial stretchability and compliance. The stretchability of crumpled ELD silver thin films improved to 400% uniaxial strain and 110% radial strain. Also, the apparent stiffness of the thin film was reduced, which resulted in a lowered stiffening effect on the electrode-elastomer composite, comparable to that of graphite powder. Due to this combination of improved stretchability and lowered stiffening effect, in all in-plane directions, high actuated area strains of 128% was attained by DEAs using crumpled ELD

silver thin film electrodes. It has also been shown that this method of crumpling metal thin films to improve actuated strains is not exclusive to ELD silver thin films.

There are a couple of added benefits that make metal thin film electrodes particularly attractive for use in DEAs. Firstly, metal thin film electrodes are able to self-clear, which makes its DEAs less susceptible to premature failures; this applies to both the micro-cracked and the crumpled versions of the metal thin films. Secondly, the sheet resistances of metal thin films are much lower than that of carbon-based electrodes, which tends to be more than $10\text{k}\Omega/\square$. This is especially important when the elastomer starts to become conductive at high fields, as the low sheet resistance of metal thin films can help to reduce the voltage drop to the dielectric elastomer. It has been shown that crumpled ELD silver thin film electrodes are able to retain a sheet resistance of under $60\Omega/\square$ when uniaxially strained by 100% for one hundred cycles.

We have found that it is possible to model a DEA's electromechanical response based purely on the stiffness of its electrodes and dielectric elastomer. However, this is on the condition that the dielectric elastomer remains a perfect dielectric. In practice, the elastomer becomes conductive at high electric fields, resulting in a closed circuit. Due to the increase in leakage current, high resistance electrodes would cause a large voltage drop across it, resulting in a lower voltage across the elastomer, thereby decreasing the DEA's actuated strains. This shows the importance of a DEA's electrodes having to be both highly compliant and stretchable.

The metal thin film electrodes presented here are both biaxially stretchable and compliant, to strains comparable to that of conventional carbon-based electrodes. Also, they have the additional benefits of low conductivity and the ability to self-clear. As such, this makes them highly suitable for use as electrodes in high-strain DEA applications.

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1 Introduction

Dielectric elastomer actuators (DEAs) are soft actuators that can be controlled electrically to produce motions similar to that of natural muscles. In addition to being able to produce large strains and stresses, DEAs are lightweight and have high specific elastic energy densities, relatively fast response speeds and high peak strain rates [7, 133, 5, 36]. As such, DEAs have been used as artificial muscles for robots that mimic mammalian movements, such as limb rotations [56, 25, 90, 132, 140] and facial expressions [90, 107, 54, 37]. For DEAs to be used as artificial muscles, an important requirement is that it should have high strains of at least 40% in order to match the strain of natural muscles.

The basic unit of a DEA, as shown in Figure 1.1, comprises of a dielectric elastomer film that is sandwiched between two compliant electrodes. When a voltage is applied across the DEA, electrostatic forces squeeze the thickness of the dielectric elastomer, causing it to reduce in thickness and consequently to expand in area. Electrostatic forces result from opposing electrical charges gathering on either surface of the elastomer, by means of the pair of electrodes. This means that the elastomer should be electrically insulating, such that it does not allow the electrical charges to pass through. On the other hand, the electrodes should be electrically conducting, so as to be able to efficiently transport electrical charges to and from the elastomer's surfaces. As the elastomer expands in area, the compliant electrodes are expected to remain electrically conductive while following the elastomer's planar movements, so that charges can be transported to the entire expanded area of the elastomer. In other words, the electrodes should be stretchable.

The magnitude of the DEA's deformation for a given electrostatic force depends on the mechanical properties of both its elastomer and compliant electrodes. In order to be able to attain large actuated strains, the elastomer should be easily deformed. An example of such an elastomer is silicone, which has a relatively low Young's modulus of about 1MPa. Similarly, as the electrodes are bonded to the elastomer, they should have a minimal stiffening effect on the elastomer, i.e. the electrodes should be compliant, such that they do not inhibit the elastomer's deformations. For example, while thick rigid metal plates (with a Young's modulus in the Giga-Pascal range) would clamp the elastomer in place and prevent any deformation, carbon grease of negligible stiffness would offer little mechanical resistance, thereby allowing the elastomer to deform freely.

Figure 1.1. The basic unit of a DEA in its un-activated state (voltage off) and activated state (voltage on). When unconstrained, the DEA undergoes an equi-biaxial deformation.

Commonly used conductive powders and greases [133, 130, 131, 94, 149] do not epitomise the perfect compliant electrode for use in DEAs. These electrodes adhere poorly to the elastomer's surface and are hence not stable over repeated actuations. For example, greases may dry up or get rubbed off, while powders may dislodge. This results in a reduction, or even a loss, in electrical conductivity over time [147]. In addition, conductive particles tend to have relatively high sheet resistances, in the kilo to mega - ohm range, which would result in electrical losses. This is especially so for larger scale dielectric elastomer applications, as Joule heating is proportional to the resistance of the electrodes. Despite the great importance that compliant electrodes have on the resulting electromechanical behaviour of DEAs, there are few studies in literature that focus on the development of compliant DEA electrodes. In contrast, most studies tend to focus on either the DEA's actuator configuration [94, 149, 34, 35, 55, 137] or on its dielectric elastomer [61, 91, 130, 131, 133, 153, 76]. In fact, most of the theoretical models that have been developed assume that the compliant electrodes are perfectly stretchable and compliant, thereby having a negligible effect on the bulk of the dielectric elastomer [187, 127, 88, 85]. However, such an ideal DEA electrode does not yet exist in practice. In order for DEAs to be commercially well-received, it would be pertinent that issues regarding its electrodes be solved together with the other aspects of the actuator.

Metals have long been recognised for their ability to conduct electricity and have therefore been the traditional material for electrical conductors. The main advantages of using metal thin films electrodes over conductive particles are that they generally adhere well to their substrates, have a low sheet resistance and can be easily deposited by reliable methods such as sputtering [130] and evaporation [135]. Apart from that, thin metal films have the advantage of having the ability to self-clear, or better known as self-healing in the metallised plastic capacitor industry. As shown in Figure 1.2, self-clearing is the phenomena wherein localised breakdowns, due to defects such as pinholes and flaws in the dielectric layer, cause the film at the defect site to puncture and the thin metal layer to vaporize [159, 160]. This causes the defect site to become insulating, thereby isolating it from the rest of the active capacitor. In this manner, the DEA is less likely to fail prematurely via short circuits, even allowing higher actuation voltages to be reached.

Figure 1.2. Schematic showing the cross-sections of a DEA with a pinhole defect, which is highly exaggerated here. DEAs using electrodes without self-clearing capabilities would result in a short circuit (left) while DEAs using electrodes with self-clearing capabilities would have a insulated region that forms to isolate the pinhole defect (right) .

In spite of the abovementioned benefits, compliant electrodes made out of metal thin films are seldom used when compared to the widespread use of conductive powders and greases. This is because metal thin films are very stiff and have low fracture strains: Metals have a Young's modulus in the giga-Pascal range, which is more than four orders higher than the Young's modulus of elastomers. As such, it has a great stiffening effect on the elastomer, which inhibits large deformations. Also, freestanding metal thin films have low fracture

strains of just a few percent [71], meaning that it would not be able to follow the large expansions of the elastomer's area, resulting in the electrodes not having a full-coverage of the elastomer.

1.1 Objectives

In this work, biaxially compliant metal thin film electrodes are developed for use in dielectric elastomer actuators. In order to be as attractive for use in DEAs as conductive powders and greases, the metal thin film electrodes need to perform as well or even better in the following aspects. First and foremost, the metal thin film electrodes need to be biaxially compliant, such that they provide little mechanical resistance to the planar deformations of the elastomer. Secondly and just as importantly, the electrodes should be highly stretchable in both biaxial directions, such that they are able to retain a full-coverage of the elastomer as it expands in area. Thirdly, in order to surpass conductive powders and greases, the metal thin film electrodes should have high conductivity and stability, in both the electrical and structural sense. Lastly, the metal thin films should have the ability to self-clear, which can help to make the DEA more robust.

1.2 Organisation of thesis

Two types of metal thin film electrodes are studied in this work: micro-cracked silver thin film electrodes and biaxially crumpled metal thin film electrodes. The metal thin films used are predominantly silver, fabricated by means of an electroless deposition (ELD) method. The mechanical and electrical properties of these two types of metal thin film electrodes are studied and related to its use in membrane DEA actuators.

Chapter 2 presents the literature review on artificial muscle technologies, with an emphasis on DEAs, and on stretchable electrodes and their potential for use in DEAs.

Chapter 3 deals with the modelling of DEAs. First, the basics of non-linear elasticity theory are covered. Following that, a simple model is used to show why electrodes that allow biaxial deformations are preferred for larger actuated strains. Next, DEAs are modelled using both a constant-force prestress and a fixed-frame prestress to check the effects of prestretch and electrode stiffness on actuated strains. The parameters that are to be used in the experiments were determined through these models.

Chapter 4 presents details of the materials used, as well as the experimental details for the DEA electromechanical activations and electrode characterisation tests.

Chapter 5 studies the use of electrolessly deposited silver thin films as electrodes for DEAs. The fabrication procedure of ELD silver thin films is first covered. Following that, the network of micro-cracks in the ELD silver thin films is studied. The electrodes are then characterised for their stretchability and stiffening effect. Finally, the ELD silver thin film electrodes were used in prestretched membrane DEAs and their electromechanical response observed.

Chapter 6 determines the feasibility of using biaxially buckled metal thin film electrodes in DEAs. The background of wrinkling is covered and a simple structural analysis is used to determine the apparent stiffness of buckled thin films and therefore the stiffening effect that they would have on the DEA elastomer. Experimentally-obtained stress-stretch behaviours of the electrode-elastomer composites were fitted to the Ogden hyperelastic model and an effective electrode determined. Using the material parameters obtained, the models are used to predict actuated strains of a prestretched membrane DEA.

Chapter 7 experimentally studies the use of biaxially crumpled metal thin films as electrodes for DEAs. The fabrication of crumpled metal thin film electrodes is first explained, and its morphological changes with compression studied. The electrodes' stretchability and stiffening effect is then determined. Following that, the crumpled metal thin film electrodes are used in prestretched membrane DEAs and their electromechanical response observed.

Chapter 8 concludes the present work and discusses future works.

2 Literature review of electroactive polymer actuators and compliant electrodes

Artificial muscles are soft and bendable actuators that have a similar performance to natural muscles, such as being able to produce irregular linear motions. This is in contrast to electromagnetic motors, which are most suitable for high-speed continuous motions, such as the rotation of a fan's blades. When it comes to precise start-stop linear motions, such as medical drug metering piston pumps, electromagnetic motors are lacking: bulky and heavy gear systems often have to be used to step down the speed and to convert rotary motions into linear ones. This not only increases cost and bulk, but also decreases the efficiency of the actuations. Newer technologies have emerged to suit irregular, linear motions, of which the most prominent would be piezoelectric ceramics. Piezoelectric ceramics are efficient, fast, high-force actuators that have been used in applications such as inkjet printers [30] and ultrasonic teeth scaling instruments [31]. However, the small strains of piezoelectric ceramics, of generally less than 1% [186, 9], limits its applications.

Like piezoelectric ceramics, electroactive polymers (EAPs) deform when subjected to an electrical stimuli, but with much larger actuated strains. Of the EAP technologies, DEAs appear to have the most potential for applications requiring large actuated strains. With such large deformations of the elastomer, which can exceed 100%, the demands on the DEA's electrodes become more challenging: the compliant electrodes have to be highly stretchable and have a small stiffening effect on the elastomer. Here, we explore alternative materials for the compliant electrodes used in DEA.

2.1 Electroactive polymers

EAPs have been slated to become the artificial muscles of the future [7], as they have high fracture toughness, large actuation strains and inherent vibration damping [124]. In this section, we review just some of the available EAP technologies. For more information on EAPs, the reader is referred to the excellent reviews done by Madden et al. [114], Mirfakhrai et al. [117], Carpi et al. [36], Anderson et al. [5] and Brochu and Pei [28], as well as the references within them.

EAPs can be divided into two types: ionic and electronic. Ionic EAPs are electrochemical polymers that change in shape due to the electrically driven motion of ions. They can be activated by low voltages but generally require a controlled wet environment, which adds to the actuator's bulk and weight. Ionic EAPs include ionic polymer-metal composites (IPMCs), carbon nanotubes (CNTs) and conducting polymers. On the other hand, electronic EAPs change in shape in response to an applied electric field. Electronic EAPs can be operated in air, but operate using high voltages, which might require bulky and expensive circuitry. In addition, electronic EAPs have relatively fast response times and can hold induced displacements under activation of a direct current voltage [9]. Electronic EAPs include relaxor ferroelectric polymers, liquid crystal elastomers (LCEs) and dielectric elastomer actuators (DEAs).

2.1.1 Ionic EAPs

Ionic polymer-metal composites (IPMCs), as its name suggests, are composites comprising of an ionic polymer sandwiched by a pair of metal electrodes. The ionic polymer, such as Nafion and Flemion, has to be able to selectively exchange either cations or anions with its own incipient ions [118]. In order to apply the metal electrodes, usually comprising either percolated gold or platinum, the ionic polymer is treated with the ionic salt solution of the metal, and then chemically reduced. When a voltage is applied, the clustered single-charged ions and solvent in the ionic polymer migrate towards the oppositely-charged electrode, causing that region to swell. Consequently, the region near the other electrode shrinks. This redistribution of charges, and its solvent, within the ionic polymer results in differential swelling and shrinkage, which in turn causes the IPMC to bend [151], as depicted in Figure 2.1. IPMCs can attain actuated strains of more than 10% at driving voltages of less than 7V, with actuation stresses of up to 30MPa [83, 120, 152]. The shortfalls of IPMCs is that it tends to exhibit relaxation, if a constant current is not supplied to it, as the solvent tends to flow back due to a pressure gradient. Also, a gradual solvent leakage from the polymer reduces its performance [117]. IPMCs have been used as muscles for soft robotics applications, such as for gripping, flying, swimming and walking [152].

Figure 2.1. Schematic showing the redistribution of charges in an ionic polymer, due to an imposed electric field, wherein cations and water migrate towards the negatively-charged electrode [114].

Carbon nanotubes (CNTs) actuators are cylindrical nanostructures of carbon, wherein an individual single-walled nanotube has a very large Young's modulus of 640GPa and a tensile strength of 37GPa [14], which is lowered when CNT bundles, held together by weak van der Waals forces, are used instead [13]. When a CNT, or a set of parallel-acting CNTs, is immersed in an electrolyte together with a counter electrode, which as shown in Figure 2.2 is another CNT, and a voltage applied, opposite charges are injected into the two electrodes. These charges are in turn balanced by the ions in the electrolyte, which cause a rearrangement

of the charges in the CNTs. At low charge injections, it is this rearrangement of charges within the CNT that results in actuations. While at high charge injections, like charges in each CNT develop repulsive electrostatic forces, resulting in the elongation of the CNT. CNTs have low actuated strains of less than 2% [53] at low driving voltages of about 1V, with high actuation pressures of 26MPa. However, CNTs have very poor electromechanical coupling, of less than 1%, due to the high stiffness of individual CNTs and its low bulk modulus of 15GPa [158]. Although most CNT actuators require an electrolyte, a solid-state CNT actuator comprising carbon nanotube aerogel sheets has been developed by Aliev et al. [3] that can generate 220% elongation.

Figure 2.2. Schematic showing the charge injections in the two nanotubes, as a result of an applied voltage [13].

Conducting polymers, such as polypyrrole and polyaniline, can be electrochemically doped with ions and accompanying solvent, so as to make them increase in volume [12]. The ions and solvents are likely to have been inserted in between the polymer chains as expansions are predominantly in the direction perpendicular to the length of the polymer chains, as shown in Figure 2.3 [64]. Conducting polymers have actuated strains of typically between 2 and 20%, at low driving voltage of about 2V, with high actuated pressures of more than 100MPa [155]. Due to the resistance between the polymer and dopants, the time required for the ions to migrate is long, resulting in a low actuation speed of much less than 1% per second [172].

Also, the electromechanical coupling and operating efficiency is very low, at about 1% [114]. Applications for conducting polymers include valves [134] and micro-catheters [148].

Figure 2.3. Schematic showing the expansion of polypyrrole. As it is oxidised, it loses electrons, thereby allowing anions to enter [114].

2.1.2 Electronic EAPs

Electrostrictive relaxor ferroelectric polymers are polymers that change shape when an electric field is applied, as a result of large molecular conformational changes that occur during a phase change [9], as shown in Figure 2.4. In the case of ferroelectric polymers, such as poly(vinylidene fluoride-trifluoroethylene), or P(VDF-TrFE), the beta phase is stable at room temperature while the alpha phase is not. However, by modifying the ferroelectric polymer via the addition of imperfections [43, 70], its Curie point can be lowered to below room temperature and the phase change energy barriers can be reduced. Doing so results in a relaxor ferroelectric polymer that is stable in the nonpolar paraelectric alpha phase during operation. By applying an electric field to the relaxor ferroelectric polymer in its alpha phase, it will change into the beta phase, which is accompanied by an expansion along the length of the polymer chain and a contraction perpendicular to the polymer chain, as shown in Figure 2.4. Relaxor ferroelectric polymers can attain actuated strains of 10% [70] and high actuation pressures of up to 45MPa [176]. However, the high driving voltages in the kilo-volt range and electric fields of about $150\text{V}\mu\text{m}^{-1}$ are required.

Figure 2.4. PVDF in the nonpolar alpha phase and the polar beta phase, looking (top) along the length of the chains and (bottom) perpendicular to the chains. A change from the alpha to beta phase results in an expansion along the length of the polymer chain and a contraction perpendicular to the polymer chain [117].

Electrostrictive liquid crystal elastomers make use of the ability of liquid crystals to change phase, and therefore orientation, to induce stresses in the polymer's backbone, which in turn leads to actuated strains. Phase changes in liquid crystal molecules, or mesogens, can result from either electric fields or heat. As shown in Figure 2.5, the mesogens are attached to the flexible polymer backbone, which allows the strains of the mesogen's orientational changes to be transferred to the polymer backbone. The field-induced phase change mechanism is similar to that described for ferroelectric elastomers. Actuated strains of about 4% at relatively low electric fields of $1.5\text{V}\mu\text{m}^{-1}$ have been attained [103].

Figure 2.5. Liquid crystal molecules or mesogens (ovals II and III) are attached to the flexible polymer backbone (rectangle I) [103].

2.2 Dielectric elastomer actuators

Dielectric elastomer actuators (DEAs) are elastomers that deform, due to electrostatic forces, when an electric field is applied, as shown in Figure 1.1. Of all the EAPs reviewed, it is able to generate the largest strains, ranging from tens of percent to hundreds of percent. Apart from large strains, actuation pressure is moderate at several mega-Pascals and has good electromechanical coupling and efficiency of up to 90%. The major disadvantage of DEAs is that it requires very high electric fields of around $100\text{V}\mu\text{m}^{-1}$ and high driving voltages in excess of 1kV, which might require circuitry that adds to the weight and bulk of the DEA. However, this can be mitigated as recent developments of small, commercial, high-voltage direct current converters (Q series, Emco High Voltage Corporation) can supply up to 5kV with just a size of about 2cm^3 and up to 10kV with a size of about 10cm^3 . Also, in order for large actuated strains to be realised, the dielectric elastomer often has to be prestretched, which also adds weight and bulk to the DEA. Table 1 compares natural muscles with the artificial muscle technologies that have been discussed. It can be seen that DEAs appear to be able to mimic natural mammalian muscles the closest, especially because of its high strains which seem to be lacking in all the other technologies, save the thermal liquid crystal elastomer, which has a slow response speed.

Table 1. Comparison of actuator technologies

Actuator	Maximum strain (%)	Maximum pressure (MPa)	Maximum efficiency (%)	Relative speed	References
Mammalian skeletal muscles	>40	0.35	35	medium	[130, 90]
Dielectric elastomers (acrylic)	380	7.2	60-80	medium	[130, 90]
Dielectric elastomer (silicone)	63	3.0	90	fast	[130, 90]
Ionic polymer metal composites	10	30	2.9	fast	[152, 114]
Carbon nanotubes	2	26	22	fast	[117, 114]
Conductive polymers (polypyrrole)	12.4	22	5	slow	[63]
Relaxor ferroelectric polymers	<7	45	80	fast	[114]
Liquid crystal elastomers	45 (thermal) 4 (field)	0.45 (thermal)	<5 (thermal)	slow (thermal)	[114]
Piezoelectric ceramics (PZT)	0.2	110	90	fast	[130, 90]
Shape memory alloys (TiNi)	>5	>200	<10	slow	[130, 90]

2.2.1 Actuator configurations and applications for DEAs

The basic unit of a DEA can be made in an assortment of shapes and sizes, and combined into different configurations that would help harness its large actuated strains. Most of the DEA configurations were based on the designs originally developed by SRI, which bear a strong resemblance to those used for piezoceramics: these include the stack, extender, roll,

tube, framed, unimorph, bimorph, diaphragm, bowtie and spider, most of which are shown in Figure 2.6.

Figure 2.6: Actuator Configurations [90] These were developed by SRI and are similar to configurations used by piezoceramics. A popular configuration that is not included in this diagram is the stack actuator.

The diaphragm, spider and bowtie configurations are able to harness the large area strains of membrane DEAs by mechanically converting them into usable linear strokes. In the case of the bowtie, an expansion in the DEA's area straightens the hinges to cause actuations in the vertical direction. As for the spider configuration, expansions in the DEA's area result in perpendicular linear strokes, in the direction of its thickness. The diaphragm, and its variant called the cone, makes use of area expansions that buckle out of plane to produce linear strokes in the direction perpendicular to its area.

The stack or multi-layer actuator comprises multiple layers of the basic dielectric actuator unit stacked on top of each other, as shown in Figure 2.7. Stacks are generally used to amplify the actuator's stroke, which constitutes the sum of the thickness deformation in all the layers. It can be quite difficult to reliably fabrication of these stacks layer by layer. To ease the manufacturing process, a long electrode can be fabricated instead, and then folded

up (like in the folds of an accordion) to form a folded stack [35, 40], as shown in Figure 2.7. Stacks also require electrodes carrying opposite charges to come into close proximity. As such, a passive border typically surrounds the electrodes to prevent a short circuit. In addition, these passive borders also help to bond the layers of DEAs together, since the electrodes are not tacky, so as to be able to better handle tensile forces that will pull the layers apart. However, the passive borders are inactive and tend to hinder the in-plane area strains of the active elastomer, thereby reducing axial strains in the thickness direction [149, 77]. Another challenge in the fabrication of stacks is the inability to connect all the electrodes carrying like-charges, so as to be able to electromechanically activate all the layers. By leaving out a layer, it becomes passive and does not contribute to actuation strains. Despite the challenges involved, DEA stacks have been successfully developed and a 16mm diameter DEA stack has been reported to be able to attain a 10% strain while lifting a load of 20 N [93].

Figure 2.7: A DEA stack that amplifies thickness deformations, (left) fabricated one layer at a time [93] and fabricated as a long monolithic strip and then folded to form a stack [35].

A roll is a stack of two or more large sheets of dielectric film, coated with compliant electrodes, that is rolled into a cylinder, as shown in Figure 2.8. The core of the cylinder may contain a spring to give a spring roll, or remain empty to give a tube. The actuation of a roll is usually either a bending motion or an extension of its length. In a spring roll, the dielectric

elastomer is often prestretched biaxially, with a prestretch that is higher along the circumference of the roll. While the end caps help to maintain the prestretch in the circumferential direction, the spring helps to keep the elastomer prestretched along the length of the roll. This is in contrast to the stack, wherein maintaining a prestretch in the elastomer would be a very challenging task. Axial strains of up to 35% could be obtained by such a spring roll [55, 92, 185]. By patterning the electrodes on a roll, spring rolls can be made to bend by up to 60 degrees to produce a walking motion [129, 128].

Figure 2.8: Roll used as artificial muscles, by Kovacs et al. (left) A coil spring core was used to provide a mechanical stress, and to retain the prestretch in both the x and y directions. (right) Completed roll actuators [55]

Kovacs et al. have indicated that the main problem with this design was the use of high prestretch of more than 400% in the dielectric elastomer. The high prestretch caused the inner films of the roll to be highly compressed by the outer ones and also brought about the need for a stiff supporting structure to be incorporated in the roll. Without the core support, instabilities at the film-structure interface would bring about stress-relaxation, which in turn causes deterioration in the actuator performance. This coil spring support structure, however, resulted in only an average of 5% of the roll actuator's cross-section area being composed of active dielectric elastomer film. Reducing prestretch, however, is not an option when using VHB 4910 as the dielectric material, as high prestretch is necessary for obtaining large actuated strains and stresses. In addition, the actuators have been reported to have poor durability, and it had been attributed to the combination of a high prestretch and the bulky and heavy supporting structure [94]. Kovacs et al. have proposed that the roll can be improved by fabricating a coreless, prestretch-free roll that is made of IPN (Interpenetrating Polymer Network) post-processed VHB 4910 films [55], which are inherently stretched.

Core-free tubular rolls that are not prestretched, as shown in Figure 2.9, have been developed by Danfoss PolyPower [163, 170]. Its electrode is a sputtered thin metal layer that is corrugated, which allows only unidirectional motions. Also, as the thin metal electrodes are quite stiff, the roll is able to stand freely. However, the maximum strain for such rolls is low, at less than 2.5%, perhaps partly due to the lack of prestretch and partly due to the metal electrodes, which might be too stiff to allow large deformations of the elastomer.

Figure 2.9. Core-free tubular actuator using corrugated sputtered silver thin film electrodes [163]

As the electromechanical performances of DEAs resemble natural muscles quite closely, it has been used in several applications that aim to mimic mammalian and insect motions. Examples include an arm wrestling robot using spring rolls [56], a hopping robot for search and rescue missions, using cone actuators [25], flapping-wing robots using bowtie actuators, inchworm robots for navigating confined spaces, using rolls, and insect-like walking robots named “Flex” [90, 132] and “MERbot” [140] that use spring rolls. DEAs have also been used to mimic facial expressions, such as mouth movements, using bowtie actuators [90] and eyeball rotations, using inflated DEAs in an agonist-antagonist configuration [107], folded stack [54, 37] and buckling actuators [37].

2.3 Dielectric elastomers

The properties of the dielectric elastomer used will affect the performance of the DEA. The elastomer should ideally be a perfect insulator, have a high relative permittivity, high

breakdown field strength, low stiffness modulus, high elongation at break and display little viscoelastic behaviour. Through the years, commercial materials that have stood out are polyacrylate elastomers (VHB™ adhesive tape series) and silicone elastomers (PDMS, polydimethylsiloxane). Out of all the elastomers used, VHB elastomers have been a popular choice in earlier research works as they enable DEAs to attain large actuated strains. However, as the development of DEAs move out of the labs towards commercialisation, more groups have chosen to work with silicone elastomers.

Although the actuated strains obtained by DEAs using silicone elastomers are not as large, silicone elastomers are less viscoelastic as compared to VHB elastomers. This helps improve the mechanical response speed of the actuator and reduces losses [10]. Apart from that, the large actuated strains attained by DEAs using VHB elastomers require a large mechanical prestretch, which might be hard to impose in some configurations. Due to stress relaxation, the performance of such highly prestretched DEAs might even be compromised over time. On the other hand, at low prestretch values, silicones generally generate larger strains than VHB elastomers, hence making them more suitable for certain configurations, such as stacks and tubes. In addition, silicone elastomers can also operate within a wide temperature range.

Despite the trend towards the use of silicone elastomers in DEAs, in this work, we have chosen to use the VHB polyacrylate elastomer as we need to utilise the large elongations that they can attain prior to rupture.

2.4 Compliant electrodes for DEAs

Electrode-covered elastomers in DEAs can expand by more than four times during actuations [80, 106] and therefore require compliant electrodes that are able to remain conductive even when highly stretched to such extents. A DEA's electrodes are used as a medium for transporting charges between the power source and the surfaces of the elastomer. Therefore, a disruption in the flow of charges would result in the whole of, or parts of, the DEA becoming inactive, resulting in smaller actuated forces and therefore strains. Equally important is the electrode's mechanical stiffness. In order to not impede large deformations of the soft elastomer, the compliant electrodes should ideally have a negligible stiffening effect on the elastomer.

The dielectric elastomer will undergo different types of deformations depending on the actuator configuration of the DEA: the most common types being uniaxial, biaxial and pure shear deformations, as illustrated in Figure 3.6. For biaxial planar actuations, such as that undergone by the DEA in Figure 1.1, the electrodes need to be stretchable in both planar directions. This is in contrast to say a spring, which is stretchable in only one direction. Of course, along with biaxial stretchability, the electrodes should also be biaxially compliant, such that they do not impede elastomer expansion in both planar directions.

Commonly used conductive powders and greases, as shown in Figure 2.10a and b respectively, [133, 130, 131, 94, 149] are not ideal compliant electrodes for DEAs as they have relatively high sheet resistances and poor electrical and mechanical stability. In order for DEAs to be commercially well-received, issues regarding its electrodes need to be solved.

Figure 2.10. Conductive carbon particles in a) powder form b) suspended in grease and c) embedded in an elastomer matrix [147].

Research activity in the field of stretchable electronics is the most relatable with regards to the compliant electrodes of DEAs. Although the flexible and stretchable electrodes required in stretchable electronics is not the equivalent of compliant electrodes for DEAs, fabrication methods and designs can be borrowed and improved on. Similar to compliant electrodes for DEAs, one of the primary concerns of stretchable electrodes for stretchable electronics is the strain to which they can be stretched, whilst remaining electrically conductive. However, as stretchable electrodes are subjected to external stresses and do not need to aid actuations,

electrode stiffness is usually not a design consideration. In the following section, a brief survey of stretchable, and possibly compliant, electrodes is carried out. A summary of the different types of electrodes used for DEAs is also shown in Table 2. For further information, the reader is referred to the comprehensive review done by Rosset and Shea [147].

Table 2. Types of electrodes used in DEAs and in stretchable electronics

Electrodes		Carbon-based	Metal-based
Particulate	Dry powder	Graphite powder [77, 149, 133] Carbon black powder [94, 39] Carbon nanotubes [183]	-
	Grease	Carbon grease [131, 39]	Silver grease [75, 161]
	Impregnated in elastomer	Carbon black in silicone [40, 108]	Gold-ion implantation [144-146] Silver nanoparticles [74] Platinum salt reduction [164]
Thin-film	Un-patterned	-	Thick (>200nm) inherently ductile copper film [104, 111] Thin inherently brittle copper film [101, 98] Sputtered gold (<100nm) [133, 136]
	Patterned in-plane	-	Zigzag – gold [89] Spiral – gold [135, 136] Meander – gold [29, 59, 58]
	Patterned out-of-plane	Crumpled graphene [184]	Corrugated – silver [18, 163, 20] Corrugated (buckled) – gold [167] Corrugated (buckled) – copper [171]

2.4.1 Impregnated conductive particles

Conductive particles can be impregnated in an elastomer matrix to form a conductive rubber, as shown in Figure 2.10c. Doing so helps to prevent the ablation of the particles [74, 164, 146, 130, 40], thereby alleviating issues regarding their mechanical stability. However, such electrodes generally have high electrical resistances, typically above the range of $k\Omega/\square$ for carbon-based electrodes. A lower electrical resistance can be attained by impregnating more conductive particles, but this leads to an increase in electrode stiffness [146, 133]. A variant of this concept is the use of a filtered cathode vacuum arc implantation method to implant metal nano-clusters, such as gold, into the top 20-30nm of an elastomer film [144-146, 154]. By doing so, the effective electrodes constitute just the part of the elastomer that had been modified by the implanted metal nano-clusters. These effective electrodes have a sheet resistance of about $1k\Omega/\square$ and can be uniaxially stretched by up to 175% [146]. The Young's modulus of the effective electrode has been calculated to be between 0.2 – 5GPa, depending on the proportion of metal clusters in the elastomer. The maximum actuated strain attainable by diaphragm DEAs using such metal ion implanted electrodes was relatively high, at an equivalent area strain of approximately 40% [146]. While promising, ion implantation is expensive [8], as opposed to the sputtering or evaporation of metal thin films.

2.4.2 Metal thin films

Metals have long been recognised for their ability to conduct electricity and have therefore been the traditional material for electrical conductors. The main advantages of using metal thin films electrodes over conductive particles are that they generally adhere well to their substrates, have a low sheet resistance and can be easily deposited by reliable methods such as sputtering [130] and evaporation [135]. Apart from that, thin metal films have the advantage of having the ability to self-clear, as shown in Figure 1.2, which results in a DEA that is less likely to fail prematurely via short circuits.

However, plain metal thin films cannot be considered as compliant electrodes. This is because metal thin films are very stiff and have low fracture strains: Metals have a Young's modulus in the Giga-Pascal range, which is more than four orders higher than the Young's modulus of elastomers. As such, it has a great stiffening effect on the elastomer, which inhibits large deformations. Also, freestanding metal thin films have low fracture strains of

just a few percent [71], meaning that it would not be able to follow the large expansions of the elastomer's area, resulting in the electrodes not having a full-coverage of the elastomer.

The stretchability of metal thin films can be improved by two main methods: the first is to utilise the inherent material behaviour of metal thin films (ductile versus brittle), while the second method is to purposefully pattern the metal thin films into forms that are more flexible.

In the first method, the metal thin films need to be well adhered to an elastomer substrate [60, 98, 97]. When metal thin films that are inherently ductile [104, 178, 105, 112, 111] are strained, multiple sites will undergo local thinning to form pre-rupture necks, while the cracks that result are short and isolated, as shown in Figure 2.11a. However, in order to attain such inherent ductility, the metal film needs to be sufficiently thick ($>200\text{nm}$ Cu), while the substrate needs to be sufficiently stiff ($>300\text{MPa}$) [112]. In addition, the adhesion between the metal film and the substrate has to be very good. Although highly stretchable by up to 60% uniaxial strain, this mechanism of ductile deformations appears to be unsuitable for use in DEA electrodes due to the high stiffness involved. In contrast, by using a metal thin film that is inherently brittle, micro-cracks will form in it, as shown in Figure 2.11b, [98, 101, 112, 62]. Its stretchability of up to more than 30% uniaxial strain has been attributed to the out-of-plane twisting and deflection of the micro-cracked network on an extremely compliant substrate when strained, which results in small actual deformations of the metal film itself [98]. As such, micro-cracked metal films can be said to be stretchable electrodes that have been naturally patterned under the right material conditions.

Figure 2.11. Micrographs of (left) 500 nm thick copper thin films exhibiting cracks formed by ductile failure and (right) 50 nm thick copper thin films exhibiting cracks formed by brittle failure [111].

As for the second method, metal thin films can be purposefully patterned in order to improve its stretchability. For example, metal thin films have been patterned into meanders and spirals [89, 135, 136], or tortuously patterned and then encapsulated by elastomers [59, 69, 29] as shown in Figure 2.12, to enable linear strains of up to 200% [69]. However, the trade-off of such electrode patterning is its reduced area coverage, which will lead to lower actuated forces.

Figure 2.12. Tortuously patterned metal thin films [59].

For better electrode-area coverage, it might be preferable to make use of electrodes that have been patterned in the out-of-plane direction. For example, continuous silver thin films have been sputtered onto un-stretched silicone substrates with wavy surfaces, as shown in Figure 2.9, to obtain electrodes that are mechanically stretchable by up to 80% linear strain [19],

while its DEAs can actuate by up to 15% linear strain [20]. Alternatively, corrugated conductive films can also be obtained via buckling: conductive films are deposited on a uniaxially stretched elastomer, which was later allowed to relax. Such electrodes can typically be stretched by up to 100% uniaxial strain [58, 156, 167, 143, 82], but can reach 300% uniaxial strain [171]. The large uniaxial strains could be sustained as the undulated film flattened out when stretched, thereby maintaining its conductivity [58]. However, most of these corrugated stretchable electrodes are only uniaxially stretchable because they are very stiff in the transverse direction [99]. As deformation in the transverse direction is constrained, the applications of such electrodes are more limited than that of stretchable electrodes that are biaxially stretchable.

Biaxially stretchable buckled metal thin films can generally be attained by making use of thermal means to expand the elastomer before the deposition of the metal thin film. When the elastomer cools down to room temperature, the metal thin film buckles into biaxial wrinkles [72, 181, 182, 22]. However, thermal expansion only strains the elastomer's area by a few percent and therefore limits the buckled electrode's stretchability. There exists a novel photo-patterning method, wherein a platinum salts embedded in an elastomer are reduced to form a platinum thin film on the elastomer's surface. Due to the submersion of the platinum salt-elastomer composite in an aqueous reducing solution, in situ swelling strains are induced. However, upon removal of the swelling strains, the platinum thin films are axially compressed and buckle into out of plane folds. As such, these low resistance ($<80\Omega/\square$) folded platinum thin film electrodes can be stretched by up to 150% linear strain [164]. However, in order to attain such stretchability and low sheet resistance, the resultant platinum thin films had to be about 700nm thick and therefore might be too stiff to be used in DEAs. In addition, such a method limits the dielectric elastomer to only photo-sensitive elastomers.

3 On the modelling of DEAs with appreciable electrode stiffness

Electrode stiffness has an impact on a DEA's actuated strain performance. Compliant electrodes, such as conductive greases, often have negligible stiffness. On the other hand, an elastomeric electrode with conductive fillers has a stiffness similar to that of the elastomer, and may therefore impede large deformations of the elastomer. Bozlar et al. have studied the effects of stiffness, of elastomeric electrodes, on a DEA subjected to a constant-force prestretch [23]. The constant-forces, which act to radially prestretch a DEA, can be exerted by dead-weights, as shown in Figure 3.1, and act on the circumferences of both the elastomer membrane and the electrodes. As the thickness of the DEA decreases when under Maxwell stress activation, the constant radial forces induce increasing axial stresses with increasing actuated strains in the DEA.

Figure 3.1. Schematic of a DEA prestretched by constant-forces that are supplied by dead weights.

In practice, it is easier and more common to prestretch a DEA by making use of a fixed rigid frame, as compared to using a system of pulleys and dead-weights. To do so, an elastomer membrane is prestretched and then affixed to a rigid frame, as shown in Figure 3.2.

Following that, electrodes are applied to the centre of both sides of the prestretched elastomer membrane, while leaving the bordering area of elastomer bare. In this setup, the DEA comprises of two regions: an active region and a passive region. The active region is the part of the elastomer membrane coated with electrodes while the passive region is the bare annular-shaped elastomer that borders the active region.

Figure 3.2. Schematic of a DEA prestretched by affixing the deformed elastomer to a rigid frame.

The overall outer diameter of this type of DEA is constant as it is bounded by a rigid frame. When un-activated, the active region is subjected to the same tension as the passive region, therefore having the same thickness. When activated by Maxwell stresses, the active region expands in area and reduces in thickness. On the other hand, the passive region undergoes stress relaxation and consequently reduces in area. Previously, Koh et al. [88] have modelled such a DEA, with an active and a passive region, but without taking electrode stiffness into account. In our model, we take into account the electrodes' stiffness and show the effect of electrode stiffness on the DEA's actuated strain performance.

In this chapter, we start by first reviewing the basics of nonlinear elasticity and hyperelastic constitutive models, which are useful for describing the large deformations in elastomers.

Next, we show that electrodes should be biaxially compliant (so as to allow biaxial deformations of the elastomer) in order to attain larger area stretches for a given applied electrostatic pressure. Following that, the model utilised by Bozlar et al. is revisited, wherein DEAs using electrodes with a finite stiffness are radially prestretched by constant-forces. Finally, we develop a model to simulate the actuated strain performance of DEAs that have electrodes of appreciable stiffness and that are prestretched by a circular rigid frame. There are many different types of elastomers that have been used for DEAs. Here, we have opted to use the popular VHB elastomer for all our models as it is highly deformable.

3.1 Theoretical background

An elastomer in a DEA tends to undergo large, nonlinear elastic deformations, due to prestretch and/or electromechanical activation. As such, assumptions made in linear elasticity, i.e. that strain is a linear function of displacement and that stress is a linear function of strain, do not hold. In view of that, this section covers the aspects of nonlinear elasticity [65, 122] that are required for the DEA modelling covered in the subsequent sections. In the modelling of DEAs, it is assumed that the elastomer is isotropic, incompressible and perfectly elastic, such that time-dependent effects are negligible.

3.1.1 Deformations

When dealing with large deformations, the deformed state of a body (current configuration at time t) is significantly different from its undeformed state (reference configuration at time $t = 0$). The reference configuration uses Lagrangian coordinates (X, Y, Z) , which are time-independent, while the current configuration uses Eulerian coordinates (x, y, z) , which transforms with changes in configuration at current time t . In order to determine relative deformations in a body, Eulerian coordinates can be expressed as a function of Lagrangian coordinates.

Figure 3.3 shows an elastomer that deforms from its initial state (reference configuration) to a deformed state (current configuration) at time t . In the reference configuration, a line element at point $(X, 0, 0)$ is aligned along the x -axis and has an initial length of dX . At time t , in the current configuration, the line element displaces to point $(x, 0, 0)$, and deforms to a length of dx . The deformation of this line element is given by the ratio of its deformed length to its

original length, or $\frac{dx}{dX}$. More generally, local deformations at a point (X,Y,Z) in the reference configuration can be characterised by the deformation gradient tensor **A**

$$\mathbf{A} = \begin{bmatrix} \frac{dx}{dX} & \frac{dx}{dY} & \frac{dx}{dZ} \\ \frac{dy}{dX} & \frac{dy}{dY} & \frac{dy}{dZ} \\ \frac{dz}{dX} & \frac{dz}{dY} & \frac{dz}{dZ} \end{bmatrix} \quad (3.1)$$

Figure 3.3. Equi-biaxial / axis-symmetric deformation of an elastomer.

The eigenvalues of the deformation gradient tensor **A** give the principal stretches ($\lambda_1, \lambda_2, \lambda_3$) of point (X,Y,Z). If the principal directions are aligned with the coordinate systems used, then the principal stretches can be denoted by $\lambda_1 = \frac{dx}{dX}$, $\lambda_2 = \frac{dy}{dY}$ and $\lambda_3 = \frac{dz}{dZ}$ respectively. In addition, there will be no shear in the principal plane, which makes **A** a diagonal matrix

$$\mathbf{A} = \begin{bmatrix} \lambda_1 & 0 & 0 \\ 0 & \lambda_2 & 0 \\ 0 & 0 & \lambda_3 \end{bmatrix} \quad (3.2)$$

The deformation gradient tensor can be further simplified to just two principal stretches, λ_1 and λ_2 , when the material is incompressible, such as is the case of most elastomers. To do so, we make use of J, which is a measure of dilatation (the variation of a small volume relative to

reference configuration), that is equal to the determinant of deformation gradient tensor $\mathbf{A}$. For incompressible materials that undergo isochoric (constant volume) deformations, J is equal to one, such that

$$J = \det \begin{bmatrix} \lambda_1 & 0 & 0 \\ 0 & \lambda_2 & 0 \\ 0 & 0 & \lambda_3 \end{bmatrix} = \lambda_1 \lambda_2 \lambda_3 = 1 \quad (3.3)$$

Therefore, for an incompressible material, the stretch in the third direction can always be represented by the stretches in the other two directions, such that

$$\lambda_3 = \frac{1}{\lambda_1 \lambda_2} \quad (3.4)$$

Take the case of biaxial deformations, as shown in Figure 3.4a, wherein stretches are imposed in the two planar directions λ_1 and λ_2 . This results in a compression of its thickness, such that the stretch in the thickness direction λ_3 is equal to $\lambda_1^{-1} \lambda_2^{-1}$. This relationship holds regardless of the boundary conditions imposed. For example, when applied to a uniaxial deformation, as shown in Figure 3.4b, wherein a stretch is imposed in one direction λ_1 , the stretch in the thickness direction λ_3 is also equal to $\lambda_1^{-1} \lambda_2^{-1}$. However, as the contraction in the two free directions are equal, such that $\lambda_2 = \lambda_3$, the stretch in the thickness direction λ_3 can be further simplified to be equal to $\lambda_1^{-0.5}$.

Figure 3.4. Relationship between deformations in the three principal directions for incompressible materials.

A DEA usually has its dielectric elastomer membrane subjected to an equi-biaxial prestretch. When activated, Maxwell stresses also induce an equi-biaxial deformation that is both large and homogeneous, as is shown in Figure 3.3. An equi-biaxial deformation has equal stretches in the planar directions ($\lambda_1 = \lambda_2$), meaning that the stretch in the thickness direction λ_3 can be described by just λ_1 , such that $\lambda_3 = \lambda_1^{-2}$. A homogeneous deformation indicates that all the points in the elastomer in the x, y - plane have the same deformation gradient $\mathbf{A}$. This indicates that the stretches in the entire body is the same as the stretches at each local point. As such, λ_1 can be simply represented by the ratio of the elastomer's overall deformed radius r to its original radius R , or $\frac{r}{R}$.

3.1.2 Stresses

When dealing with nonlinear elasticity, one has to balance stresses in the current configuration, while constitutive relations, which relate stress and strain, must be imposed relative to the reference configuration of the material [68]. For elastomers, the theory of hyperelasticity is an expedient method of constructing the constitutive relations. A hyperelastic material, also known as a Green elastic material, has a strain energy function W that measures the stored energy in a material due to deformation. The strain energy function is a scalar function of deformation gradient tensor $\mathbf{A}$ and satisfies the relationship $\mathbf{S} = \frac{\partial W(\mathbf{A})}{\partial \mathbf{A}}$, wherein $\mathbf{S}$ is the nominal stress tensor, whose transpose is known as the first Piola-Kirchhoff stress tensor. By taking into consideration just the principal stresses, the principal nominal stresses s_i are given by

$$s_i = \frac{\partial W(\lambda_1, \lambda_2, \lambda_3)}{\partial \lambda_i} - \frac{p}{\lambda_i} \quad i = 1, 2, 3 \quad (3.5)$$

wherein p is the arbitrary hydrostatic pressure.

This nominal stress tensor $\mathbf{S}$ is in turn related to the true stress tensor $\mathbf{T}$, also known as the Cauchy stress tensor, by $\mathbf{T} = J^{-1} \mathbf{S} \mathbf{A}^T$. By taking into consideration just the principal stresses, the principal true stresses, or the Cauchy stresses, σ_i are given by

$$\sigma_i = s_i \lambda_i = \lambda_i \frac{\partial W(\lambda_1, \lambda_2, \lambda_3)}{\partial \lambda_i} - p \quad i = 1, 2, 3 \quad (3.6)$$

For any material that is incompressible, such as are most elastomers, the relationship between principal stresses and stretches can be further simplified by making use of equation (3.4). By doing so, λ_3 does not have to be considered in the strain energy function, such that

$$\tilde{W}(\lambda_1, \lambda_2) = W(\lambda_1, \lambda_2, \lambda_1^{-1} \lambda_2^{-1}) \quad (3.7)$$

By using $\tilde{W}$ and by eliminating the scalar p from equation (3.6),

$$\sigma_i - \sigma_3 = \lambda_i \frac{\partial \tilde{W}(\lambda_1, \lambda_2)}{\partial \lambda_i} \quad i = 1, 2 \quad (3.8)$$

In the modelling of DEAs, the prestretching of the elastomer results in in-plane stresses σ_1 and σ_2 , while electrostatic forces result in stresses in the z -direction σ_3 , as shown in Figure 3.5. In the prestretched state, the DEA is not yet activated and hence σ_3 is equal to 0. By substituting $\sigma_3 = 0$ into equation (3.8), one can determine the in-plane stresses σ_1 and σ_2 acting on the elastomer, as result of deformations in the elastomer resulting from the mechanical prestretch λ_1 and λ_2 .

Figure 3.5. Stresses in the different states in a DEA.

When the DEA is activated, electrostatic pressure σ_e compresses the thickness of the elastomer, such that

$$\sigma_3 = -\sigma_e = -\epsilon_o \epsilon_r E^2 = -\epsilon_o \epsilon_r \left(\frac{V}{Z} \right)^2 = -\epsilon_o \epsilon_r \left(\frac{V}{Z \lambda_3} \right)^2 \quad (3.9)$$

wherein ϵ_o is the permittivity of free space, ϵ_r the relative permittivity, E the electric field and V the voltage across a thickness z , of original thickness Z and thickness stretch λ_3 . Again, by substituting equation (3.9) into equation (3.8), one can either determine the in-plane stresses if deformations in the elastomer are known, or determine the deformations in the elastomer if the in-plane stresses are known.

3.1.3 Constitutive models of rubber elasticity

In the modelling of DEAs, the main requirement of the constitutive model is that it should be able to accurately predict the behaviour of the elastomer in the DEA configuration, based on material parameters extracted from simple deformation experiments; this is because the strain energy function of a constitutive model depends on the deformed elastomer's principal stretches, as presented in section 3.1.2. The most convenient way to determine material parameters is to perform uniaxial extensions on the elastomer and then fit a constitutive

model to that stress-stretch data. While there are many constitutive models that are able to fit the uniaxial stretch data, not many are able to predict the elastomer's biaxial deformation behaviour based on material parameters obtained from just the uniaxial deformation data. Predictions can be improved by fitting the model to data from multiple deformation experiments: for example, fitting to both uniaxial and biaxial extension data [123]. However, as the experimental setup for biaxial extension is not as easily available as that for uniaxial extension, it might be more challenging to obtain biaxial extension data. In order to make use of the model, it should also be simple enough to be amenable to mathematical analysis [121]. In addition, it would also be useful if the constants in the constitutive model had a significant relationship with the material properties of the material. For example, the Young's modulus in Hooke's law gives a quantitative measure of stiffness.

Generally, constitutive models for rubber elasticity comprise of statistical and phenomenological theories. The statistical theory is based upon the assumption that elastomers comprise of a network of long and flexible cross-linked chains of molecules [162]. When an elastomer is stretched, its randomly oriented molecular chains become more ordered, which leads to a reduction in its configurational entropy. The work done on the elastomer is in turn directly related to this change in entropy, which can be described by mathematically-simple Gaussian statistics [179]. However, as breaking strain is approached at large deformations, the finite extensibility of chain stretch has to be accounted for. This can be done by assuming a network structure that obeys Langevin chain statistics, such as used by models such as the Arruda and Boyce eight-chain model [6]. Although such statistical models, based on the theory of finite chain extensibility, are able to predict the behaviour of rubber up to large strains, they tend to be mathematically complex [179].

While statistical theory aims to explain an elastomer's mechanical properties based on its underlying microscopic structure, phenomenological theory is a continuum mechanics treatment that does not take into account molecular concepts: The phenomenological theory merely aims to mathematically represent the behaviour of the elastomer. One of the earliest phenomenological theories for rubber elasticity was developed by Mooney [119]; its strain energy function, in terms of principal stretches, is given by

$$W(\lambda_1, \lambda_2, \lambda_3) = C_{10}(\lambda_1^2 + \lambda_2^2 + \lambda_3^2 - 3) + C_{01}(\lambda_1^{-2} + \lambda_2^{-2} + \lambda_3^{-2} - 3) \quad (3.10)$$

where C_1 and C_2 are material constants. Although the Mooney (also the Mooney-Rivlin [142]) equation is only able to describe relatively small strain behaviour. In order to better describe large deformations, wherein there is a strain-hardening effect [66], other models, such as the Ogden [121] and Yeoh [180] models can be used.

The strain energy function for the Ogden model has the form

$$W(\lambda_1, \lambda_2, \lambda_3) = \sum_{i=1}^M \frac{\mu_i}{\alpha_i} (\lambda_1^{\alpha_i} + \lambda_2^{\alpha_i} + \lambda_3^{\alpha_i} - 3) \quad (3.11)$$

where M is a positive integer that determines the number of terms used, while μ_i and α_i are material constants. The shear modulus μ of the material can be determined from the material constants by

$$\mu = \frac{1}{2} \sum_{i=1}^M \mu_i \alpha_i \quad (3.12)$$

As for the Yeoh model, its strain energy function is given by

$$W(\lambda_1, \lambda_2, \lambda_3) = C_{10}(\lambda_1^2 + \lambda_2^2 + \lambda_3^2 - 3) + C_{20}(\lambda_1^2 + \lambda_2^2 + \lambda_3^2 - 3)^2 + C_{30}(\lambda_1^2 + \lambda_2^2 + \lambda_3^2 - 3)^3 \quad (3.13)$$

where C_{10} and C_{20} and C_{30} are material constants.

Phenomenological models based on limiting chain extensibility models have also been formulated, of which the Gent strain energy function [57] is one of the simplest [66]

$$W(\lambda_1, \lambda_2, \lambda_3) = -\frac{\mu}{2} J_m \ln \left(1 - \frac{\lambda_1^2 + \lambda_2^2 + \lambda_3^2 - 3}{J_m} \right) \quad (3.14)$$

Where μ is the shear modulus and J_m is the constant limiting value for $\lambda_1^2 + \lambda_2^2 + \lambda_3^2 - 3$. A variant of the Gent strain energy function has been formulated to include dependence on the second invariant, i.e. $\lambda_1^{-2} + \lambda_2^{-2} + \lambda_3^{-2}$ [139], which has been referred to as the ‘Gent + Gent’ model [123]

$$W(\lambda_1, \lambda_2, \lambda_3) = -\frac{\mu}{2} J_m \ln \left(1 - \frac{\lambda_1^2 + \lambda_2^2 + \lambda_3^2 - 3}{J_m} \right) + C_2 \ln \left(\frac{\lambda_1^{-2} + \lambda_2^{-2} + \lambda_3^{-2}}{3} \right) \quad (3.15)$$

Once the material parameters have been obtained, they can be substituted into the respective constitutive model and used to model deformations with differing boundary conditions.

3.2 Effect of deformation-type on actuated strains

The amount of deformation in the dielectric elastomer is dependent on the magnitude of the electrostatic pressure, which acts in the direction of the elastomer’s thickness. In order to reduce the driving voltage, it would be preferable to generate large area stretches using small amounts of electrostatic pressure. As mentioned in section 2.3, the in-plane mechanical stiffness of the electrode affects the manner in which the elastomer deforms: a very stiff electrode would not permit any deformations in the elastomer while a perfectly compliant electrode does not give any resistance to elastomer deformations. For example, grease electrodes would result in an equi-biaxial deformation, corrugated metal film electrodes would result in a pure shear deformation, while zigzag-patterned metal film electrodes would result in a somewhat uniaxial deformation. In other words, without the use of any external loading, an equi-biaxial deformation requires biaxially compliant electrodes; a pure shear deformation requires electrodes to be compliant in one direction but stiff in the other; while uniaxial deformation requires electrodes that are expandable in one direction yet compressible in the other. The resultant elastomer geometries for these types of deformations are shown in Figure 3.6.

Figure 3.6. Schematic showing the common types of deformations in a DEA's elastomer: uniaxial, pure shear and equi-biaxial.

In this section, we show that the type, or directions, of electrode compliance matters. To do so, discrete values of electrostatic pressure are first prescribed to DEAs that can only deform biaxially, in pure shear and uniaxially. Following that, for each of the three types of DEA, the extent of deformation in the elastomer that can be maintained by each prescribed magnitude of electrostatic pressure is calculated. Using the resultant area stretch as a measure, we aim to show that electrodes that allow equi-biaxial deformations are preferred.

A simple membrane DEA, without mechanical prestretch, is considered here. Without prestretch, the principal stretches are generally expected to be less than 1.5, and taking these relatively small stretches into consideration, the Mooney-Rivlin hyperelasticity model is used. Taking the incompressibility of elastomers into consideration, the stretch in the length, breadth, thickness directions ($\lambda_1, \lambda_2, \lambda_3$) and area stretch (λ_{Area}), due to an applied electrostatic pressure, are related by

$$\lambda_{\text{Area}} = \lambda_1 \lambda_2 = \frac{1}{\lambda_3} \quad (3.16)$$

For equi-biaxial deformations, the deformations in the length and breadth directions are equal, such that

$$\lambda_2 = \lambda_1; \lambda_3 = \frac{1}{\lambda_1^2}; \lambda_{Area} = \lambda_1^2 \quad (3.17)$$

For pure shear deformations, the length deforms while the breadth does not, such that

$$\lambda_2 = 1; \lambda_3 = \frac{1}{\lambda_1}; \lambda_{Area} = \lambda_1 \quad (3.18)$$

For uniaxial deformations, the length deforms while the breadth contracts, such that

$$\lambda_2 = \frac{1}{\sqrt{\lambda_1}}; \lambda_3 = \frac{1}{\sqrt{\lambda_1}}; \lambda_{Area} = \sqrt{\lambda_1} \quad (3.19)$$

From the stress-stretch relation shown in equation (3.8), wherein $\sigma_1 = 0$, and by substituting the Mooney-Rivlin strain energy function, shown in equation (3.10), and the electrostatic pressure relation, shown in equation (3.9), electrostatic pressure σ_e is shown to be related to the stretches in the elastomer by

$$\sigma_e = 2C_{10} \left(\lambda_1^2 - \frac{1}{\lambda_{Area}^2} \right) + 2C_{01} \left(\lambda_{Area}^2 - \frac{1}{\lambda_1^2} \right) \quad (3.20)$$

Substitution of equations (3.17) in (3.20), (3.18) in (3.20) and (3.19) in (3.20), and by using a VHB elastomer, wherein C_{10} and C_{01} are 45 kPa and 41.2 kPa respectively [173], one can determine the relationships between an applied electrostatic pressure with the resultant area stretches in the different types of deformations. For example, when an electrostatic pressure of 200kPa is applied to a DEA that can deform equi-biaxially, the area stretch that can be maintained by such a compressive stress can be determined by using equations (3.17) and (3.20),

$$\begin{aligned}
2 \times 10^5 &= 2(45 \times 10^3) \left(\lambda_{Area} - \frac{1}{\lambda_{Area}^2} \right) \\
&\quad + 2(41.2 \times 10^3) \left(\lambda_{Area}^2 - \frac{1}{\lambda_{Area}} \right) \\
\Rightarrow \lambda_{Area} &\approx 1.44
\end{aligned} \tag{3.21}$$

The graph in Figure 3.7 shows the resultant area stretches due to a range of electrostatic pressure that had been applied to DEAs that can deform only either equi-biaxially, in pure shear or uniaxially. It is apparent that the generated area stretch is largest for biaxial deformations, given the same applied electrostatic pressure. By substituting equation (3.16) into equation (3.9), one can see that the electrostatic pressure is related to the square of the area stretch. This means that apart from being able to attain higher actuated area stretches at the same electrostatic pressure, the same electrostatic pressure can be attained at a lower driving voltage, given that the elastomers have the same original thickness. As such, in order to reduce the driving voltage of a DEA, it would be preferable to use electrodes that allow biaxial deformations, so as to generate larger area stretches using smaller amounts of electrostatic pressure and driving voltages. In addition, several applications, such as a diaphragm pump [7], tuneable liquid lens [38] or a loudspeaker, require biaxial deformations.

Figure 3.7. Difference between the generated area stretches in the VHB elastomer for the three deformation modes.

3.3 DEAs prestretched by constant-forces

The quasi-static mechanical response of a prestretched membrane DEA results from stresses induced in its active region: these induced stresses come from both electrical and mechanical sources. Electrically-induced stresses are brought about by the electrostatic forces generated by the charges in the electrodes, which results in the compressive forces acting in the direction of the active region's thickness. On the other hand, mechanically-induced stresses are brought about by tensile forces acting in the plane of the active region, which results in a prestretch in the elastomer. The elastomer in a DEA can be prestretched by external means, such as dead-weights or rigid structures. Alternatively, and perhaps preferably for the sake of reducing the actuator's weight and size, the elastomer can be prestretched via built-in features, such as using an IPN elastomer [61]. The introduction of a prestretch in the elastomer of a DEA thins it, which results in higher electrostatic pressure for a given voltage, and induces axial stresses that work in favour of improving actuated stretches. However, indefinitely increasing the prestretch in the elastomer does not equate to better electromechanical performances. In this section, the effects of using different magnitudes of biaxial prestretch, brought about by constant axial forces, are studied. Another salient factor that affects actuated stretches in DEAs is the stiffness of its electrodes. Due to the incompressible nature of elastomers used in DEAs, electrodes that are very stiff would constrict both planar expansions and thickness contractions during electromechanical activation. Electrode stiffness can range from zero, which can be attained by spraying charges on the elastomer [79], to more than a thousand times that of the elastomer, such as when using thick metal plate electrodes. Therefore, in this section, the extent to which the magnitude of electrode stiffness affects electromechanical response is also studied.

3.3.1 Model overview of DEAs prestretched by constant-forces

All deformations here are assumed to be homogeneous and equi-biaxial. The electrodes are applied to the elastomer in its prestretched state and as they will have to undergo moderate strains, they are approximated as hyperelastic materials. In order to simplify the analysis, prestretch is assumed to be a result of constant applied forces, such as that supplied by dead-weights. However, this form of prestretch is not commonly used in experiments. The fixed-frame method of maintaining prestretch results in forces that varies with actuated stretch and will be discussed in section 3.4. The model used is modified from that presented by Bozlar et

al. [23]. Our model utilises a thinner elastomer ($250\mu\text{m}$) and studies actuated stretches of only up to 2, which corresponds to a 300% area strain.

Referring to Figure 3.8, an elastomer with an original radius of R and thickness Z is stretched by dead-weights, with a total force of P . This stretch is termed as the prestretch λ_{pre} and the stress induced in the elastomer is called the prestress. In this prestretched state, a pair of electrodes, each having a thickness of $H/2$, is applied to the elastomer, such that they all have the same radius. It should be noted that the electrodes are stress-free at this point and stresses are only induced in the electrodes when the DEA is electromechanically activated. In the activated state, let the mechanical force acting on the elastomer and electrodes be P_1 and P_2 respectively, such that the total force acting on the DEA composite $P = P_1 + P_2$.

Figure 3.8. Schematic of a DEA prestretched by constant-forces, with its electrodes applied in its prestretched state.

When activated, the elastomer stretches by λ_{act} , such that its radius increases from $R\lambda_{\text{pre}}$ to $R\lambda_{\text{pre}}\lambda_{\text{act}}$. It experiences axial stresses brought about by force P_1 that act on its thickness $Z / \lambda_{\text{pre}}^2 \lambda_{\text{act}}^2$, along its circumference $2\pi R\lambda_{\text{pre}}\lambda_{\text{act}}$. At the same time, electrostatic stresses compress its thickness. By taking into account equations (3.8), (3.9) and (3.14),

$$\begin{aligned}
& \frac{P_1}{(2\pi R\lambda_{\text{pre}}\lambda_{\text{act}})\left(\frac{Z}{\lambda_{\text{pre}}^2\lambda_{\text{act}}^2}\right)} + \epsilon_o\epsilon_r \left(\frac{V}{\frac{Z}{\lambda_{\text{pre}}^2\lambda_{\text{act}}^2}}\right)^2 \\
& = \lambda_{\text{pre}}\lambda_{\text{act}} \frac{\partial \tilde{W}_{\text{elastomer}}(\lambda_1 = \lambda_{\text{pre}}\lambda_{\text{act}}, \lambda_2 = \lambda_{\text{pre}}\lambda_{\text{act}})}{\partial \lambda_1}
\end{aligned} \tag{3.22}$$

On the other hand, the pair of circular electrodes undergoes an actuated stretch of λ_{act} during activation, such that their total thickness becomes $H / \lambda_{\text{act}}^2$. Due to the induced radial force P_2 that acts on its circumference, and as stress in the thickness direction is zero, the pair of electrodes is subjected to axial stresses given by:

$$\frac{P_2}{(2\pi R\lambda_{\text{pre}}\lambda_{\text{act}})\left(\frac{H}{\lambda_{\text{act}}^2}\right)} = \lambda_{\text{act}} \frac{\partial \tilde{W}_{\text{electrode}}(\lambda_1 = \lambda_{\text{act}}, \lambda_2 = \lambda_{\text{act}})}{\partial \lambda_1} \tag{3.23}$$

By combining equations (3.22) and (3.23) and substituting $P_2 = P - P_1$,

$$\begin{aligned}
& \frac{Z}{Z + H\lambda_{\text{pre}}^2} \left[\frac{P_1}{(2\pi R\lambda_A)\left(\frac{Z}{\lambda_A^2}\right)} \right] + \frac{H\lambda_{\text{pre}}^2}{Z + H\lambda_{\text{pre}}^2} \left[\frac{P - P_1}{(2\pi R\lambda_A)\left(\frac{H\lambda_{\text{pre}}^2}{\lambda_A^2}\right)} \right] \\
& + \frac{Z}{Z + H\lambda_{\text{pre}}^2} \left[\epsilon_o\epsilon_r \left(\frac{V}{\frac{Z}{\lambda_A^2}}\right)^2 \right] \\
& = \frac{Z}{Z + H\lambda_{\text{pre}}^2} \left[\lambda_A \frac{\partial \tilde{W}_{\text{elastomer}}(\lambda_1 = \lambda_A, \lambda_2 = \lambda_A)}{\partial \lambda_1} \right] \\
& + \frac{H\lambda_{\text{pre}}^2}{Z + H\lambda_{\text{pre}}^2} \left[\frac{\lambda_A}{\lambda_{\text{pre}}} \frac{\partial \tilde{W}_{\text{electrode}}(\lambda_1 = \frac{\lambda_A}{\lambda_{\text{pre}}}, \lambda_2 = \frac{\lambda_A}{\lambda_{\text{pre}}})}{\partial \lambda_1} \right]
\end{aligned} \tag{3.24}$$

and after simplification,

$$\begin{aligned}
& \left[\frac{P\lambda_A}{2\pi RZ} \right] + \left[\epsilon_0 \epsilon_r \frac{V^2}{Z^2} \lambda_A^4 \right] \\
&= \left[\lambda_A \frac{\partial \tilde{W}_{\text{elastomer}}(\lambda_1 = \lambda_A, \lambda_2 = \lambda_A)}{\partial \lambda_1} \right] \\
&+ \frac{H\lambda_{\text{pre}}^2}{Z} \left[\frac{\lambda_A}{\lambda_{\text{pre}}} \frac{\partial \tilde{W}_{\text{electrode}}(\lambda_1 = \frac{\lambda_A}{\lambda_{\text{pre}}}, \lambda_2 = \frac{\lambda_A}{\lambda_{\text{pre}}})}{\partial \lambda_1} \right] \quad (3.25)
\end{aligned}$$

The prestress σ_{pre} , which is the axial stress induced by constant-force P acting on the elastomer, can be determined from the prestretch λ_{pre} in the elastomer by

$$\sigma_{\text{pre}} = \frac{P}{2\pi RZ} \lambda_{\text{pre}} = \lambda_{\text{pre}} \frac{\partial \tilde{W}_{\text{elastomer}}(\lambda_1 = \lambda_{\text{pre}}, \lambda_2 = \lambda_{\text{pre}})}{\partial \lambda_1} \quad (3.26)$$

A suitable strain energy function can be substituted into equations (3.25) and (3.26) in order to determine the relationship between the applied voltage and the actuated stretch in the DEA, which is defined $\lambda_A / \lambda_{\text{pre}}$. Here, the Gent model is used, see equation (3.14), wherein the material parameters for the elastomer (VHB) are $\mu = 50$ kPa and $J_m = 125$ [23]. However, other combinations of hyperelastic models were also studied and gave qualitatively the same results. The original thickness of the elastomer is $Z = 250$ μm , while its permittivity is given by its relative permittivity $\epsilon_r = 4.5$ multiplied by the permittivity of free space $\epsilon_0 = 8.854 \times 10^{-12} \text{Fm}^{-1}$. It is subjected to a prestretch of between 1 and 5. The electrode has an original thickness H of $1\mu\text{m}$, and is modelled to have a shear modulus of equal to, and a hundred times, that of the elastomer. The MATLAB code is shown in appendix A1.

3.3.2 Effects of prestretch and electrode stiffness

Here, we study the actuated strain performance of DEAs by first varying the extent to which the elastomer is prestretched, and then by varying the electrode stiffness of the DEA. By subjecting the elastomer of a DEA to different magnitudes of prestretch, it can be seen in

Figure 3.9a that the relationship between the elastomer's stretch and driving voltage changes. When the gradient of a curve's tangent first turns zero, it signifies an event in which the DEA would continue to expand in area even though the voltage supplied remains unchanged. This indicates mechanical instability and will likely result in actuator failure; "snap-through" deformation [88, 106, 187] is not considered as it does not occur within the range of stretch studied here. The two dashed-curves correspond to breakdown strengths of 100MV/m and 150MV/m; it is assumed that prestretch does not affect breakdown field. Its intersection with a curve indicates that the electric field experienced by the DEA has reached the breakdown strength, resulting in a short circuit through the elastomer and hence failure of the actuator. It can be seen that generally, the actuator is more likely to fail via mechanical instability at lower magnitudes of prestretch and more likely to fail by electrical breakdown at higher magnitudes of prestretch.

Figure 3.9. The effects of prestretch are shown by subjecting the elastomers to prestretch magnitudes of between 1 and 5. The electrodes are taken to have a thickness of $1\mu\text{m}$ and a shear modulus that is equal to the shear modulus of the elastomer.

As our primary interest is in actuated strains of the DEA, and not in the absolute stretch in its elastomer, we can make a clearer comparison by looking at Figure 3.9b. The marked points on the peaks of the curves indicate the start of mechanical instability; the solid curves end at a breakdown field of $100\text{V}\mu\text{m}^{-1}$ and the dotted curves end at a breakdown field of $150\text{V}\mu\text{m}^{-1}$. By increasing the amount of prestretch in the elastomer, lower driving voltages are generally required to attain the same actuated stretch. This is due to the elastomer being thinner, which has the combined effect of a higher generated electrostatic pressure, at the same driving voltage, and higher axial stresses, given the same applied axial force. However, it is also noted that this method of using a larger prestretch to lower driving voltage becomes less effective at higher prestretch values. With regards to actuator failure modes, higher prestretch magnitudes enables mechanical instability to take place at larger actuated stretches, but it also results in electrical breakdowns occurring at lower actuated stretches.

The maximum actuated stretch prior to either mechanical instability or dielectric breakdown (at $100\text{V}\mu\text{m}^{-1}$ and $150\text{V}\mu\text{m}^{-1}$) is plotted in Figure 3.9c. Maximum actuated stretches for DEAs with prestretch values of less than 2 are determined by failure due to mechanical instability while those with prestretch values of more than 2 are determined by failure due to electrical breakdown. Depending on the dielectric strength of the elastomer and the stiffness of the electrodes, and by balancing the need to attain large actuated stretches with the use of lower driving voltages, we can see that the optimal prestretch for the modelled elastomer is in the vicinity of 2.5.

In order to study the effect of electrode stiffness on the actuated stretch performance of DEAs, we now fix the prestretch in the elastomer at $\lambda_{\text{pre}} = 2.5$ and let the electrode have a shear modulus that is 0, 10 and 100 times the elastomer's shear modulus. The MATLAB code for studying the effect of electrode stiffness is shown in appendix A2. As can be seen in Figure 3.10, in order to attain the same actuated stretch, higher driving voltages are required by DEAs using electrodes with a larger shear modulus. A similar effect is obtained by using electrodes that are thicker. This is because an increment in both shear modulus and thickness result in a stiffer electrode that requires more electrostatic pressure to deform. The high driving voltages required by DEAs with stiffer electrodes tend to result in electrical breakdowns occurring at lower stretches. For example, assuming that the dielectric breakdown occurs at 150 MV/m , a DEA using an electrode that has a shear modulus 500 times that of the elastomer has a relatively small maximum actuated stretch of less than 1.14.

In addition, it is also apparent that DEAs with stiffer electrodes also cause mechanical instability to occur at a lower stretch, which also tends to result in actuator failure. From this, it is clear that thinner electrodes with a lower shear modulus are preferred for better electromechanical performance. However, it should be kept in mind that the primary requirement of electrodes is that they must still be able to remain conductive and continuous even under high stretches.

Figure 3.10. Effect of electrode stiffness. The DEAs are modelled using electrodes with a shear modulus of between 0 and 500 times the shear modulus μ of the elastomer, which has an original thickness of $250\mu\text{m}$ and a prestretch of 2.5.

3.4 DEAs prestretched by a rigid frame

In most experimental setups, a rigid frame is used to maintain a prestretch in the elastomer as it is convenient to implement and results in a very simple DEA setup. However, in such a setup, the axial forces exerted on the active DEA varies with actuated stretch, unlike what was described in section 3.3, wherein axial forces were constant. When introducing a prestretch via a rigid frame, an elastomer is first mechanically stretched biaxially, and then secured onto a fixed-frame, as is depicted in Figure 3.2. Subsequently, a pair of electrodes is applied to the centre of the prestretched elastomer to form the active region. The bare part of the prestretched elastomer, which borders the active region, is termed the passive region as it is not electrically connected. However, it plays the vital role of inducing a prestress in the active region. Initially, when the driving voltage is zero, stretch in both the active and passive regions are equal to the prestretch. However, when electromechanically activated, stretch in the active region increases, which causes the radial stretch in the passive region to decrease, as a result of its outer boundary being fixed by the rigid frame. The model used here is based on that presented by Koh et al. [88], but modified to include the effects of electrodes that have appreciable stiffness.

3.4.1 Model overview of DEAs prestretched by a rigid frame

The three main states of a DEA prestretched by a rigid frame are shown in Figure 3.11. In the reference state, the undeformed elastomer has a thickness of Z , an overall diameter of $R = R_B$ and its active region has a diameter of $R = R_A$ (the “active region” is not actually active here, but denotes the material points that will subsequently be coated with electrodes). When radially or equi-biaxially prestretched by λ_{pre} , the entire elastomer deforms homogeneously, and the outer radius of the elastomer is fixed at $r_B = R_B \lambda_{pre}$. In this prestretched state, a pair of stress-free electrodes, of total thickness H , is applied onto either side of the active region. Upon activation, the active region, together with the electrodes, expand to a radius of $r_A = R_A \lambda_{pre} \lambda_{act}$, wherein λ_{act} is the actuated stretch, while r_B remains unchanged as it is anchored to the rigid frame.

Figure 3.11. Schematic of a circular membrane DEA in its (top) reference or undeformed state, (middle) prestretched state and (bottom) activated state.

The membrane tensions acting on the circular active region are equal to those acting on the annular passive region. As shown in Figure 3.12, the total axial force P acting on the active region comprised of force P_1 acting on the dielectric elastomer and force P_2 acting on both electrodes. The axial forces acting on the circular active region and electrodes can be calculated in the same way as for equi-biaxial deformations, as discussed in section 3.3.1. Deformations in the annular-shaped passive region are axisymmetric and vary with radius. The radial interfacial stress between the active and passive regions varies with actuated stretch. For each prescribed actuated stretch, the stretches in the annular passive region is solved as a boundary value problem. Once the stretches at the interface have been

determined, the axial force acting on the passive region P can be determined, together with component forces P_1 and P_2 acting on the active region.

Figure 3.12. Balancing of forces at the interface of the passive and active regions.

3.4.2 Passive region

This section first shows the formulation of the boundary value problem required to solve for the deformations in the annular passive region. Following that, the effects of prestretch and geometrical dimensions on deformations, and therefore stresses, in the passive region are studied. The deformations in the passive region are axis-symmetric and vary with radius. Here, it would be more convenient to use polar coordinates in place of Cartesian coordinates. Also, the Lagrangian coordinate system is used (for reasons discussed in section 3.1): With reference to Figure 3.11, R describes a point in the reference state while r describes a point in either of the deformed states.

The principal stretches are assumed to be aligned to the polar coordinate system used. As such, the deformations at each point can simply be described by its radial and tangential

stretches. The radial stretch describes deformations in the radial direction, or the change in an element's length, from dR to dr ,

$$\lambda_R = \frac{dr}{dR} \quad (3.27)$$

while tangential stretch describes deformations in the tangential direction, which in this axis-symmetric case is simply the change in the circumference along the radius of the point

$$\lambda_\theta = \frac{2\pi r}{2\pi R} = \frac{r}{R} \quad (3.28)$$

The forces experienced by each point along the radius can be balanced by using the momentum equation for polar coordinates (no acceleration and no body forces)

$$\frac{ds_R}{dR} + \frac{s_R - s_\theta}{R} = 0 \quad (3.29)$$

wherein s_R and s_θ are the nominal stresses (see section 3.1.2) in the radial and tangential directions, respectively. It should be noted that the differential equation is with respect to coordinates in the undeformed reference state (Lagrangian coordinates). It would now be convenient to express the stresses in terms of deformations by making use of constitutive relations. Due to the orthogonality property of polar coordinates, directions 1 and 2 of the Cartesian coordinates can be substituted by the radial and tangential directions, respectively.

The change in radial stretch with respect to position R can be determined by substituting equation (3.29), $\sigma_1 = s_R \lambda_R$, $\sigma_2 = s_\theta \lambda_\theta$, $\lambda_1 = \lambda_R$ and $\lambda_2 = \lambda_\theta$, in equations (3.7) and (3.8), to obtain [88]

$$\frac{d\lambda_R}{dR} = \frac{\frac{\partial \tilde{W}(\lambda_R, \lambda_\theta)}{\partial \lambda_R} - \frac{\partial \tilde{W}(\lambda_R, \lambda_\theta)}{\partial \lambda_\theta}}{R \left[\frac{\partial^2 \tilde{W}(\lambda_R, \lambda_\theta)}{\partial \lambda_R^2} \right]} \quad (3.30)$$

The change in tangential stretch with respect to position can be obtained by differentiating equation (3.28) with respect to R,

$$\frac{d\lambda_\theta}{dR} = \frac{1}{R}(\lambda_R - \lambda_\theta) \quad (3.31)$$

In order to determine the deformations in the annular passive region of the DEA, differential equations (3.30) and (3.31) are solved while taking into consideration the boundary conditions at the passive region's inner at outer radii. The position of the rigid frame, which corresponds to the prestretched outer radius of the passive region, remains unchanged throughout, such that

$$\lambda_\theta(R = R_B) = \frac{r_B}{R_B} = \lambda_{pre} \quad (3.32)$$

The second boundary condition is imposed at the interface between the active and the passive regions, wherein the outer radius of the active region should equal to the inner radius of the passive region,

$$\lambda_\theta(R = R_A) = \frac{r_A}{R_A} = \lambda_{pre}\lambda_{act} \quad (3.33)$$

The stretches can be obtained by solving differential equations (3.30) and (3.31), while taking into account boundary conditions (3.32) and (3.33). This has been carried out by making use of MATLAB's bvp4c function, which is an in-house algorithm designed for solving boundary value problems. The MATLAB code is shown in appendix A3.

The nominal radial stress s_R , at each actuated stretch, the radial and tangential stretches at a point on the passive region's inner radius, such that.

$$s_R = \frac{\partial \tilde{W}(\lambda_R(R = R_A), \lambda_\theta(R = R_A))}{\partial \lambda_R} \quad (3.34)$$

The present model has been used to simulate the change in membrane stresses, in a 2.5 times equi-biaxially prestretched DEA. The relative size of the passive region to the active (electrode-covered) region is reflected in its diameter ratio, which is defined as the ratio of the fixed-frame's diameter to the electrode's initial diameter (R_B/R_A). Here, the Gent model is used ($\mu = 50\text{kPa}$, $J_m = 125$) and the elastomer is taken to have an initial thickness of $Z = 250\mu\text{m}$. If not mentioned, the electrodes are assumed to be 1 micron thick and to have a shear modulus equal to the elastomer's shear modulus.

Due to the fixed boundary, activation of a prestretched membrane DEA reduces the membrane tension. Figure 3.13 shows that the true stress acting on the active region reduces with increasing actuated stretch. Higher actuated stretches may even cause a loss of membrane tension, causing tensile stresses to become compressive stresses. For example, 1.5 times equi-biaxially prestretched DEA loses membrane tension at an actuated stretch of 1.7. This loss of membrane tension may induce wrinkling and even electromechanical instability [86, 78].

Figure 3.13. Effect of prestretch on the change in true radial stress with increasing actuated stretch. This is simulated at a R_B/R_A ratio of 3 (diameter of the fixed-frame to the diameter of the prestretched active-region at zero voltage).

The effect of prestretch on the DEA membrane is also studied. A larger force and stress is required to maintain a higher membrane prestretch, especially since a higher prestretch results in a thinner elastomer membrane. In addition, the strain stiffening effect of the elastomer can be observed with increasing prestretch. In the region of the prestretch values used, as shown in Figure 3.14, the stress increases at an increasing rate with respect to stretch. However, it can also be observed that the drop in stress, with increasing actuated stretch, is larger for elastomers with a higher prestretch. This is because, for a given actuated stretch, although the absolute reduction in the passive area is the same, the relative reduction is larger for an elastomer with a higher prestretch, due to its smaller original dimensions (see appendix A4).

Figure 3.14. Stress-stretch behaviour of a biaxially deformed elastomer, based on the Gent model.

The decrease in stress, with increasing actuated stretch, is also exacerbated by using smaller geometric ratios, wherein the passive region's outer diameter becomes smaller with respect to the electrode's initial diameter. In contrast, when using a constant-force prestretch, the stress increases with actuated stretch. Figure 3.15 shows the effect of geometric ratio on the behaviour of an elastomer that had been prestretched by 2.5 times. By increasing the geometric ratio, wherein the fixed-frame's diameter becomes larger with respect to the electrode's diameter, the reduction in stress with increasing actuated stretches becomes less severe. However, this effect reduces with increments at higher ratios, and there is little benefit in going beyond a geometric ratio of 10. In a DEA, larger geometric ratios equate to greater weight and volume, which is undesirable from the practical point of view.

Figure 3.15. Effect of ratio (diameter of the rigid frame to the diameter of the prestretched active-region at zero voltage) on the change in pre-stress with increasing actuated stretch. This is studied at a prestretch of 2.5.

3.4.3 Coupling of the active and passive regions

The prestretched membrane DEA has a circular active region that is bordered by an annular passive region. At the interface, the axial forces acting on the active region is equal to that acting on the passive region. According to equation (3.25) describing equi-biaxial deformations, the total axial force acting on the thickness of the active region at $R=R_A$ is

$$\begin{aligned}
 P = \frac{2\pi R_A Z}{\lambda_{act} \lambda_{pre}} & \left\{ \left[\lambda_{act} \lambda_{pre} \frac{\partial \tilde{W}_{elastomer}(\lambda_1 = \lambda_{act} \lambda_{pre}, \lambda_2 = \lambda_{act} \lambda_{pre})}{\partial \lambda_1} \right] \right. \\
 & + \frac{H \lambda_{pre}^2}{Z} \left[\lambda_{act} \frac{\partial \tilde{W}_{electrode}(\lambda_1 = \lambda_{act}, \lambda_2 = \lambda_{act})}{\partial \lambda_1} \right] \\
 & \left. - \left[\epsilon_o \epsilon_r \frac{V^2}{Z^2} (\lambda_{act} \lambda_{pre})^4 \right] \right\} \quad (3.35)
 \end{aligned}$$

where $P = P_1 + P_2$, and P_1 and P_2 are the axial forces acting on the elastomer and the electrodes, respectively.

On the other hand, the force P acting on the passive region, at $R = R_A$, can be determined from equation (2.34), which uses polar coordinates.

$$P = \frac{2\pi R_A Z}{\lambda_{\text{act}} \lambda_{\text{pre}}} \left[\lambda_R \frac{\partial \tilde{W}(\lambda_R(R = R_A), \lambda_\theta(R = R_A))}{\partial \lambda_R} \right] \quad (3.36)$$

P depends on both the radial and tangential stretches (λ_R and λ_θ) in the passive region, at $R = R_A$, which can be determined from the boundary value problem described in section 3.4.2.

The voltage V required to cause a prescribed actuated stretch can be determined by equating equation (3.35) to equation (3.36), such that

$$V = \frac{Z}{\sqrt{\epsilon_o \epsilon_r} (\lambda_{\text{act}} \lambda_{\text{pre}})^2} \left\{ \lambda_{\text{act}} \lambda_{\text{pre}} \frac{\partial \tilde{W}_{\text{elastomer}}(\lambda_1 = \lambda_{\text{act}} \lambda_{\text{pre}}, \lambda_2 = \lambda_{\text{act}} \lambda_{\text{pre}})}{\partial \lambda_1} \right. \\ + \frac{H \lambda_{\text{pre}}^2 \lambda_{\text{act}}}{Z} \frac{\partial \tilde{W}_{\text{electrode}}(\lambda_1 = \lambda_{\text{act}}, \lambda_2 = \lambda_{\text{act}})}{\partial \lambda_1} \\ \left. - \lambda_R \frac{\partial \tilde{W}(\lambda_R(R = R_A), \lambda_\theta(R = R_A))}{\partial \lambda_R} \right\}^{\frac{1}{2}} \quad (3.37)$$

3.4.4 Effect of prestretch, geometric ratio and electrode stiffness

As described in section 3.4.2, a DEA that is prestretched by a fixed frame is subjected to a reduction in axial stress due to actuated strains. This reduction in stress is illustrated in Figure 3.13, where the passive annular region reduces in area and becomes thicker as the active region expands and thins under Maxwell stress. The extent of axial stress reduction depends on the ratio of the diameters of the passive to the active region. In turn, the stress reduction affects the electromechanical response of DEAs. Figure 3.16 shows the effect of varying electrode size relative to the fix frame (varying R_B/R_A) on the actuation of a DEA with

prestretch of 2.5. A DEA with a relatively small electrode (large ratio of $R_B/R_A=10$) requires 10% less voltage to achieve the same actuated stretch as compared with the DEA with relative bigger electrode (at $R_B/R_A = 3$). This can be attributed to the smaller extent of stress reduction with increasing actuation as described for the DEA with small electrodes (large ratio of $R_B/R_A=10$).

In a similar trend, the DEA, which is prestretched by constant-forces, requires much less voltage to achieve the same actuated stretch. This confirms that fixed-frame prestretch does not exert as much prestress as the constant-force prestretch during electromechanical activation. In addition, the DEAs with a fixed-frame prestretch exhibit electromechanical instability around 2500V while the DEA under constant-force prestretch does not.

Next, we study the effect of prestretch on the electromechanical response of DEAs. Consider the initially 250 μ m-thick elastomer under varying prestretch while having one fifth of its elastomer's diameter coated with electrodes (i.e. $R_B/R_A=5$). Prestretch will reduce the membrane thickness of dielectric elastomer and consequently help increase the electrostatic force generation over the active (electrode-covered) region. Figure 3.17 shows that increasing the prestretch reduces the voltage requirement to attain the same actuated stretch. For example, an actuated stretch of 1.2 requires a driving voltage of 3800V, 2400V, 1880V and 1760V by DEAs prestretched by 1.5, 2.5, 3.5 and 4 respectively.

In addition, higher prestretch helps to suppress electromechanical instability due to the strain stiffening effect. At a prestretch of 3.5 or 4, the DEAs are free from electromechanical instability and can possibly attain large actuations if the dielectric material is able to sustain very high electric fields. However, their actuated stretch increases a decreasing rate with respect to increasing driving voltage. This tapering of actuation can be attributed to a reduced prestress with increasing actuation, as shown in Figure 3.13. On the other hand, if the material has a limited breakdown strength of 100V/ μ m, a 2.5 times pre-stretched DEA can produce a maximum actuated stretch of 1.26 at a 2500V breakdown voltage; whereas, a 4 times prestretched DEA can produce a maximum actuated stretch of 1.08 at a 1300V breakdown voltage.

Figure 3.16. Effect of geometric ratio on electromechanical response. The DEAs have a prestretch of 2.5 and use 1 micrometre thick electrodes that have the same shear modulus as the elastomer.

Figure 3.17. Effect of prestretch on electromechanical response. (left) The DEAs have a geometric ratio of 5 and use 1 micrometre thick electrodes that have the same shear modulus as the elastomer. The short curves that cross the voltage-stretch curves represent a breakdown field of $100\text{V}/\mu\text{m}$ and $150\text{V}/\mu\text{m}$. The marked points show the start of mechanical instability.

Here, we also study the effect of electrode stiffness on the electromechanical response of the DEA. Although the electrodes are just thin coatings, of say $1\mu\text{m}$ -thick, on a comparatively thick dielectric elastomer membrane, they are made of a stiffer material, which has a shear modulus that ranges up to 1000 times the shear modulus of the elastomer, μ . Stiff electrodes are expected to reduce the electromechanical actuation of soft dielectric elastomers. Figure 3.18 shows that a DEA with stiffer electrodes requires a higher driving voltage to attain the same actuated stretch. In other words, the stiffened DEA achieves a lower maximum actuated stretch at the same driving voltage. In addition, electromechanical instability in DEAs with stiffer electrodes also occurs at a lower actuated stretch. If the dielectric strength is limited to 100MV/m , the stiffened DEA sustains lower actuated stretches as compared to the DEA with compliant electrodes.

Figure 3.18. Effect of electrode stiffness on electromechanical behaviour. The DEAs have a geometric ratio of 5 and a prestretch of 2.5. Their electrodes are 1 micrometre thick and have a shear modulus that is equal to (1μ), 10 times (10μ), 50 times (50μ), 100 times (100μ), 500 times (500μ) and 1000 times (1000μ) the shear modulus of the elastomer. Points of mechanical instability are marked; the curves cross the voltage-stretch curves at a breakdown field of $100\text{V}/\mu\text{m}$ and $150\text{V}/\mu\text{m}$.

3.5 Discussion on the modelling of DEAs

Biaxial deformations result in larger actuated area strains for a given electrostatic pressure, therefore it would be preferable for electrodes to be biaxially stretchable and biaxially compliant. Two DEA setups were modelled: DEAs with a prestretch induced by a constant-force and by a rigid-frame. Based on these models, we have simulated the DEA's biaxial deformations due to the application of electrostatic pressures. The general behaviours for both setups were found to be qualitatively the same. Generally, lower voltages were required to attain the same actuated strain at a higher prestretch. Also, at a lower prestretch, the DEA tends to fail via mechanical instability while at a higher prestretch, the DEA tends to fail via electrical breakdown. However, in the rigid-frame prestretch setup, although a larger prestretch results in a larger prestress, there is a larger drop in prestress with increasing actuated strain. In contrast, the prestress increases with actuated strain for the constant-force prestretch setup. From the simulated results, a prestretch of about 2.5 appears to be optimal for attaining large actuated strains and will therefore be used in the experiments. The effect of electrode stiffness was also studied using the two models. Generally, by using stiffer electrodes in the DEA, higher voltages are required to attain the same actuated strains. In addition, with stiffer electrodes, mechanical instability and electrical breakdown tended to occur at lower actuated stretches. Therefore, one should aim to use electrodes of a low stiffness in order to attain high actuated strains in a DEA. The size of the rigid frame that holds the DEA's prestretch was also found to affect the actuated strains. A larger frame, with respect to the size of the electrodes, resulted in a smaller drop in prestress with increasing actuated strain. However, as the effect diminishes with increasing frame size, a practical fixed-frame to electrode diameter ratio of five will be used in the experiments.

4 Experimental details

In this chapter, we describe the experiments that have been utilised to evaluate the stretchability and the stiffening effect of the electrodes used in DEAs. This is because DEAs require electrodes that are compliant, such that the electrodes are able to follow the large area expansions of the elastomer, without impeding the deformations. So far, conductive particles suspended in grease have proven to be the most compliant, as it is liquid-like and can therefore flow. Even conductive particles that have been impregnated in elastomers have a stiffening effect on the elastomer. As compared to conductive particles, plain metal thin film electrodes are very stiff and have much lower fracture strains. Here, we make use of the results obtained from the model discussed in chapter 3, which has shown that the actuated strains of a DEA is affected by the mechanical stiffness of the electrodes and dielectric elastomer, as well as its prestretch and the size of the rigid frame that holds it.

The compliance and stretchability of a DEA's electrodes manifest in the attainable actuated strains in activated DEAs. The actuated area stretch in a DEA is defined as the ratio of the expanded area A , due to a supplied voltage, to the original area A_o , when no voltage was supplied, as shown in Figure 4.1a. Generally, compliant electrodes allow large area expansions while stiff electrodes limit area expansions, as depicted in Figure 4.1b.

Figure 4.1. a) Schematic showing the electromechanical activation tests that were used to determine the b) relationship between area stretch and driving voltage.

In order to quantify the stretchability and compliance or stiffening effect of the electrodes, we have made use of tensile tests conducted on electrode-covered elastomers, similar to the

sandwich composites used in the DEA configurations. The tensile tests were conducted using uniaxial deformations, as shown in Figure 4.2a, and radial deformation, as shown in Figure 4.2b. The stiffening effect of the electrodes is determined by comparing the amount of stress required to deform an electrode-coated elastomer with the amount of stress required to deform a bare elastomer. The stresses and stretches can be calculated from the force P and geometry: width w , thickness z and radius r , as shown in Figure 4.2c, in order to obtain the relationship between stress and stretch, as shown in Figure 4.2d. The stretchability of the electrodes is determined by measuring the change in the resistance across the electrodes with increasing amounts of deformations, as shown in Figure 4.2e.

Figure 4.2. Schematic showing the tensile tests for deformations that are a) uniaxial and b) radial. c) Stress and stretch relationships were used to find the d) stiffening effect of the electrode on the elastomer and the e) stretchability of the electrode.

4.1 Prestretched membrane DEAs

This section describes the sample preparation and experimental setup used for the electromechanical activation of prestretched membrane DEAs. A single layer membrane DEA, comprising one dielectric layer and two electrodes was used, as shown in Figure 4.3.

All of the samples had dielectric layers comprising VHB F9473PC elastomers that were prestretched by 2.5 times, unless otherwise stated.

Figure 4.3: Setup of a single layer membrane DEA prestretched by a rigid frame.

Fabrication of membrane DEAs basically requires one to first stretch an elastomer, fix it to a rigid frame and then apply the electrodes on, as shown in Figure 4.4. In order to prestretch the elastomer, a square piece was cut out from the roll of VHB tape. Following that, concentric squares of 28mm and 20mm were then marked out. The elastomer was then manually stretched such that its markings matched the inner (50mm) and outer (70mm) dimensions of the square acrylic frame. Once adhered to the frame, a pair of 10mm diameter electrodes was applied to either side of the elastomer. Thin conductive trails were also applied, such that they extended from each circular electrode from opposite sides. These trails were in turn connected to thin aluminium strip conductors, with silver conductive grease at their interface in order to ensure good electrical contact. At least five samples were tested for each type of electrode and the average was used for comparison.

Figure 4.4. Schematic showing the fabrication of prestretched membrane DEAs.

As illustrated in Figure 4.5, each DEA was connected to a high voltage source (610E, Trek Inc.) and its voltage and current, with respect to time, was logged by means of a data acquisition card (PCI 6052E, National Instruments). Simultaneously, a digital camera (Canon EOS 550D) fitted with a macro lens (Tamron AF 90mm f/2.8 Di SP A/M 1:1 Macro Lens) was used to take photographs or videos of the electrode as it was subjected to discrete values of voltage. These photographs show the top-view of the electrodes, such as that shown in Figure 4.6 of circular graphite electrodes.

Figure 4.5. Schematic of the experimental setup used to activate a membrane DEA, while logging its voltage, current and actuated strain.

Figure 4.6. Area expansion during electromechanical activation of these DEAs using graphite powder electrodes can clearly be seen by comparing the electrode area at a particular voltage with the electrode area at zero voltage.

Videos were converted to still-photographs (QuickTime 7 Pro, Apple) and all photographs were analysed using MATLAB in order to determine the changes in area. In order to determine the actuated area strain of a DEA, at a particular voltage, the number of pixels in the photograph of the electrode was normalised by the number of pixels in the photograph of the electrode at 0V. The MATLAB code used, as shown in appendix A5, makes use of the contrast between the dark electrodes and the light background, which can be seen through the transparent VHB elastomer. In order to exclude the electrode trails from the analysis, as they do not actively contribute to actuated strains, white ink (Uni-ball, Mitsubishi pencil) was applied to mark the active electrode boundary, as shown in Figure 4.7; the ink does not have a significant impact on actuated strains.

Figure 4.7. A DEA with circular sputtered silver thin film electrodes, which have cracked when strained due to actuation. Its trails are marked with white ink in order to improve the contrast for imaging purposes.

4.2 Materials in a DEA

This section introduces the materials used for the dielectric elastomer and the electrodes.

4.2.1 Elastomer

A few basic properties need to be taken into consideration when selecting an elastomer that can help enhance the performance of the DEA: these include a high dielectric constant, a high breakdown electric field and a sufficiently low Young's modulus to allow the required deformation. Throughout literature, a wide variety of elastomers have been tried out [130], but the commercial materials that have stood out are the acrylic and silicone elastomers. Acrylic elastomers generally have a high permittivity and DEAs using acrylic elastomers, specifically the VHB adhesive tape series, have shown much greater actuated strains than DEAs using silicone elastomers. However, a large prestretch is required to produce these large actuated strains, which can encumber and limit the development of certain types DEA configurations. Acrylic elastomers are also more viscoelastic than silicone elastomers, which results in larger hysteresis and creep behaviours, as well as a slower response speed [10]. Unlike silicone elastomers, acrylic elastomers are unable to operate within a wide temperature range.

As the focus of this work is on the electrodes used in DEAs, the electrodes should be tested to high stretches in order to determine their viability as compliant electrodes. In view of this, acrylic elastomers have been chosen, as they can be stretched by more than five times their

original lengths. The most commonly used types of acrylic elastomer in the study of DEAs are VHB 4905 and VHB4910, which are 500 μ m and 1mm thick respectively [94, 149]. In a quest to reduce driving voltage, while reducing forces on the external support (caused by large magnitudes of prestretch), a thinner acrylic elastomer from the same VHB series (250 μ m-thick VHB F9473PC) has been used for most of the experiments carried out. In addition, according to 3M data sheets, the dielectric strength of an undeformed VHB F9473PC (5500V/mil [2]) is higher than that of an undeformed VHB 4910 (630V/mil [1]). Subsequently, unless specifically named, all mention of VHB refers to VHB F9473PC.

4.2.2 Electrodes

The choice of electrodes in a DEA is just as important as the dielectric elastomer used. DEA electrodes should preferably have high conductivity, even at high strains, low stiffness, stability and fault tolerance. In order to serve as a comparison to the focus of this work, i.e. ELD silver thin-film electrodes, a few other types of electrodes were used: graphite powder, silver grease, sputtered silver thin-film and beaten gold leaf (see Figure 4.8).

Figure 4.8. Different types of electrode materials used. a) graphite powder, b) silver grease, c) sputtered silver and d) gold leaf.

- a) Graphite powder: The TIMREX KS6 (TIMCAL Graphite & Carbon) is a synthetic graphite, with a 6.5 μ m particle size, that is commonly used as conductive additives in the positive electrode of lithium batteries. It has been used as electrodes for DEAs by applying the powder directly onto the dielectric layer [102] and by embedded the particles in a PDMS [32]. In the present work, the graphite powder was adhered to the tacky VHB surface by using a soft-bristled brush, in a gentle tapping motion, to press the powder on. The resultant graphite layer obtained was about 10 μ m thick.

- b) Silver grease: The silver conductive grease used (CW7100, CircuitWorks, Chemtronics) is a suspension of silver particles in a silicone lubricant and has been used as electrodes for DEAs [75, 113]. Here, the silver grease was applied onto the dielectric layer by simply brushing it on.
- c) Sputtered silver thin film: Sputter deposition is a commonly used method, in microelectronics, for fabricating thin-films. It makes use of energetic gaseous ions to bombard a metal target in order for metal to sputter off the target's surface to coat the substrate [41]. Magnetron sputtering has been chosen over arc evaporation as it has a water-cooled target, which results in smaller heat generation that can potentially damage the sample. A modified sputtering system was used to fabricate the corrugated silver thin films used in Danfoss PolyPower's DEAs, which are known to have self-clearing properties [163]. The sputtered silver thin films in the present work were deposited until 150nm thick.
- d) Gold leaf: Gold that had been beaten down into sheets of gold leaves is commonly used for ornamental gilding. 24 carat gold leaves (Wrights of Lymm Ltd.) were used in this work and had a thickness of about 120nm. The gold leaves come in the form of transfer sheets for easy handling. The gold leaf was first laid onto the tacky VHB surface and then using a soft-bristled brush, the gold sheet was pressed on to ensure the adhesion of the gold leaf to the elastomer.

The thickness of metal thin films were obtained by using an atomic force microscope to measure the step-height of the thin-films that had been deposited on glass slides, while the thickness of the graphite powder coating was measured from its topography using a confocal microscope.

4.3 Stretchability

The stretchability of an electrode is its ability to deform while remaining electrically conductive. This is an important property in DEAs as the electrodes need to be able to supply charges to the surface of the elastomer, which can deform to more than twice its original area. This section describes the sample preparation and experimental setup for uniaxial and biaxial stretchability tests. These tests aim to determine how much an electrode can be stretched before losing its electrical conductivity.

4.3.1 Uniaxial stretchability

For the samples used in the uni-axial tensile tests (Figure 4.9), VHB 4905 films were cut into strips of 25mm by 75mm. The electrode material, to be tested, was applied to both sides of the centre section spanning 25 mm. Two electrical leads were placed on each end of the exposed area, with silver grease applied at the interface, in order to improve electrical contact. Following that, two pairs of acrylic plates were used to clamp the ends of the VHB film, such that only the middle square section was free to deform. These samples were stretched uniaxially, at a rate of 3 millimetres per minute, by a mechanical testing system (Instron 5565), and the resistance across the stretched length was logged by a digital multimeter (34410A, Agilent Technologies), which was connected using the four-probe resistance measurement method. At least three samples were tested for each type of electrode and the average was used for comparison.

Figure 4.9. An electrode-covered elastomer being stretched uniaxially (vertically) by an Instron mechanical testing machine.

4.3.2 Radial stretchability

A homemade radial stretching jig, as shown in Figure 4.10, was fabricated in order to radially stretch and relax an elastomer. Circular pieces of diameter 45mm were cut out from a roll of

VHB F9473PC tape and in order to mitigate tearing, the circumferences of the elastomers were reinforced by a layer of elastomer of 5mm width. Following that, 40mm diameter electrodes were applied to the elastomers. The reinforced circumference of each elastomer was fastened to sixteen crocodile clips, which were in turn attached to a stepper motor via sixteen cords. Depending on the rotation of the motor, the elastomer could be extended or retracted by equal distances, at varying speeds. For the radial-stretchability tests, a rate of 3.6 times, the original diameter of the elastomer, per minute was used. At least three samples were tested for each type of electrode and the average was used for comparison.

Figure 4.10. (top) a homemade motorised radial stretcher that is used to radially stretch and relax an elastomer. (bottom) schematic of the cross section of the radial stretcher.

The radial stretchability of the electrode was determined by the monitoring the resistance across its diameter as it was radially stretched. The four-probe resistance measurement method was used to measure the resistance change (34410A, Agilent Technologies). In contrast to diametric resistance, the van der Pauw method [165] can be used to determine the resistivity of disc-shaped thin films. However, the van der Pauw method requires at least two sets measurements for each state of stretch. Due to the large range of stretch that the electrodes have to undergo, we have deemed it more prudent to utilise diametric resistance to represent the stretchability of the electrode.

4.4 Stiffening effect

Applying a pair of electrodes onto an elastomer membrane stiffens it. DEAs with stiffer electrodes will require more force to deform, which translates to requiring a higher driving voltages than DEAs using compliant electrodes. In order to measure the stiffening effect that electrodes have on the elastomer, electrode-elastomer membrane composites were uniaxially and radially deformed by applying increasing amounts of uniaxial and radial forces, respectively.

4.4.1 Uniaxial deformations

The stiffness of a material has traditionally been determined by relating the stress required to uniaxially deform it. The samples used are the same as that described in section 4.3.1 and as shown in Figure 4.9. While the 500µm thick, 25 by 25mm samples were uniaxially stretched, at a rate of 3 millimetres per minute, the mechanical testing system (Instron 5565) logged the force and extension data. The uniaxial stress σ_{uni} in the electrode-coated elastomer membrane can be calculated, by using equation (3.19), from the logged force F and extension Δl

$$\sigma_{uni} = \frac{F}{w_o t_o} \lambda = \frac{F}{w_o t_o} \left(\frac{l_o + \Delta l}{l_o} \right) \quad (4.1)$$

where λ is the stretch in the length direction and l_o , w_o and t_o are the electrode-coated elastomer's original length, width and thickness, respectively. With increasing uniaxial deformation, the stress induced in the elastomer increases, as shown in Figure 4.11. By using

the stress-stretch behaviour of plain VHB elastomer as a baseline, one can see that sputtered silver electrodes stiffen the elastomer more than do graphite electrodes.

A measure of stiffness is the Young's modulus, which can be determined from the gradient of the stress-stretch curves in the linear region. Alternatively, the curves can be fitted to a hyperelastic model and the Young's modulus approximated from the fitted material parameters. Either way, it should be noted that this Young's modulus represents the stiffness of the electrode-coated elastomer composite and not the stiffness of the electrodes itself. This means that the quantitative results of this experiment would be applicable only to models using the same electrode-thickness to elastomer-thickness ratio. In addition, as the electrodes of DEAs are typically applied after the elastomer has been prestretched, while the electrodes were applied to the elastomer in its undeformed state for these mechanical tests, the Young's modulus of each electrode-coated elastomer composite cannot be applied directly to model the response of prestretched DEAs that undergo biaxial deformations.

Figure 4.11. Stress versus uniaxial stretch curves for VHB elastomers coated with graphite and sputtered silver electrodes.

4.4.2 Radial deformations

Radial deformations of the electrode-elastomer composites are studied in order to better determine their stress-stretch behaviours in a prestretched DEA configuration. As such, the samples were prepared to emulate the response of those used in the membrane DEAs. Bare VHB F9473PC elastomers were radially stretched by 2.5 times and secured onto acrylic

frames. Following that, electrodes were coated onto the prestretched elastomers to form 40mm diameter electrodes that were relatively stress-free. The radial stiffness measurement jig used is similar to that described in Section 4.3.2. However, in this case, as the elastomer has to be deformed by a measurable force, the motor was replaced by an eight-pulley system linked to dead weights, as shown in Figure 4.12. Sixteen loading points, as was used in the stretchability tests, would result in a more uniform deformation of the elastomer than the eight loading points used here. However, due to the additional force required to overcome the clamping forces of the clips, in order to stretch the clipped portions of the elastomer, the use of sixteen clips would make the elastomer appear to be stiffer than it actually is. As such, eight loading points was used in the radial deformation stiffness tests.

In order to load an electrode-coated, prestretched elastomer onto the jig, the sample was carefully peeled off from the acrylic frame and secured to the crocodile clips. The transfer was carried out one point at a time, until all eight clips were secured to eight equi-distant points on the circumference of the elastomer. As each clip was secured, equal amounts of weights were added to each of the eight cords to compensate the tension that was provided by the frame, thereby allowing the elastomer to remain radially stretched by 2.5 times. Subsequently, in order to deform the elastomer and the electrodes, weights were added, at a rate of 2.6 grams per minute, to each of the eight crocodile clips. At least three samples were tested for each type of electrode and the average was used for comparison.

Figure 4.12. (top) a homemade radial stiffness measurement jig that utilises constant-force dead weights to radially stretch the elastomer. (bottom) schematic of the cross section of the radial stiffness measurement jig.

The radial stress σ_{rad} in each electrode-elastomer membrane can be determined by finding the force acting on a unit area of the elastomer, with a thickness of t and a diameter of D , and taking into account equation (3.17),

$$\sigma_{\text{rad}} = \frac{\sum F}{\pi D t} = \frac{\sum F}{\pi D_o t_o} \lambda \quad (4.2)$$

where t_0 and D_0 are the electrode-elastomer membrane's initial thickness and initial diameter. As stretch ratio $\lambda = D/D_0$ increases, radial stress increases, as shown in Figure 4.13.

Figure 4.13. Stress versus radial stretch curves for VHB elastomers coated with graphite and sputtered silver electrodes.

A simple measure of the electrode-coated VHB membrane's stiffness is its radial tangential modulus E_{rad} , which is calculated from the gradient of its stress-stretch curve,

$$E_{\text{rad}} = \frac{\Delta\sigma_{\text{rad}}}{\Delta\lambda} \quad (4.3)$$

in which $\Delta\sigma_{\text{rad}}$ is the radial stress increment over the stretch-ratio increment $\Delta\lambda$. The electrode's stiffening effect can be observed when the radial tangential modulus of each electrode-coated VHB membrane was compared against that of the plain VHB membrane, which was found to be 402kPa. Sputtered silver-coated VHB was three times stiffer than plain VHB, while graphite-coated VHB was only 8% stiffer than plain VHB.

4.5 Discussion on the stiffening effect of graphite powder

The high stiffening effect of the sputtered silver thin film electrodes on the elastomer is not surprising, due to its large Young's modulus. On the other hand, graphite powder is commonly perceived to have a negligible effect on the elastomer's stiffness. However, as shown in the stress-stretch curves in Figure 4.11 and Figure 4.13, this is not the case. The

graphite powder used has a particle size of $6.5\text{ }\mu\text{m}$, as shown in Figure 4.14, which is large compared to the 150 nm thick sputtered metal thin films. In the radial-stretch experiments, the prestretched VHB elastomer was $40\text{ }\mu\text{m}$ thick, while each layer of graphite powder electrode was about $10\text{ }\mu\text{m}$ thick. As a result, the thick graphite layer had an appreciable clamping effect on the dielectric elastomer, thereby adding an appreciable stiffening effect on the DEA composite.

Figure 4.14. Micrograph of TIMREX KS6 graphite powder [33].

5 ELD silver thin film electrodes for DEAs

Metals, such as gold and silver, are low-resistivity materials that can efficiently transport electrical charges with minimal losses. Due to its low resistivity, metals can be made very thin, while still maintaining a low sheet resistance. The use of metal thin films can help to reduce the bulk of the DEA, as compared to electrode materials of high resistivity, wherein thicker electrodes would have to be used in order to attain the same sheet resistance. A thinner film will also help to reduce the electrode's stiffening effect on the elastomer, as planar stiffness is directly proportional to the thickness of the metal thin film electrodes. However, as have been proven by the small actuated strains of DEAs using plain sputtered metal thin film electrodes [133, 136], it is evident that metal thin films are still not stretchable enough and/or are too stiff to allow large deformations of the elastomer.

Metal thin films can be patterned into meanders in order to improve its stretchability and to reduce its stiffness. However, doing so reduces the area-coverage of the elastomer, which results in reduced electrostatic pressure, given the same elastomer footprint; this would in turn limit the actuated strains of its DEAs [136]. High-resolution patterning via lithography can help to alleviate this loss of electrode-coverage, but this requires extra time and cost. In this chapter, we show that sub-micron cracking in ELD silver thin films helps to improve the actuated strains of its DEAs. This is mainly caused by two changes in the properties of the ELD silver thin films: a reduced stiffening effect on its elastomer substrate and an improved stretchability. These micro-cracks formed spontaneously during the fabrication of the ELD silver thin films and retain an almost full-coverage of the elastomer in the DEA's inactivated state. In comparison, sputtered silver thin films without micro-cracks are stiffer and not as stretchable, which results in its DEAs being able to attain only small actuated strains.

5.1 Electroless deposition of silver

Metal thin films of less than half a micrometre thick are usually fabricated by physical vapour deposition methods such as evaporation and sputter deposition [81]. Evaporation deposition involves the melting of metals at high temperatures in order to make their atoms evaporate and deposit onto the substrate, while sputter deposition utilises energetic gaseous ions to bombard a metal target, thereby causing the metal to sputter off the target's surface to coat the substrate [41]. Although these methods of depositing metal thin films are very popular in

microelectronics, they might not be as suitable for the manufacture of electrodes for DEAs: these fabrication processes occur in a vacuum at high temperatures and may therefore alter the properties of the substrate. In addition, most of these types of equipment have vacuum chambers that are quite small and might not be able to fit larger-scale DEA substrates. An alternative method for fabricating metal thin films is electroless deposition (ELD), which is a chemical reaction that does not require any external current and can be carried out under normal conditions [50]. Compared to physical vapour deposition methods, ELD is an inexpensive and simple procedure that does not require expensive clean room equipment. A common and widespread application of ELD is the making of mirrors, wherein films of silver are deposited by either pouring or spraying the chemicals onto sheets of glass [48]. Silver serves as a good material candidate for DEA electrodes, as it is stable in clean air and water under normal conditions, and has one of the lowest electrical resistivity amongst all metals. Although it is not as stable as gold, because silver can tarnish when reacting with the hydrogen sulphide in air, it is much less expensive. But more importantly, it is much less dangerous to fabricate ELD silver, as compared to fabricating ELD gold, because almost all ELD gold processes produce cyanide ions [125]. In this chapter, the use of ELD silver thin film electrodes in DEAs is explored by studying its electrical and mechanical properties, as well as its performance when used in membrane DEAs.

5.1.1 Electroless deposition

ELD silver films are formed by an autocatalytic process that involves the mixing of a silver salt with a reducing sugar, such that the silver ions are reduced to form solid silver. This is not unlike the chemical reactions that take place in the Tollens' test for aldehydes. For example, when the diammine silver complex $\text{Ag}(\text{NH}_3)_2^+$ is mixed with formaldehyde CH_2O , it gets reduced to aqueous ammonia NH_3 and elemental silver Ag , which precipitates out of solution. Simultaneously, the formaldehyde oxidises to formate ion CHOO^- ,

Although the basic principle is simple, the success of such autocatalytic deposition processes depends on a few important considerations [50]. Firstly, the surface needs to be properly

activated, in order for the silver to deposit onto the surface, otherwise colloidal silver will form instead. To do so, a sensitizer made out of stannous chloride SnCl_2 can be applied to the surface, both to speed up the precipitation of silver and to increase the adhesion of silver to the elastomer. Secondly, the concentrations of both the oxidizing and reducing agents need to be balanced, as a very quick reaction would result in the formation of colloidal silver. Thirdly, the optimal parameters that influence redox reactions, such as temperature and pressure, also have to be determined.

5.1.2 Fabrication process

In the present work, a three-part commercial silvering solutions set (HE-300) from Peacock Laboratories, Inc. was used. Although the exact compositions of the chemicals are unknown, the set basically comprises a diammine silver complex, a hydroxide activator and an aldehyde reducing agent. In addition, the No. 93 Sensitizing Solution, also from Peacock Laboratories, Inc., which contains stannous chloride, was used to activate the elastomer's surface prior to the silvering.

In order to attain a silver thin film of $150 \pm 30\text{nm}$ thickness, the following steps were carried out in room conditions (refer to Figure 5.1):

- i. Dilute the sensitizer with deionised (DI) water using a 1:30 ratio. Dilute each of the three-part silvering solutions, in separate beakers, using a 1:40 ratio.
- ii. Attach a mechanically prestretched dielectric elastomer onto a rigid frame.
- iii. Use a silicone PDMS mask to ensure that the chemicals only come into contact with the portion of the elastomer that is to be silvered.
- iv. Drip on the diluted sensitizer and slightly agitate the solution, such that it covers the entire surface that is to be silvered.
- v. Wait for 1 minute then discard the solution and wash with DI water. (When washing, one can determine that the elastomer's surface is hydrophilic if the water forms a film. If water droplets with large contact angles form, then the surface is hydrophobic and the surface wetting procedure has to be revised.)
- vi. Mix together equal amounts of each of the three parts of diluted silvering solutions in a separate beaker.
- vii / viii. Using a pipette, drip the mixed solution onto the wetted area and slightly agitate it in order to cover the entire masked out surface. After 1 minute, a silver film would have

formed on the surface of the elastomer and the chemicals can be discarded and the mask removed.

ix. Wash the silver film with DI water and then blow dry using cool compressed air.

Figure 5.1. ELD silver fabrication process.

It was observed that micro-cracks formed in the ELD silver thin films after the fabrication process. In order to deposit ELD silver thin films, silvering chemicals had to be left on the free elastomer films, which caused the films to sag. This in turn caused a tensile stress on the metal thin film, which had been deposited on the opposite side of the elastomer, thereby causing it to crack. Also, when the ELD silver films were dried using compressed air, the pressure from the air would deform the free elastomer film, thereby causing tensile stresses in the ELD silver thin films and causing them to crack. When the silvering was carried out on elastomers mounted on a rigid glass slide instead of being free to deform, cracks were not observable, as shown in Figure 5.2.

Figure 5.2. Difference in surface morphologies for ELD silver thin films deposited on VHB substrates that were a) mounted on a glass backing and b) affixed to a fixed frame and therefore free to deform where the films were deposited onto.

5.2 Morphology and cracks in ELD silver thin films

The micro-cracks present in the ELD silver thin films can help to improve its stretchability, as micro-cracked metal thin films adhered to elastomers have been used as stretchable electrodes [98, 101, 112, 62]. In addition to improving the stretchability of the electrodes, the micro-cracks are expected to also make it easier to stretch the ELD silver thin films.

The micro-cracks in the ELD silver thin films are copious and zigzag about the entire area as shown in Figure 5.3, with relatively small crack spacings ($L \sim 800\text{nm} - 9500\text{nm}$) and crack openings ($\delta \sim 140\text{nm}$). The network of cracks is two, if not three dimensional, as is evident in its highly textured surface shown in Figure 5.4. The zigzagging nature of the micro-cracks is a result of the ELD silver grains, which are small, of average grain size of 50nm , and equiaxed or globular, as shown in Figure 5.5. The grain boundaries act as dislocation barriers, forcing the cracks to propagate around them to form inter-granular fractures [169, 111]. The resultant formation of zigzagging micro-cracks means that that numerous conductive pathways will still exist, as shown in Figure 5.3, enabling the micro-cracked silver film to remain conductive. If the silver grain size were larger, cracks would be more likely to propagate through them to form channel cracks that would cut off all conductive paths to

render the silver film non-conductive. In addition, if cracks do not propagate through the grains, then plastic deformations are avoided, which leads to a lower film toughness [17]. In the field of stretchable electronics, only the stretchability of the conductor is of importance. Thicker ductile films can be stretched by more than 50% [112] and fail by first deforming plastically and then undergoing trans-granular fracture. Albeit highly stretchable, such films are stiff, unlike the more brittle nature of failure by inter-granular fracture [111], as experienced by small-grained metal films, such as our 150nm thick ELD silver. As having a low stiffness is just as important as having a high stretchability for DEA electrodes, thin micro-cracked metal films appear to be more suitable than thick ductile films.

Figure 5.3. Micrographs of the micro-cracks in the ELD silver electrodes with increasing magnification.

Figure 5.4. Topology of the undeformed ELD silver thin film's surface, as measured by a Confocal microscope.

Figure 5.5. The ELD silver thin film comprised silver grains that are small and equiaxed or globular.

In contrast to the micro-cracked ELD silver thin films are the sputtered silver thin films, which had no visible crack, as shown in Figure 5.6. It can be seen that the sputtered silver thin film is slightly wrinkled. These wrinkles formed due to the higher temperature in the growing metal film, as compared to the substrate during sputtering, which results in

compressive stresses in the metal film upon cooling [150]. Such wrinkling has been known to improve the stretchability of metal thin films [97, 82, 143, 171, 101, 100] and this is shown in the sputtered silver electrode's uniaxial stretchability of 21%. However, the extent of wrinkling in the sputtered thin films due to thermal mismatch is minimal and will flatten out after just a few percent stretch.

Figure 5.6. Surface morphology of silver thin film electrodes. Micro-cracks exist in the (left) ELD silver electrodes but not in the (right) sputtered silver electrodes.

5.3 Stretchability of ELD silver thin film electrodes

DEA electrodes have to be able to remain conductive even when highly stretched, as the electrode-covered area can expand to two times or more its original size during actuations. In order to determine the stretchability of the ELD silver electrodes, samples comprising ELD silver thin films on VHB elastomers were subjected to both uniaxial and radial stretches. In their undeformed states, the uniaxial samples were 25mm squares while the radial samples were 40mm-diameter circles. The resistance across the electrodes were monitored during the stretching process and were deemed to be non-conductive when a resistance of $1\text{G}\Omega$ was reached.

As shown in Figure 5.7, the resistance across the ELD silver electrode started low at about 10Ω and conductivity was lost at 33% uniaxial strain and 11% radial strain. The sputtered silver film electrode also had a low starting resistance of 1Ω , but its conductivity was lost at a lower uniaxial strain of 21%. In contrast to the metal thin film electrodes is the graphite powder electrode, which had a much higher starting resistance of about $100\text{ k}\Omega$, but managed to retain its conductivity up to much higher strains of 140% uniaxial strain and 55% radial strain.

Figure 5.7. (left) uniaxial stretch and (right) radial stretch tests show the change in resistance with respect to increasing strains.

The relatively high stretchability of the ELD silver thin film electrodes can be attributed to its micro-cracks. The small crack openings in the ELD silver films ensure that conductivity is maintained due to electron wave function delocalisation and tunnelling effects [62].

5.4 Stiffening effect of ELD silver thin film electrodes

DEA electrodes have to be mechanically compliant, such that they minimally inhibit expansions of the elastomer during actuations. In order to determine the stiffening effect that ELD silver thin film electrodes have on the elastomer, samples comprising ELD silver thin films on VHB elastomers were subjected to both uniaxial and radial stretches. As shown in Figure 5.8, for both uniaxial and radial deformations, it is qualitatively apparent that the VHB elastomer was stiffened the least by graphite powder, followed by ELD silver thin films, and then sputtered silver thin-films. This order coincides with the stretchability tests, wherein

graphite could be stretched the most, followed by ELD silver and then sputtered silver. For uniaxial deformations, the extent to which each electrode stiffened the elastomer was quantified by approximating the Young's Modulus of each electrode-coated VHB 4905 sample using the Mooney-Rivlin model [119]. The Young's Modulus of a 500 μm -thick VHB elastomer coated with graphite, ELD silver and sputtered silver was 1.03, 1.20 and 1.58 times that of an uncoated VHB elastomer (156kPa). As for radial deformations, the stiffening effect of the electrode on the VHB elastomer was quantified by comparing the radial modulus of each electrode-coated elastomer with the radial modulus of the plain elastomer, as discussed in section 4.4.2. The radial modulus of a 250mm-thick VHB coated with graphite, ELD silver and sputtered silver was 1.08, 2.6 and 3 times the radial modulus of the uncoated VHB elastomer (402kPa). Although this type of quantification makes it easy to observe the stiffening effect of the different electrode-types, it should be noted that they indicate the stiffness of the electrode-coated elastomer composite and not the stiffness of the electrodes itself.

Figure 5.8. Stress-strain curves of electrode-covered VHB elastomer composites that have undergone (left) uniaxial and (right) biaxial deformations.

In the stress-stretch relationship for the VHB elastomers coated with ELD silver thin films, it is apparent that there is a gradual reduction in gradient with increasing stretch. This suggests a reduction in the electrode's stiffness. If we look at the morphology of the micro-cracked ELD silver thin films, as shown in Figure 5.9, we can see that the crack spacings and crack openings in the thin films increase with increasing stretch. The reduction in the crack

spacings in the silver thin films results in a smaller effective modulus of the DEA composite [16], which is beneficial for attaining large actuated strains.

Figure 5.9. Widening of the micro-cracks in an ELD silver film when uniaxially strained by 30%.

5.5 Electromechanical activation of DEAs using ELD silver thin film electrodes

In this section, the actuated strains in membrane DEAs using ELD silver thin film electrodes is studied, as they are subjected to differing amounts of prestretch and ELD silver film thickness.

5.5.1 Performance with respect to other electrodes

Membrane DEAs using ELD silver thin film electrodes can attain actuated area strains of 50%, as shown in Figure 5.10. By supplying the DEA with a driving voltage of 9kV, the region covered by the ELD silver thin films expand by 50%, so much so that the electrodes become less opaque.

Figure 5.10. An actuated area strain of 50% was attained by increasing the voltage supplied to a membrane DEA, using ELD silver thin film electrodes, from a) 0V to b) 9kV ($337\text{V}\mu\text{m}^{-1}$).

This actuated strain is high compared to DEAs using sputtered silver thin films of the same thickness, but low when compared to DEAs using graphite powder electrodes, as shown in Figure 5.11. As driving voltage was increased, the actuated strain for DEAs using ELD silver electrodes increased at a decreasing rate. From 7.5kV, actuated strains plateau and do not increase with increasing voltage. This can be attributed to the high planar stiffness that metal thin film electrodes have as compared to graphite powder electrodes. As such, the maximum actuated area strain of DEAs with ELD silver electrodes was about half that of DEAs with graphite electrodes. Despite this, the breakdown voltage and breakdown field strength of DEAs using ELD silver electrodes was more than three times that of DEAs using graphite powder electrodes. This means that DEAs using ELD silver can generate higher forces than DEAs using graphite powder electrodes as electrostatic force is proportional to the square of the electric field.

Figure 5.11. Relationship between actuated area strain and driving voltage for single film DEAs. Five samples were tested for DEAs using ELD silver electrodes and error bars indicate the standard deviation.

5.5.2 Effect of elastomer prestretch on actuated strains

The effect of elastomer prestretch on the actuated strains of DEAs using ELD silver electrodes is studied here. The amount of prestretch in the elastomer would affect the actuated strains in the DEA, wherein a lower prestretch would require more voltage to attain the same actuated strain while a higher prestretch would reach the breakdown field earlier. As shown in Figure 5.12, this proves to be true. In order to attain an actuated strain of 35%, DEAs with a biaxial prestretch of 2 requires a driving voltage of 7kV while that with a prestretch of 4 requires 4kV. Also, while DEAs with a biaxial prestretch of 2 and 2.5 allow driving voltages of more than 10kV, that with a prestretch of 4 breaks down at a driving voltage of 6.5kV. For DEAs using ELD silver thin film electrodes and VHB F9473PC elastomers that are 250 μ m thick in its undeformed state, it appears that a biaxial prestretch of 2.5 yields the best actuated strains.

Figure 5.12. Effect of elastomer prestretch on actuated strains of DEAs using ELD silver thin film electrodes.

5.5.3 Self-clearing

The self-clearing ability of ELD silver thin films is a major contributing factor to the high breakdown fields that its DEAs can attain. Due to self-clearing, premature failures of the DEA can be circumvented, even allowing the use of higher driving voltages. It does not, however, modify the properties of the elastomer as even higher breakdown field strengths ($450 \text{ V}/\mu\text{m}$) has been obtained by simply immersing the DEA in a dielectric liquid [96]. Due to defects in the dielectric layer, such as voids or impurities, a localised breakdown, which is characterised by a surge in current together with a drop in voltage, can bring about a short circuit. However, due to the self-clearing ability of ELD silver electrodes, a transient arc discharge will vaporise the metal electrode in the neighbourhood of the defect, thereby making the region non-conductive [160]. Figure 5.14 shows the time histories of current and voltage as the driving voltage was increased in steps of 500V. The first self-healing event occurred at 5kV, as characterised by a spike in the current and a corresponding drop and recovery of the voltage. This phenomenon was not observed in DEAs using graphite electrodes. At higher electric fields, leakage current increased steadily.

Figure 5.13. Self-clearing in DEAs using ELD silver electrode, characterised by a recovery following a plunge in voltage, together with a corresponding spike in current.

A self-cleared site is shown in Figure 5.14, wherein a pinhole is burnt through the dielectric layer, while the surrounding region of electrode is oxidised, thereby making the site inert. However, this results a smaller active electrode area, thereby causing the performance of the actuator to drop. Therefore, while self-clearing is a remarkable property that can allow larger strains, it should be employed only as a safety feature in preventing terminal failure. In other words, when designing an actuator, is it best to have the operating range below voltages at which breakdowns tend to occur. Alternatively, like in the manufacturing procedures for metalized plastic capacitors, the DEA can be aged before functional use. This involves the deliberate burning off of the dielectric layer's points of weakness by increasing the applied voltage slowly till an appropriate value. Aging can enable a more reliable and predictable performance of the DEA.

Figure 5.14. A self-cleared site in a DEA with ELD silver electrodes.

5.6 Discussion on ELD silver electrodes

ELD silver thin films are suitable electrodes for DEA applications requiring moderate strokes. In addition, due to the high electric fields of more than $375\text{V}/\mu\text{m}$ attained by its DEAs, there is potential for large actuated forces. While piling in comparison to the more conventional DEA electrodes such as graphite powder, DEAs using ELD silver thin film electrodes were able to attain significantly higher actuated strains than DEAs using sputtered metal thin films, as shown in Table 3. At a voltage of 2kV, DEAs using ELD silver thin film electrodes managed to attain an actuated area strain of 20%. As the other works in literature make use of silicone elastomers as their dielectric layer, while the present work makes use of a polyacrylate elastomer, a direct comparison is not possible. However, when ELD silver thin film electrodes were applied onto a silicone elastomer, the DEA managed to attain actuated area strains of 21% at a driving voltage of 2.5kV [109]. Although the current setup uses unpatterned silver thin films, it is still able to attain higher strains than the DEAs using electrodes that have been patterned to improve its stretchability and to reduce its stiffness. As such DEAs using ELD silver thin film electrodes appear to have greater potential for larger actuated strains than DEAs using sputtered metal thin films.

Table 3. Comparison between actuated strains of membrane DEAs using metal thin film electrodes.

	Pelrine et al., 1998	Pimpin et al., 2007	Pimpin et al., 2007	Benslimane et al., 2009	Previous work 2011	Present Work
Dielectric Material	PDMS	PDMS Sylgard 186, Dow Corning	PDMS Sylgard 186, Dow Corning	PDMS Elastosil RT625, Wacker	PDMS TC5101, BJB Enterprises	Polyacrylate VHB F9473PC, 3M
Dielectric Thickness	50 μ m (unknown)	33 μ m (no prestretch)	33 μ m (no prestretch)	40 μ m (no prestretch)	80 μ m (minimal prestretch)	40 μ m (prestretch of 2.5)
Actuator Size	<i>unknown</i>	2 mm diameter	2 mm diameter	31 by 51 mm	10 mm diameter	10 mm diameter
Electrode Material	Sputtered gold thin film	Sputtered gold thin film	Sputtered gold thin film	Sputtered silver thin film	ELD silver thin film	ELD silver thin film
Electrode Thickness	< 100 nm	Top: 150 nm Bottom: 130 nm	Top: 150 nm Bottom: 130 nm	100 nm	250 nm	150 nm
Electrode pattern	Un-patterned	Un-patterned	Concentric circle pattern	Corrugated in the actuation direction	Un-patterned	Un-patterned
Maximum Strain	4% area strain	1.7% area strain @ 2kV	5.6% of the actuator diameter @ 2kV	10% linear strain @ 3.6 kV	21% area strain @ 2.5kV	20% area strain @ 2kV 56% area strain @ 9.5kV

The ELD silver thin films are stretchable up to 33% uniaxial strain and 11% radial strain, with resistances more than two orders lower than that of graphite powder, within that range. Its stiffening effect on the dielectric elastomer is not as strong as that by the sputtered silver thin film electrodes of the same thickness. This is due to the network of micro-cracks in the ELD silver thin films that lowers its planar stiffness, while maintaining conductivity. The ELD silver thin films, like other metal thin film electrodes, are able to self-clear, which enabled relatively high breakdown fields of up to 570 V/ μ m in VHB DEAs that had been biaxially prestretched by 4 times. Apart from improving the breakdown field and actuated strain of the DEA, self-clearing electrodes are very useful for preventing catastrophic failures that give no warning.

A major advantage of using ELD silver thin film electrodes is that it is a relatively inexpensive procedure, especially when compared to other methods that require clean room equipment, such as sputtering and evaporation. In addition, as this method is commonly used to silver mirrors of large areas, there is potential for fabrication of the electrodes at a larger scale. Conversely, patterning in the micro-scale has been possible via micro-contact printing [177]. Our group has attempted micro-contact printing of ELD silver and found that widths of down to $200\mu\text{m}$ is possible, as shown in Figure 5.15.

Figure 5.15. Micro-contact printing of ELD silver on a VHB elastomer. (left) overall pattern. (right) magnification of one segment of the pattern, with a width of a $200\mu\text{m}$.

6 Stiffness analysis of buckled metal thin films

Flat metal thin film electrodes are stiff relative to its elastomer substrate and hence restrict the elastomer's deformations. Corrugations have been shown to be able to make silver thin films more compliant and stretchable [163]. However, corrugations only allow uniaxial deformations of the elastomer. In order to fully maximise electrostatic force generation and therefore actuated strains in a DEA, like conductive greases, the metal thin film electrodes should be biaxially compliant and stretchable. Currently, biaxially stretchable metal thin film electrodes do not appear to be compliant enough and have not been able to demonstrate large actuations in DEAs [72, 181, 182, 22, 46, 136]. We intend to make metal thin films as biaxially compliant as graphite powders by buckling the metal thin films on an elastomer substrate. In doing so, we aim for DEAs using these biaxially buckled or crumpled ELD silver thin film electrodes to achieve large actuated strains that are similar to that achieved by DEAs using graphite powder electrodes. In this chapter, we conduct a basic feasibility study of biaxially buckled metal thin film electrodes by using structural mechanics theories to determine its stiffening effect on the dielectric elastomer. Following that, we predict the actuated strains of DEAs using such buckled metal thin film electrodes by making use of experimentally-obtained material parameters.

6.1 Overview of wrinkling

Wrinkling is a common phenomenon that occurs in everyday life, of which one of the most relatable examples is the increasingly marked appearance of wrinkles in our faces as we get older. As skin ages, the numerous but fine wrinkles in youthful skin turn into few but deep wrinkles that appear to be folds [11, 95]. Common cosmetic methods for combatting the deepening of wrinkles include the application of a moisturiser on the skin's surface, in order to hydrate the outer most layer of skin (stratum corneum), and the injection of botulinum toxin A, better known as Botox, under the wrinkled layer of skin, in order to firm up the supporting tissues (living epidermis and upper dermis). This is because the stratum corneum is known to become drier with age, which results in an increase in its stiffness [126], while the living epidermis and upper dermis become less stiff with age, due to the alteration of collagen bundles [11]. Simplistically explained, it is because of the increasing mismatch

between the Young's modulus of the stiffening outer skin layer and the softening inner skin support that skin wrinkling becomes more pronounced with age.

One of the simplest models that has been used to study the phenomenon of wrinkling is that of a thin stiff film, of thickness h , supported by a soft substrate of infinite thickness [4], as shown in Figure 6.1. Referring back to the example of facial skin, the stiff film is analogous to the outer skin while the soft substrate is analogous to the inner skin support. When the compressive force P reaches a critical value, there is a loss of mechanical stability of its pre-buckling equilibrium state, resulting in the bifurcation into a buckled equilibrium state [73], of amplitude ζ and half-wavelength l . In accordance with Euler's observations regarding the buckling of columns [15], the magnitude of compressive force required to cause the supported film to buckle is generally dependent on its Young's modulus, geometric dimensions and boundary conditions. In this case, the boundary conditions are imposed by the supporting substrate.

Figure 6.1. Schematic of the wrinkling of a long thin film supported by a soft substrate, in its a) pre-buckled state and in its b) buckled configuration.

The forces acting on the film include its internal elastic stresses, as a result of its bending, the external compressive axial load P and the reactive lateral stress σ_z from the substrate due to its deformation, see Figure 6.1b, wherein

$$D \frac{d^4 w}{dx^4} + P \frac{d^2 w}{dx^2} = b \sigma_z \quad (6.1)$$

where D is the flexural rigidity, b the width and w the vertical displacement in the z direction of the film, which can be approximated by a sinusoidal wave [4, 157, 22] with a half-wavelength of l and an amplitude of ζ .

Corresponding to the critical load is the critical strain, at which on the onset of buckling occurs. Assuming that the substrate is an incompressible elastomer, the critical strain depends on just the ratio between the Young's modulus of the film E_f and of the substrate E_s [22], approximated by

$$\varepsilon_{\text{crit}} = 0.52 \left[\frac{E_s}{E_f} \right]^{\frac{2}{3}} \quad (6.2)$$

The resultant buckled film will have a critical half-wavelength l_{crit} that is approximately [4, 84]

$$l_{\text{crit}} = \pi h \left[\frac{E_f}{3E_s} \right]^{\frac{1}{3}} \quad (6.3)$$

It can be seen that the larger the mismatch between the Young's modulus of the film and substrate, the smaller the critical strain and the larger the resultant wavelength. This matches the phenomenon regarding aging skin, wherein wrinkles are fewer and more spaced out in old skin than in young skin, as a result of the outer skin layer becoming stiffer and the supporting tissues becoming softer.

Skin wrinkling occurs when a person frowns, wherein the contractions of the underlying muscles result in compressive axial forces acting on the skin. When the degree of contraction of the underlying muscles intensifies, gentle wrinkles turn into deep furrows. In the post buckling regime, wherein the applied compressive strain is larger than the critical strain, the geometrically nonlinear behaviour of the wrinkles have to be taken into account [42].

For compressive strains that are only slightly higher than the critical strain, the wavelength remains at its critical value l_{crit} , while the amplitude ζ of the wrinkles increases with strain ε , given by [84]

$$\frac{\zeta}{l_{crit}} = \frac{2\sqrt{1+\nu_f}}{\pi} \sqrt{|\varepsilon| - \varepsilon_{crit}} \quad (6.4)$$

where ν_f is the Poisson's ratio of the film, while l_{crit} and ε_{crit} are defined by equations (6.5) and (6.4) respectively.

Subsequently, at larger compressive strains, wrinkle amplitude continues to increase while wrinkle wavelength starts to decrease [24]. This trend continues up until the wrinkles become unstable, resulting in morphological changes in the wrinkles, such as period-doubling bifurcations involving the nonlinear coupling of two modes [24] and a hierarchical pattern comprising self-similar buckles of varying wavelengths [52]. In the latter study, it is said that the growth of the amplitude of the initially-formed wrinkles will saturate due to the nonlinear effects from the stretching and shearing of the substrate. The composite, comprising the buckled film and the affected portion of the stretched substrate, will form the effective film for the next hierarchy of wrinkles. As this effective film will be thicker and stiffer than the original film, the wavelength of the successive hierarchy of wrinkles will be larger than that of its preceding hierarchy of wrinkles to give nested wrinkles.

6.2 Apparent stiffness of buckled thin films

A low planar stiffness of electrodes is essential for a DEA to attain large actuations. As it is easier to bend a film than to stretch it, it is expected that the planar stiffness of a buckled thin film is lower than the planar stiffness of a flat film. As discussed in section 6.1, the morphology of the wrinkling of a thin film depends on many factors, such as material properties, boundary conditions and loading conditions, and is therefore difficult to predict. This is especially so when the substrate is viscoelastic [84] or when compressive strains are large [24], resulting in complex morphologies such as nested wrinkles [52]. In this section, we ignore the effects of the substrate by stripping down the model to just a thin film that buckles under uniaxial and biaxial compressive stresses. In order to quantify the planar stiffness of the buckled thin film, the amount of tensile forces required to flatten the

buckled thin film is determined via simple analyses using structural plate theory [157]. Doing so enables us to show that the apparent stiffness of uniaxially and biaxially buckled thin films is lower than that of flat thin films.

6.2.1 Uniaxial buckling

The apparent axial modulus of a corrugated thin film can be estimated by considering a representative strip of thin film, which is subjected to a pair of compressive forces per unit length, N_x . The lateral (z-direction) displacement w of the buckled thin film is assumed to have a sinusoidal profile [157]:

$$w = \zeta \sin\left(\frac{\pi x}{l}\right) \quad (6.5)$$

in which ζ is the amplitude of the corrugation and l is the half-wavelength of the corrugation, along the x-axis, as shown in Figure 6.2b.

Using a small-displacement approximation, the arc length l_0 of the buckled thin film is

$$l_0 = \int_0^l \sqrt{1 + \left(\frac{dw}{dx}\right)^2} dx \approx \left(1 + \frac{\pi^2 \zeta^2}{4l^2}\right) l \quad (6.6)$$

Figure 6.2. Dimensions and deformations of a (a) flat (b) uniaxially buckled and (c) biaxially buckled thin-film.

Assuming that there is negligible axial strain in the middle plane of the thin film, the buckled thin film bends from its original length of l_0 to get packed into a shorter half-wavelength l .

Hence, the apparent axial strain is defined as the ratio of half-wavelength reduction over the arc length, given by

$$c_x = \frac{l_o - l}{l_o} = \frac{\pi^2 \zeta^2}{4l^2 + \pi^2 \zeta^2} \quad (6.7)$$

By assuming small deflections, the strain energy U in the thin film is equal to the bending energy

$$U = 0.5D \int_0^1 \int_0^1 \left(\frac{\partial^2 w}{\partial x^2} \right)^2 dx dy = \frac{lb}{4} D \zeta^2 \frac{\pi^2}{l^2}$$

wherein D is the flexural rigidity of the film, given by

$$D = \frac{Eh^3}{12(1 - \nu^2)} \quad (6.8)$$

The work done T by the axial force per unit length N_x to buckle the thin film is given by

$$T = 0.5 \int_0^b \int_0^1 N_x \left(\frac{\partial w}{\partial x} \right)^2 dx dy = \frac{lb}{4} N_x \zeta^2 \frac{\pi^2}{l^2}$$

By using the principle of virtual work, wherein

$$\frac{\partial U}{\partial \zeta} \delta \zeta = \frac{\partial T}{\partial \zeta} \delta \zeta$$

the critical compressive force per unit length is

$$N_x = \frac{Eh^3}{12(1 - \nu^2)} \left(\frac{\pi^2}{l^2} \right) \quad (6.9)$$

wherein h is the film thickness, while E and ν are the Young's modulus and Poisson's ratio of the thin film, respectively.

Assuming elastic deformation, the compressive forces required to buckle the thin film is equal to the tensile forces required to flatten it. The apparent Young's modulus is therefore the ratio of the compressive axial stress over the apparent strain:

$$E_{x,\text{uniaxial}} = \frac{N_x}{hc_x} = \left(\frac{\pi^2}{12(1-\nu^2)} \right) \left(\frac{h}{l} \right)^2 \left(\frac{1}{c_x} \right) E \quad (6.10)$$

Using equation (6.10) and noting that the thin film's thickness is typically in the nano-scale, while the wavelength is typically in the micro-scale [82, 156], it can be seen from Figure 6.3 that the apparent axial Young's modulus of a uniaxially buckled thin film is lower than that of a flat thin film.

Figure 6.3 Change in the apparent axial modulus of the thin film (normalised by the Young's modulus of the material) due to uniaxial and biaxial compressions for a given half-wavelength.

Although uniaxial buckling is able to make thin films more compliant in one direction (along the x-axis), it stiffens the thin film in the transverse direction (along the y-axis). In fact, in large structures, corrugated plates are often used for their enhanced stiffness along the y-axis. Due to an increased cross-sectional area over the same half-wavelength, the uniaxially buckled thin film has a transverse apparent Young's modulus of

$$E_{y,\text{uniaxial}} = \left(\frac{1}{1 - c_x} \right) E \quad (6.11)$$

which is greater than that for the flat thin film as $1 - c_x$ is less than one.

6.2.2 Biaxial buckling

Unlike uniaxial buckling, which makes a metal thin film more compliant in one direction but stiffer in the other, biaxial buckling makes a metal thin film equally compliant in both the axial and transverse directions. The apparent axial modulus of a biaxially buckled thin film is estimated by considering a representative square strip of metal thin film, which is compressed biaxially by two orthogonal pairs of equal forces per unit length, i.e. $N_x = N_y$. The lateral (z -direction) displacement w of this biaxially buckled thin film is assumed to be [157] (see Figure 6.2c)

$$w = \zeta \sin\left(\frac{\pi x}{l}\right) \sin\left(\frac{\pi y}{l}\right) \quad (6.12)$$

wherein l is the identical half-wavelengths along the orthogonal axes of x and y .

By assuming small deflections, the strain energy U in the thin-film is equal to the bending energy

$$U = 0.5D \int_0^b \int_0^l \left(\frac{\partial^2 w}{\partial x^2} + \frac{\partial^2 w}{\partial y^2} \right)^2 dx dy = \frac{lb}{8} D \zeta^2 \left(\frac{\pi^2}{l^2} + \frac{\pi^2}{b^2} \right)^2$$

The work done T by the axial forces per unit length N_x and N_y to buckle the thin film is given by

$$T = 0.5 \int_0^b \int_0^l N_x \left(\frac{\partial w}{\partial x} \right)^2 + N_y \left(\frac{\partial w}{\partial y} \right)^2 dx dy = \frac{lb}{8} \zeta^2 \left(N_x \frac{\pi^2}{l^2} + N_y \frac{\pi^2}{b^2} \right)$$

By using the principle of virtual work, wherein

$$\frac{\partial U}{\partial \zeta} \delta \zeta = \frac{\partial T}{\partial \zeta} \delta \zeta$$

and by assuming that $N_x = N_y$, the force per unit length required to buckle the thin film is given by

$$N_x = 4D \left(\frac{\pi^2}{l^2} + \frac{\pi^2}{b^2} \right)$$

By assuming a square plate wherein $b = l$,

$$N_x = 2D \left(\frac{\pi^2}{l^2} \right)$$

As such, the equi-biaxial compressive force per unit length is

$$N_x = \frac{Eh^3}{6(1-\nu^2)} \left(\frac{\pi^2}{l^2} \right) \quad (6.13)$$

Again, assuming elastic deformation, the compressive forces required to buckle the thin film is equal to the tensile forces required to flatten it. Hence, the apparent axial modulus in the x axis can be calculated as the ratio of the apparent axial stress to the apparent strain, following [157]:

$$E_{x, \text{biaxial}} = \frac{N_x - \nu N_y}{hc_x} = \left(\frac{\pi^2}{6(1+\nu)} \right) \left(\frac{h}{l} \right)^2 \left(\frac{1}{c_x} \right) E \quad (6.14)$$

Due to symmetry and equi-biaxial loading, the axial moduli along the two orthogonal directions are equal, such that

$$E_{y, \text{biaxial}} = \frac{N_y - \nu N_x}{hc_y} = E_{x, \text{biaxial}} \quad (6.15)$$

As seen in Figure 6.3, like uniaxially buckled thin films, the axial apparent Young's modulus of a biaxially buckled thin film is also lower than the Young's modulus of a flat thin film. However, when the thin film is biaxially buckled, its stiffness is reduced in both the axial and transverse directions; this is in contrast to uniaxially buckled thin films, wherein stiffness is

increased in the transverse direction. As such, biaxially buckled metal thin films are suitable for use as biaxially compliant electrodes for DEAs.

6.3 Stiffening effect of buckled thin films

In this section, we aim to determine the stiffening effect that buckled thin film electrode have on its elastomer substrate. By doing so, we can determine its suitability for use as compliant electrodes in DEAs. By using the apparent Young's modulus of the buckled thin films, which had been derived in the previous section, we can apply the rule of mixtures to determine the Young's modulus of the composite comprising both the electrodes and its elastomer substrate, such as that shown in Figure 6.4.

Figure 6.4. Schematic showing the composite comprising buckled thin film electrodes and a dielectric elastomer.

For the dielectric elastomer, let us assume that it has a Young's modulus of $E_{VHB} = 156\text{kPa}$ (see section 0) and a thickness of $Z = 40\mu\text{m}$, as used in the DEA configurations. As for the metal thin films, let us assume that it has a Young's modulus of $E_{ELD} = 52\text{MPa}$ (derived using the data from section 0¹, which is lower than the bulk Young's modulus of silver due to the

¹ From uniaxial stretch tests, $Z = 500\mu\text{m}$, $H = 300\text{nm}$, $E_{VHB} = 156\text{kPa}$ and the ELD silver coated VHB composite, $E_{ELDcomp} = 187\text{kPa}$. Using the rule of mixtures, the Young's modulus of the micro-cracked ELD silver E_{ELD} can be determined by

$$E_{ELD} = \left(\frac{Z+H}{H}\right) E_{ELDcomp} - \left(\frac{Z}{H}\right) E_{VHB} = 52\text{MPa}$$

presence of micro-cracks), a Poisson's ratio of 0.37, a thickness of $h = 150\text{nm}$ and a compression of $c_x = 0.4$. The half-wavelength l of a crumpled ELD silver thin film, which had been biaxially compressed by 40% of its original length, is between of $l \sim 400\text{nm} - 1250\text{nm}$, as shown in Figure 6.5.

Figure 6.5. Morphology of a biaxially crumpled ELD silver thin film. l is the half-wavelength of the buckles, and $2l$ the peak to peak wavelength.

By substituting these values into equation (6.16), we get the apparent Young's modulus of a biaxially buckled ELD silver thin film E_{buckled} to be

$$E_{\text{buckled}} = \left(\frac{\pi^2}{6(1 + \nu)} \right) \left(\frac{h}{l} \right)^2 \left(\frac{1}{c_x} \right) E_{\text{ELD}} \quad (6.16)$$

$$E_{\text{buckled}}(l = 400\text{nm}) = 21.9\text{MPa}$$

$$E_{\text{buckled}}(l = 1250\text{nm}) = 2.25\text{MPa}$$

Due to the buckling of the thin film, the effective thickness of the electrodes increases, to give a total effective thickness of H for the two electrodes. By substituting $l = 400\text{nm}$ & 1250nm and $c_x = 0.4$ into equation (6.7), we find that $H(l=400\text{nm}) = 416\text{nm}$ and $H(l=1250\text{nm}) = 1300\text{nm}$. Therefore, using the rule of mixtures, the Young's modulus of the buckled ELD silver thin film on VHB composite E_{comp} is given by

$$E_{\text{comp}} = \left(\frac{Z}{Z + H} \right) E_{VHB} + \left(\frac{H}{Z + H} \right) E_{\text{buckled}} \quad (6.17)$$

$$E_{\text{comp}}(l = 400nm) = 410kPa = 2.2E_{VHB}$$

$$E_{\text{comp}}(l = 1250nm) = 252kPa = 1.3E_{VHB}$$

From this, we can expect the Young's modulus of the crumpled ELD covered VHB elastomer composite to be between 1.3 to 2.2 times the Young's modulus of the VHB elastomer. This is compared to un-buckled ELD silver thin films, whose composite would have a Young's modulus that is 3.5 times the Young's modulus of a VHB elastomer of similar geometry.

6.4 Modelling of DEAs with biaxially crumpled ELD silver thin films

Due to the complexity of the morphology of crumpled ELD silver electrodes, it is difficult to theoretically determine their stiffness. In this section, we aim to make use of experimentally-determined electrode stiffness properties in order to predict the electromechanical response of a DEA. The stiffness properties of electrode-coated VHB elastomers were experimentally measured via tensile deformations, as discussed in chapter 4. As will be discussed in chapter 7, crumpled ELD silver electrodes are formed by allowing the silver film to buckle on the VHB elastomer. This buckling causes the silver thin film to fold into the VHB elastomer, as well as to pull the VHB elastomer upwards. As such, the effective electrode comprises of both the buckled silver thin film as well as the elastomer in its vicinity. Here, we extract the mechanical stiffness of the effective electrode and then make use of the model discussed in section 3.4 in order to predict the DEA's actuated strains. The main assumptions made are that the electrodes are highly stretchable and are able to retain a full coverage of the stretched elastomer throughout the deformation process. In addition, it is assumed that the VHB elastomer is perfectly incompressible, which implies that a biaxial area expansion is equal to a uniform thickness compression, and that it is a perfect dielectric that does not conduct electricity.

6.4.1 Material parameter extraction

In order to model the electromechanical actuations of membrane DEAs, one can make use of the various hyperelastic constitutive models to describe the elastomer's deformations. To do

so, material properties have to first be determined from the uniaxial and/or radial stretch-stress behaviours of the various electrode-elastomer composites. Figure 6.6 shows the uniaxial stress-stretch curves of 500 μ m-thick VHB 4905 strips coated on both sides with graphite, sputtered silver thin film, ELD silver thin film and crumpled ELD silver thin film electrodes. The end of each curve marks the uniaxial strain at which the electrode ceased to be electrically conductive. All of the elastomer-electrode composite strips experience zero stress at a stretch of one, except for the crumpled ELD silver thin film composite. This is because the compressed silver thin film exerts tensile stresses on the elastomer, causing the elastomer to be stretched by 1.05 times even without any external stress applied.

Figure 6.6. Amount of mechanical stress required to uniaxially stretch the different types of electrode-coated VHB strips, up until each electrode lost its conductivity. For the crumpled ELD silver electrodes, there is a 1.05 pre-stretch in the VHB strips due to the compressed silver films.

Here, the stiffening effect that each electrode has on the dielectric elastomer can be perceived both visually from the stress-strain curves and by comparing the shear moduli shown in Table 4. These values were calculated by first fitting each curve to the one parameter Ogden model [173] by using MATLAB's nonlinear curve-fitting function (lsqcurvefit). Following that, the shear modulus was calculated from the parameters obtained. It was found that sputtered silver thin films stiffened the elastomer the most (by 69%), followed by ELD silver thin films (by 20%), crumpled ELD silver thin films (by 6%) and then graphite powder (by 1%).

Material properties can be determined from the uniaxial stress-stretch curves by fitting strain energy functions, such as those in equations (3.10), (3.11), (3.13), (3.14) and (3.15), to the stress-stretch curves, by using the relationship indicated in equation (3.8). In these purely mechanical-stretching experiments, σ_1 is the stress in the length direction while σ_3 , which is the stress in the thickness direction, would be equal to zero. For example, by making use of the one-parameter Ogden model, the relationship between stress and uniaxial or radial stretch can be obtained

$$\sigma_{\text{uni}} = \mu \left(\lambda^\alpha - \lambda^{-\frac{1}{2}\alpha} \right) \quad (6.18)$$

$$\sigma_{\text{rad}} = \mu (\lambda^\alpha - \lambda^{-2\alpha})$$

wherein μ and α can be determined by making use of MATLAB's curve fitting tool (cftool).

In stiffness measurements, while the stress-stretch behaviours of the composites can be obtained, one cannot directly measure the apparent modulus of the electrodes. Moreover, in the case of crumpled metal thin film electrodes, it is difficult to determine the stiffness of just the metal thin films without considering the elastomer in its neighbourhood. This is because they have an inter-dependent relationship, wherein the buckled metal thin film induces tensile stresses in the elastomer while the stretched elastomer influences the buckling profile of the metal thin film. In addition, the other electrodes are generally discontinuous mediums that cannot be modelled easily: graphite powders are particles while metal thin films are micro-cracked, see Figure 4.7, Figure 5.3 and Figure 7.2. Despite being discontinuous, these electrodes still cause a clamping effect on the elastomer in its vicinity, thereby stiffening it. This is evident in the increased stiffness of the graphite and micro-cracked silver thin film on elastomer composites, as was discussed in section 4.5. The electrodes are assumed to only

have an effect on up to 1 μm of the elastomer beneath it. As such, in order to simplify the characterisation of electrode stiffness for modelling purposes, the electrodes' buckled or discontinuous nature are ignored. Instead, an effective electrode that is hyperelastic and of thickness 1 μm is used for modelling purposes, as shown in Figure 6.7. The equivalent modulus of the effective electrode is extracted from the stretch-stretch data obtained from the tensile tests.

Figure 6.7. Schematic of electrodes modelled as hyperelastic materials with an equivalent modulus. The silver thin film thickness t_{Ag} is 150 nm while the graphite powder has a particle size of 6.5 μm . The proportion of the electrode's thickness to the elastomer's thickness is approximately to scale.

As mentioned, the stress-stretch curves show the behaviour of the composites comprising both the electrodes and the elastomer. In order to determine the equivalent modulus of the electrodes, one can utilise the rule of mixtures,

$$\sigma^{\text{comp}} = \left(\frac{Z}{Z + H} \right) \sigma^{\text{VHB}} + \left(\frac{H}{Z + H} \right) \sigma^{\text{electrode}} \quad (6.19)$$

wherein σ^{comp} , σ^{VHB} and $\sigma^{\text{electrode}}$ are the stresses acting on the composite, VHB elastomer and effective electrode, respectively, while Z is the undeformed thickness of the elastomer and H the combined undeformed thickness of the pair of electrodes.

The Gent model, which we used in section 3.4, is often employed due to its mathematical simplicity and its ability to model the limiting chain extensibility of the elastomer, as characterised by its stiffening at high strains. However, in the range of the actuated strains experienced by the DEAs used here, strains are not high enough for such behaviour to be observed. As such, it has been found that the Ogden model, see equation (3.11), results in a better fit to the stress-stretch behaviour of the electrode-elastomer composites than the Gent model. Good fits for the plain VHB elastomer and the electrode-elastomer composites could be attained by using just a one-parameter Ogden fit, with material parameters obtained from uniaxial tensile tests, shown in Table 4.

The material parameters of each effective electrode could be determined by making use of equation (6.19) and the stress-stretch behaviours of its composite and of the plain VHB elastomer. The composite's tensile stress σ^{comp} can be related to the uniaxial stretch λ ,

$$\sigma^{\text{comp}} = \left(\frac{Z}{Z+H} \right) [\mu^{\text{VHB}} (\lambda^{\alpha^{\text{VHB}}} - \lambda^{-0.5\alpha^{\text{VHB}}})] + \left(\frac{H}{Z+H} \right) [\mu^{\text{electrode}} (\lambda^{\alpha^{\text{electrode}}} - \lambda^{-0.5\alpha^{\text{electrode}}})] \quad (6.20)$$

wherein μ and α are the one-parameter Ogden model material parameters and the superscripts indicate the material.

Table 4. One-parameter Ogden material parameters from uniaxial stress-stretch experiments

Composite	Shear modulus (kPa)	α_1	μ_1 (kPa)	Effective electrode	Shear modulus (kPa)	α_1	μ_1 (kPa)
Plain VHB	47.9	0.6109	156.8				
Graphite powder on VHB	48.5	1.2840	75.6	Graphite powder	536.9	5.329	201.5
Crumpled ELD silver	50.8	0.4454	228.1	Crumpled ELD	579.2	0.004388	264e3

on VHB				silver			
ELD				ELD			
silver	57.3	0.009138	12540	silver	1936	0.001148	3374e3
on VHB							
Sputtered				ELD			
silver	82.7	0.03007	5388	silver	8030	0.003212	5000e3
on VHB							

6.5 DEA model assuming a fixed-frame prestress

The membrane DEAs used in experiments typically maintain a prestretch in the active region by fixing its bordering passive region to a rigid frame. This section models the electromechanical behaviour of such DEAs, which are subject to decreasing magnitudes of prestress with increasing actuated strain, as discussed in section 3.4. In accordance with the experiments conducted, see section 4.1, the membrane DEAs are modelled to have a prestretch of 2.5, an electrode diameter of 10mm and a rigid-frame diameter of 50mm. Apart from the assumption of a varying-force prestress, all material properties and assumptions are the same as in section 3.4. At each prescribed stretch in the elastomer λ_A , the stretch in the passive region, at $R=R_A$, λ_R can be determined by solving differential equations (3.30) and (3.31).

For the DEAs with graphite and flat metal electrodes, by substituting the one parameter Ogden model into equation (3.37), the voltage V required to attain each prescribed stretch in the elastomer λ_A is given by

$$\begin{aligned}
 V = \frac{\sqrt{Z}}{\lambda_A^2 \sqrt{\epsilon_0 \epsilon_r}} & \left\{ Z \left[\mu^{VHB} (\lambda_A^{\alpha^{VHB}} - \lambda_A^{-2\alpha^{VHB}}) \right] \right. \\
 & + H \lambda_{pre}^2 \left[\mu^{electrode} \left(\left(\frac{\lambda_A}{\lambda_{pre}} \right)^{\alpha^{electrode}} - \left(\frac{\lambda_A}{\lambda_{pre}} \right)^{-2\alpha^{electrode}} \right) \right] \\
 & \left. - Z \left[\mu^{VHB} (\lambda_R^{\alpha^{VHB}} - \lambda_R^{-2\alpha^{VHB}}) \right] \right\}^{\frac{1}{2}} \quad (6.21)
 \end{aligned}$$

As for the DEAs with crumpled ELD silver electrodes, which are not stress-free in the prestretched state, the voltage V required to attain a prescribed stretch in the elastomer λ_A is given by

$$\begin{aligned}
 V = \frac{\sqrt{Z}}{\lambda_A^2 \sqrt{\epsilon_0 \epsilon_r}} & \left\{ Z \left[\mu^{VHB} (\lambda_A^{\alpha^{VHB}} - \lambda_A^{-2\alpha^{VHB}}) \right] \right. \\
 & + H \left[\mu^{electrode} (\lambda_A^{\alpha^{electrode}} - \lambda_A^{-2\alpha^{electrode}}) \right] \\
 & \left. - Z \left[\mu^{VHB} (\lambda_R^{\alpha^{VHB}} - \lambda_R^{-2\alpha^{VHB}}) \right] \right\}^{\frac{1}{2}}
 \end{aligned} \tag{ 6.22 }$$

In the prestretched configuration, the active region comprises both a stretched elastomer and a pair of stretched electrodes, while the passive region comprises just the bare stretched elastomer. As a result, the prestretch in the passive region is higher than the in the active region. This is effect is equivalent to the larger constant-force prestress in the crumpled ELD silver DEAs that will be discussed in section 7.3.2. The simulated actuated strains, using the material parameters in Table 4, are shown in Figure 6.8.

Figure 6.8. Simulated actuated strain versus (top) driving voltage and (bottom) electric field behaviour of DEAs prestretched by 2.5 times, with the assumption of a varying-force prestress and using a one-parameter Ogden model, from uniaxial stress-stretch data.

It appears that DEAs using crumpled ELD silver thin film electrodes and graphite powder electrodes would require the least amount of driving voltage in order to obtain a large actuated strain. This is followed by DEAs using ELD silver thin film electrodes and then DEAs using sputtered silver thin film electrodes. This shows that the buckling of the ELD silver thin film does in fact lower the electrode's stiffening effect on the DEA composite, thereby potentially improving actuated strains of the DEA.

6.6 Discussion on the stiffening effect of buckled thin films

From the calculations made using simple structural mechanics, it appears that the buckling of metal thin films will help to reduce its stiffening effect on the comparatively soft elastomer. In the above analyses, the apparent planar stiffness of the buckled thin films was approximated from the buckling load. The assumption made in the trends shown in Figure 6.3 was that due to the application of a critical load, the thin films buckled to form profiles of the same wavelength l , regardless of the extent of compression. Consequently, the amplitudes increased with increasing extents of compression, due to geometric packing, leading to a lowered stiffness at higher compressions. In reality, it is likely that the application of larger compressive strains result in post-buckling deformations because critical strains are often very small, as predicted by equation (6.2). As the axial stiffness of a film is lower when compressed in the post-buckling stage [141], the analyses conducted previously can be considered as conservative as they result in a higher planar stiffness. Simulations based on experimentally-obtained material parameters show that buckled metal thin films does indeed lower the electrode's stiffening effect on the DEA composite, resulting in higher actuated strains than DEAs using flat thin film electrodes.

As discussed in section 6.1, the simple sinusoidal profile of the buckled thin film can rearrange into more complex morphologies at larger compressive strains. In the case of a change to a higher mode, the effective stiffness will increase as a result of a decrease in wavelength, as suggested by equation (6.14). In the event of the formation of nested wrinkles, as was described in section 6.1, the effective thickness of the film will increase, thereby increasing the apparent stiffness of the electrodes. As such, although increasing the compressive strain in thin films generally result in buckled films of a lower apparent stiffness, a compressive strain that is too large can also counteractively result in a higher apparent stiffness, due to a change in the morphology of the buckled thin film.

7 Crumpled ELD silver electrodes in DEAs

From the discussions in section 6, it appears that the buckling of metal thin films will help to reduce its stiffening effect on the soft elastomer. In this section, we aim to experimentally show that ELD silver thin film electrodes are indeed more stretchable and compliant when buckled. When used in a DEA, these biaxially buckled or crumpled ELD silver thin film electrodes are able to achieve large actuated strains, similar to that achieved by DEAs using graphite powder electrodes.

7.1 Fabrication of crumpled metal thin films

For our purpose of developing stretchable and compliant electrodes for DEAs, the conductive metal thin films can be induced to buckle by contracting the elastomer beneath it, and allowing the wrinkles to flatten out when the elastomer expands due to electrostatic forces. To fabricate buckled metal thin film electrodes, the metal film is first deposited on a highly stretched dielectric elastomer. Once the metal thin film is well adhered to the dielectric layer, the dielectric layer is allowed to contract, thereby introducing compressive axial forces in the metal thin film. To do so, one can heat the substrate to thermally expand it before depositing the metal thin film; wrinkles will form in the metal thin film when the composite cools to room temperature [72, 181, 182, 22]. However, only small strains of a few percent can be realised with thermal expansion. In order for the metal thin film to remain highly conductive even when subjected to large planar strains, its wrinkles should not flatten out prior to reaching the desired strain. To do so, more metal film has to be compressed into the same area, which requires a great strain in the dielectric elastomer prior to the deposition of the metal film. In addition to the requirement of large actuated strains, a large prestretch is commonly imposed to the dielectric elastomer prior to the application of the electrodes. As thermal methods are incapable of such large expansions, mechanical means have to be used to expand the dielectric layer prior to the deposition of the metal thin film.

For the biaxially crumpled metal thin films used in this study, compressive biaxial forces were induced in the metal thin films via mechanical means, in order to bring about buckling so severe that that metal film appears to be crumpled. The preparation of crumpled metal thin films is schematically shown in Figure 7.1. First a stress-free elastomer of diameter D_0 is highly stretched to a diameter of D_1 . Following that, a pair of metal thin films is deposited on

either side of the highly-stretched elastomer. In this state, the elastomer is highly-stretched while the metal thin films are relatively stress-free and flat. By reducing the stretch in the elastomer, from a diameter of D_I to D_{II} , induced biaxial compressive strains cause the metal films on the elastomer to buckle, thereby forming crumpled metal thin film electrodes.

Figure 7.1. Schematic illustrating the fabrication and activation of a membrane DEAs with crumpled metal thin-film electrodes.

In a pre-stretched state, the elastomer substrate is under tension and has a radial prestretch ratio of

$$\lambda_p = \frac{D_{II}}{D_0} \quad (7.1)$$

while the buckled metal thin-films are subjected to an apparent compressive strain of

$$c = \frac{D_I - D_{II}}{D_I} = 1 - \frac{D_{II}}{D_I} \quad (7.2)$$

Upon activation by Maxwell stress, the membrane DEA expands in area to an activated diameter of D_{act} . The electrically-induced area strain is given by:

$$s_A = \left[\left(\frac{D_{act}}{D_{II}} \right)^2 - 1 \right] 100\% \quad (7.3)$$

This area expansion is accompanied by the unfolding and flattening of the crumpled metal thinfilm electrodes.

A radial stretching jig, described in section 4.3.2, was used to control and stretch the elastomer. First, an elastomer was highly-stretched to a stretch ratio higher than λ_p . Next, the stretched elastomer, held by sixteen crocodile clips, was temporarily fixed to a portable circular frame by means of bolts and nuts. Once secured to the frame, the stretched elastomer was unhooked from the jig to make the application of electrodes to either side of the stretched elastomer easier. After the stress-free metal films had been deposited on the stretched elastomer, it was hooked back to the jig and unfastened from the portable frame. Subsequently, as the stretch in the elastomer was reduced to λ_p , the induced compressive strain c , as defined in equation (7.1), in the metal thin films caused them to crumple. In order to vary the amount of compressive strain in the crumpled electrodes, while maintaining the same prestretch, the stretch at which the electrodes were deposited was altered. For example, to attain a DEA with $\lambda_p = 2.5$ and $c = 0.4$, metal thin films had to be deposited on an elastomer that had been highly-stretched by 4.2 times. The stretching and relaxation of the 250 μ m-thick dielectric elastomer (VHB F9473PC, 3M) was carried out at a rate of 0.06 D_0 per second. The ELD silver thin films were deposited in the same way as described in section 5.1.

7.2 Morphology of crumpled ELD silver thin films

The morphology of the buckled metal thin film depends on the amount of compressive strain that it had been subjected to. As shown in Figure 7.2, the buckling of ELD silver thin films becomes more severe with increasing amounts of compressive strains.

Figure 7.2. Change in morphology as the ELD silver thin film was subjected to varying amounts of compressive strain. A lower magnification is shown on the left column while a higher magnification is shown on the right column.

The morphologies are generally dissimilar to the sinusoidal profile described in the classical studies on wrinkling [22, 4]. This is because the compressive strains used here are biaxial and are far larger than the critical strain of just 0.006%, according to equation (6.2). In addition, the VHB elastomer is known to exhibit viscoelastic properties [67, 116], which would affect the rate at which the thin films buckle, which in turn affects the resultant morphological and dynamical features observed for large deformations [84].

Initially, the stress-free ELD silver thin film was rather flat, conforming to the surface morphology of the prestretched VHB membrane. When subjected to 20% compressive strain, deep folds and fine cracks formed. Despite the compressive nature of the stresses, biaxial compressive strains will induce tensile membrane forces in the buckled film [157], resulting in the formation of cracks that help relieve the tension. In fact, this morphology is reminiscent of the theory of plate tectonics, wherein the movement of the earth's crust, resting on a bed of molten lava, folds to form mountain ridges and cracks to form fault lines [47, 166]. As the amount of compressive strain increased further, the ELD silver thin film buckled further, resulting in deeper folds of shorter wavelengths. At 50% compressive strain, fine twisted folds formed on the existing wrinkles, giving a nested hierarchy [52]. Under large biaxial compressions, ELD silver thin films transform into a complex combination of deep folds and tightly packed wrinkles. As such, we have coined these electrodes as crumpled metal thin film electrodes.

7.3 Mechanical properties of crumpled ELD silver thin films

The mechanical properties of the electrodes would affect the electromechanical performance of its DEAs. If the electrodes are not stretchable enough, then charges cannot be transported to the dielectric elastomer's surfaces, which would hamper large deformations. Also, if the electrodes are very stiff, then it would clamp the dielectric elastomer, thereby restricting its deformations. Here, the stretchability and stiffening effect of the electrodes are explored.

7.3.1 Stretchability of crumpled ELD silver thin film electrodes

Generally, electrode resistance increases with increasing tensile strain and different types of electrodes exhibit different initial resistances and stretchability. Here, the uniaxial stretchability of crumpled ELD silver thin film electrodes is compared against the stretchability of graphite powder, sputtered silver thin film and ELD silver thin film

electrodes. The crumpled ELD silver thin films were prepared by allowing the elastomer to relax totally to a 1.05 residual stretch in the elastomer, such that the ELD silver thin films had a compressive strain of 75%, as defined by equation (7.2). Each sample, initially a 25mm square, was uniaxially stretched until it ceased to be conductive ($>1\text{G}\Omega$). Figure 7.3 shows the electrode resistance for the various electrode materials as a function of uniaxial strain. It can be observed that metal film electrodes have a much lower initial sheet resistance than graphite powder electrodes: graphite had a sheet resistance of $> 10\text{ k}\Omega/\square$, while flat sputtered silver thin film and ELD silver thin film had a sheet resistance of $1\text{ }\Omega/\square$ and $10\text{ }\Omega/\square$ respectively. The crumpled ELD silver thin film showed a slightly higher resistance ($21\text{ }\Omega/\square$) than the flat ELD silver thin film because of the cracks that formed in the silver films during the buckling process, such as those observed in the silver film that had been compressed by 20% (Figure 7.2). However, crumpled silver thin film electrodes had the highest stretchability among the electrode materials considered. It managed to retain its electrical conductivity up to 404% uniaxial strain. In contrast, graphite powder electrodes lost their conductivity at 140% while flat sputtered silver thin films and flat ELD silver thin films lost their conductivity at 21% and 33%, respectively.

Figure 7.3. Change in resistance across each electrode as they were subjected to increasing amounts of uni-axial mechanical strain. The electrodes were deemed to be non-conductive at a resistance of $1\text{G}\Omega$.

When the samples, initially 40mm-diameter circles, were radially stretched, a similar trend of increasing resistance with increasing stretch was observed, as shown in Figure 7.4. However, as compared to uniaxial stretching, the radially stretched electrodes lose their conductivities at lower strains. The calculated sheet resistance of a radially-stretched circular electrode was found to be just slightly lower than measured resistance, as discussed in appendix A6. Therefore, the measured resistance of the circular electrode can be taken to be approximately the same as its sheet resistance. ELD silver thin films, with the lowest initial resistance of 10 Ω , could only be stretched up to 11% radial strain, beyond which it cracked into fine isolated grains and lost its electrical continuity [110]. Graphite powder electrodes, which had a higher initial resistance of 51k Ω , could be stretched by up to 50% radial strain, beyond which the network of conductive particles disconnected. The crumpled ELD silver thin films used here had a compressive strain of 40%, as defined by equation (7.2), and could be stretched by up to 110% radial strain. This 11-fold improvement over the plain ELD silver thin film can be attributed to the unfolding capability of the crumpled form, which helps delay the widespread crack formation in the metal thin-film under high tension. Like in the uniaxial tests, the 40% crumpled ELD silver thin film electrode had a higher initial resistance of 90 Ω , as compared to that of the plain flat ELD silver thin film electrode.

Figure 7.4. Radial stretchability of different types of electrodes.

Related to its ability to maintain a low resistance even at high stretches, crumpled ELD silver thin film electrodes are able to retain a full coverage of the radially-stretched elastomer throughout the deformation process. Theoretically, the capacitance C should be directly proportional to the forth power of radial stretch λ ,

$$C = \frac{\epsilon_o \epsilon_r A}{z} = \frac{\epsilon_o \epsilon_r A_o \lambda^2}{Z \left(\frac{1}{\lambda^2} \right)} = C_o \lambda^4$$

where ϵ_o is the permittivity of free space, ϵ_r the relative permittivity, A and A_o the instantaneous and original areas, z and Z the instantaneous and original thickness of the elastomer and C_o the original capacitance of the DEA. As shown in Figure 7.5, corresponding to theory, the measured capacitance of the DEA using crumpled ELD silver thin film electrodes varies linearly with respect to the forth power of stretch. This shows that the crumpled ELD silver thin film electrodes are able to maintain a full coverage of the elastomer throughout the stretching process.

Figure 7.5. Change in capacitance with respect to (left) stretch and (right) forth power of stretch.

The crumpled ELD silver thin film electrodes are also able to sustain a cyclic uniaxial loading of between 0% and 100% strain. The crumpled ELD silver thin film electrodes were 10mm by 10mm in size and had a compressive strain of 75% at zero apparent strain. The electrodes were cyclically strained, between 0% and 100% uniaxial strain, for one hundred

times at a rate of 50% strain per minute. As shown in Figure 7.6, the resistance of the crumpled silver thin film electrodes remained low, below 60 Ω , throughout the one hundred cycles. After the first cycle, the resistance of the unstrained electrode, at 0% strain, increased from 33 Ω to 45 Ω . Thereafter, the unstrained electrode resistance decreased through the first 30 cycles and increased slightly through cycles 30 to 40. After the first 40 cycles, the electrode resistance stabilized; the unstrained electrode resistance was maintained at $42 \pm 0.2 \Omega$ while the 100%-strained electrode resistance was kept at $52 \pm 0.2 \Omega$.

Figure 7.6 Change in resistance across the electrode as the crumpled ELD silver electrode was cyclically strained between 0% and 100% for a hundred times.

7.3.2 Stiffening effect of crumpled ELD silver thin films

Uniaxial stretching, as was discussed in section 6.4.1, is the simplest and most common method of determining the stiffness of materials. However it might not be able to accurately depict the actual working conditions of the biaxially deformable membrane DEAs described in section 4.1. In order to more accurately measure the stiffening effect of the different types of electrodes on a 250 μm -thick VHB F9473PC elastomer, elastomer membranes, coated on both sides with electrodes, were radially stressed, starting from an initial equi-biaxial prestretch of 2.5.

Radial stress σ_R increased with increasing stretch ratio (D / D_0) and the type of electrode material was found to influence the apparent radial stiffness of the composite to different extents, as shown in Figure 7.7. Here, the gradient of the electrode-elastomer membrane's stress-stretch ratio is used as a measure of the radial modulus E_R of the electrode-elastomer

membranes. Each electrode's stiffening effect is apparent when the radial modulus of its electrode-elastomer composite was compared to the radial modulus of the plain VHB elastomer. As a reference, the plain prestretched VHB elastomer was found to have a radial modulus of 402kPa. Flat silver thin films was found to stiffen the elastomer membrane the most, wherein sputtered silver elastomer composites were three times stiffer and ELD silver elastomer composites were 2.6 times stiffer than plain elastomers. Graphite powder coatings were found to minimally stiffen the elastomer membrane by just 8%. As for the 40% crumpled ELD silver thin films, little stiffening effect was also observed, wherein the crumpled ELD silver-elastomer composite was just 13% stiffer than the plain VHB elastomer. Unlike other electrodes, which are stress-free when the elastomer is in its prestretched state, the crumpled metal electrodes add a residual stress to the electrode-elastomer composite, thereby requiring a higher radial prestress at the same apparent prestretch of 2.5.

Figure 7.7. Stiffening effect of different types of electrodes on the electrode-elastomer membrane composites that were subject to radial stretching.

Figure 7.8 shows the stress-strain curve of a plain VHB sample, a graphite and a crumpled ELD silver on VHB sample that were radially stretched from their stress-free states. There was an initial stretch of 1.05 in the elastomer of the crumpled ELD silver composite, caused by the compressive forces in the silver. It is apparent from these stress-strain curves that the

crumpled ELD silver on VHB composite is stiffer than the bare VHB elastomer. Unlike the samples that were left to relax after being biaxially stretched by 2.5 times, these continually loaded samples have a smaller radial modulus between a stretch of 2.5 and 4.

Figure 7.8. Amount of mechanical stress required to radially stretch a plain VHB disc and VHB composites with graphite and crumpled ELD silver thin film electrodes. Due to the compressive stresses in the crumpled silver film, tensile stresses were induced in the VHB elastomer, which in turn caused a 1.11 prestretch in the crumpled ELD silver composite in its stress-free state.

7.4 Electromechanical performance of crumpled ELD silver thin film DEAs

Crumpled metal thin film electrodes can be used as compliant electrodes for DEAs that require biaxial deformations as they have been found to be highly stretchable and only slightly stiffens its elastomer substrate. Here, membrane DEAs using 40% crumpled ELD silver thin-film electrodes were prepared as described before: a 4.2 times stretched elastomer membrane, on which initially stress-free ELD silver thin-films were deposited, was relaxed to a 2.5 times pre-stretch. However, due to the stiffness of the electrode trails, the initially circular stress-free silver electrodes crumpled into an oval shape of approximately 10mm in diameter. This pre-stretched membrane DEA was subsequently transferred to a 50 mm square acrylic frame for support.

Activation of the DEAs, using a high voltage power supply, caused the electrodes to expand with a large area strain. As shown in Figure 7.9, the actuated area strain was 103% at 1.6 kV ($80\text{V}\mu\text{m}^{-1}$) and further increased to 128% at 1.8 kV ($102\text{V}\mu\text{m}^{-1}$). During area expansion, the crumpled electrodes were observed to unfold from the fine wrinkles into the coarse ones, as shown in Figure 7.10. Such large actuated strains, of DEAs using crumpled ELD silver thin film electrodes, are comparable to the actuation strains produced by DEAs using graphite powder electrodes, as shown in Figure 7.11, and is a marked improvement over the actuation capability of DEAs using plain silver thin film electrodes. This can be attributed to the combination of improved stretchability and reduction of the silver thin-films' apparent Young's modulus due to buckling. The actuated strains of the DEA using ELD silver thin film electrodes can be seen to deviate from that of DEAs using crumpled ELD silver thin film electrodes from about 10% strain. This is largely due to the lower stretchability of the ELD silver thin film electrodes as the micro-cracked ELD silver thin films actually get less stiff with increasing stretch, as was discussed in section 5.3. As shown in Figure 7.4, the resistance of micro-cracked ELD silver thin films increase rapidly after being stretched by about 7%, while the resistance of crumpled ELD silver thin films remain relatively unchanged when stretched by up to 40%. This indicates that at higher strains, there would be higher electrical losses in DEAs using flat ELD silver thin films than in DEAs using crumpled ELD silver thin films.

Figure 7.9. Photographs showing the top view of a 2.5 times biaxially prestretched membrane DEA with crumpled ELD silver electrodes that had an initial compressive strain of 40%.

Figure 7.10. The micrographs (taken from a different sample as in Figure 7.9) illustrate the unravelling of the folds during activation at 2kV. The corresponding actuated area strain is approximately 90%.

Figure 7.11. Actuated area strain as a function of driving voltage for DEAs using different electrode materials. Ten samples for the crumpled ELD silver DEAs were tested and error bars indicate the standard deviation.

The simulated actuated strains, discussed in section 6.5, appear to fit experimental data quite well, but only up to an electric field of about $40\text{-}50\text{V}\mu\text{m}^{-1}$, as shown in Figure 7.12. At higher electric fields, the simulations veer towards higher actuated strains and then mechanical instability, while the experimental data tapers off. For the DEAs using metal thin film

electrodes, the model deviates from the experimental results more drastically, as compared to DEAs using crumpled ELD silver and graphite powder electrodes. The model used here is based purely on the mechanical planar stiffness of the VHB elastomer and its electrodes. However, in practice, the actuated strain of the DEA depends on both mechanical and electrical properties of materials, as well as the coupling of mechanical and electrostatic stresses. Two possible reasons have been discussed in appendix A8.

Figure 7.12. Simulated actuated strain versus (top) driving voltage and (bottom) electric field behaviour of DEAs prestretched by 2.5 times, with the assumption of a varying-force prestress and using a one-parameter Ogden model, from uniaxial stress-stretch data.

So far, it has been shown that crumpling ELD silver thin films by 40% results in a lower apparent Young's modulus; it would be interesting to find out if the extent of crumpling has an effect on its apparent Young's modulus, such that it manifests in the actuated strains of DEAs. As described in section 7.2, on the changes in morphology of the crumpled ELD silver thin films with greater extents of electrode compression, the closer and deeper the wrinkles and folds. This in turn affects its apparent Young's modulus, as suggested by equation (6.14), thereby influencing the achievable electromechanical actuation of the DEA. As shown in Figure 7.13, the maximum actuated strain achievable by DEAs varied with the extent of electrode crumpling, even though they all had the same elastomer prestretch. That is to say, for a 40% crumpling, ELD silver thin films were deposited on a 4.2 times stretched

elastomer, before reducing the stretch to a prestretch to 2.5; as for a 10% crumpling, ELD silver thin films were deposited on a 2.8 times stretched elastomer, before reducing the stretch to a prestretch of 2.5. DEAs with plain ‘flat’ ELD silver thin film electrodes produced the least actuation due to the large stiffening effect of its electrodes. As shown in Figure 7.14, DEAs with crumpled metal thin film electrodes generally fared better with more electrode crumpling, up to 40% compressive strain. However, too much crumpling made the electrodes stiffer. This was caused by a reduction in the wavelength, refer to equation (6.14), and an increase in the effective thickness of the electrodes, due to a nested hierarchy of wrinkles and folds, as apparent in Figure 7.2. Consequently, DEAs using ELD silver thin film electrodes that had been crumpled by 50% had smaller actuated strains than DEAs using ELD silver electrodes that had been crumpled by 30% and 40%.

Figure 7.13. Actuated area strain, with respect to the applied electric field, of DEAs using ELD silver thin film electrodes that had been crumpled by 0%, 20%, 30%, 40% and 50% compressive strains.

Figure 7.14. Actuated area strain (at an electric field of $80 \text{ V}\mu\text{m}^{-1}$) obtained by membrane DEAs with ELD silver electrodes that were crumpled to varying degrees.

7.5 Self-clearing properties

Silver thin film electrodes have been reported to be able to self-clear [110, 20], which can help to stop localized electrical breakdown from prematurely damaging DEAs. In this case, the crumpled silver thin film electrodes were also found to be able to self-clear on DEAs. Typically, a DEA has an electrical breakdown localized at a defective spot. For example, the sample shown in Figure 7.15 was subjected to a localized breakdown, wherein purplish sparks were observed at the defective spot near the electrode edge. During the breakdown, the leakage current surged and the driving voltage fell as shown in Figure 7.16. The electrical shorting caused the area strain of the DEA to decrease to 31%, lesser than the achievable strain at the pre-set voltage of 2kV. The elastomer membrane was punctured, as shown in Figure 7.17a, due to the high current flow and joule heating at the shorting spot. The electrical breakdown oxidized the silver thin film surrounding the defective spot and eventually isolated the spot, thereby stopping the breakdown. As the localized defect has been self-cleared, the current leakage was stopped and the pre-set driving voltage restored. Consequently, this self-cleared DEA sample actuated area strain increased to 84% at 2kV, as shown in Figure 7.15.

Self-clearing can occur multiple times in a single DEA prior to total failure. As shown in Figure 7.16, upon increasing the driving voltage by another 100V, a second localised breakdown occurs, which then self-clears again to enable an elevated driving voltage of 2.1kV. This sequence takes place once more before the DEA fails at an applied voltage of 2.2kV. Although self-clearing electrodes allow higher driving voltages, they should only be used for preventing localised breakdowns at a few spots. This is because self-clearing results in the passivation of the surrounding electrode material, which makes those regions non-conducting, thereby resulting in a compromise of actuated strains. Ill-positioned self-cleared sites in the middle of a DEA, as shown in Figure 7.17b/c, result in the formation of large wrinkles, which also reduces in-plane actuated strains. As such, while self-clearing electrodes cannot compensate badly manufactured DEAs that are full of flaws, they are very useful for preventing the premature breakdown of DEAs that have just a few localised defects.

Figure 7.15. Photographs of the top view of a DEA with crumpled ELD electrodes that undergo self-clearing. Before the event, at 1.9 kV, the area strain was 63%. After the driving voltage was increased to 2 kV, an electrical breakdown occurred, causing the area strain to decrease to 31%. However, after the self-clearing event, at the same driving voltage of 2 kV, the area strain increased to 84%.

Figure 7.16. Time histories of voltage and current show an electrical breakdown that is followed by self-clearing, as indicated by a spike in current and a corresponding plunge and recovery in voltage. Four self-clearing event are shown, of which the voltage change during the 1st and 2nd events are magnified.

Figure 7.17. a) a self-cleared site in a DEA membrane sample with crumpled ELD silver thin film electrodes. b) arc discharge at self-clear site in the middle of the actuator. c) wrinkling of the actuator due to passivated self-clear sites. (the white borders around the electrodes in (b) and (c) is ink used for imaging purposes)

7.6 Discussion on crumpled ELD silver electrodes

There are several benefits of using buckled or crumpled metal thin films over ‘flat’ metal thin films as DEA electrodes. First and foremost, the buckling of a metal thin film greatly improves its stretchability: crumpled ELD silver thin films that have been compressed by

75% can be uniaxially strained by more than 400% while flat ELD silver thin films can be uniaxially strained to just over 30%. Although uniaxial stretchability of the electrodes is often used as a characterisation metric [101, 29, 171], many DEAs also need to undergo biaxial actuations. As such, a DEA's electrodes should also ideally be biaxially stretchable. By crumpling ELD silver thin films by 40%, the electrodes can be radially strained by up to 110% while flat ELD silver thin films can only be radially strained by up to about 10%. Buckling of metal thin films also lowers the apparent stiffness of the thin film, which in turn has a smaller stiffening effect on the electrode-elastomer composite, as compared to flat metal thin films. In the case of crumpled ELD silver thin films, a biaxial or radial buckling was used, in contrast to uniaxial corrugations, so as to lower the apparent stiffness of the electrodes in all in-plane directions. Due to this combination of improved stretchability and a lowered stiffness, in all in-plane directions, the actuated strains attained by DEAs using crumpled ELD silver thin film electrodes was greater, at more than 120% area strain, than the 50% area strain attainable by DEAs using flat ELD silver thin film electrodes. In addition, the higher actuated strain was attained at a lower driving voltage (1.8kV versus 9kV), taking into account that the elastomers in both DEAs had a similar initial thickness. Improvements to a DEA's actuated strains, by using metal thin films in a crumpled form over a flat form, is not exclusive to ELD silver. DEAs using gold leaf electrodes have also been found to have greater actuated area strains when its electrodes were crumpled (73%) as compared to when its electrodes were flat (6%), as discussed in appendix A7. Recently, it has also been reported that DEAs using crumpled graphene electrodes were able to attain more than 100% actuated area strain [184].

Apart from improvements to the metal thin film's stretchability and mechanical compliance, the buckling of a metal thin film does not adversely affect its existing favourable traits. Crumpled metal thin film electrodes are still able to self-clear, which helps to prevent premature failures of the DEA. In addition, although the sheet resistance of the crumpled ELD silver thin film electrodes is slightly higher than that of their flat counterparts, it remains low, especially when compared to the $>10\text{k}\Omega/\square$ sheet resistance of graphite powder. The crumpled ELD silver thin film electrodes managed to retain a sheet resistance of under $60\Omega/\square$ when uniaxially strained by 100% for one hundred cycles. It appears that crumpled ELD silver thin film electrodes have the potential for use in larger-scale dielectric elastomer actuators, due to its high stretchability and relatively low sheet resistance, which can help to reduce parasitic resistive losses [27], as compared to carbon-based electrodes. In addition,

taking into consideration that the capacitance of the DEA increases with stretch, maintaining a low resistance at high stretches can help to improve the time required to charge the DEA, i.e. the RC time constant τ , which is directly proportional to both the resistance of the electrodes and the capacitance of the DEA. Therefore, using electrodes with a low resistance can also help to potentially improve the bandwidth of DEAs with fast mechanical responses.

While the crumpling of metal thin films can benefit its DEA's actuated strains, it is not without its disadvantages. The most obvious drawback is the additional fabrication steps and equipment required to induce the metal thin films to crumple. For example, if one is attempting this method using metal thin films deposited by evaporation or sputtering, the size of the deposition chamber would limit the size of the highly stretched elastomer and its supporting structure. However, the metal thin films used in this chapter, i.e. ELD silver thin films and gold leaves, could be deposited simply and under ambient conditions. Apart from having a high conductivity, silver was originally chosen due to its low reactivity. However, silver is generally susceptible to attack by sulphates and chlorides. When kept in clean nitrogen, flat ELD silver thin film samples did not deteriorate after more than three years. However, when unprotected, the flat ELD silver thin film samples became unusable after a month. On the other hand, the crumpled ELD silver samples could be kept in its crumpled state for much longer: the square resistance of unused crumpled ELD silver thin film samples remained below $200\Omega/\square$ after one year. In view of this, a protective layer will have to be introduced if the DEA is to be used in harsh environments and its effects on the electromechanical behaviour studied.

8 Conclusions

The work in this thesis has shown that it is possible to make metal thin film electrodes to be almost as biaxially stretchable and biaxially compliant as conventional carbon-based electrodes, with the added benefits of being more conductive and having the ability to self-clear. Metal thin films, primarily in the form of silver that had been deposited electrolessly, have been made to be biaxially stretchable and compliant via two mechanisms: an inherently brittle thin film that results in a network of zigzagging micro-cracks and a biaxially compressed thin film that buckles to form crumples.

Electroless deposition is a relatively simple and inexpensive method that can be used to fabricate metal thin films under room temperature and pressure conditions. This is especially so when compared to other more commonly-used metal thin film deposition methods, such as sputtering and evaporation, which require costly equipment that utilises a vacuum chamber. Electroless deposition is commonly used to silver mirrors of large areas and has also been used to create micro-scale patterns, thereby giving ELD silver the potential to be used for both large and small scale DEA applications.

When 150nm-thick ELD silver films were deposited on a soft elastomer substrate, the network of zigzagging micro-cracks that formed enabled them to be moderately stretchable, at up to 33% uniaxial strain and up to 11% radial strain. This is a stark improvement over the 2-3% fracture strains of freestanding metal films. In addition, the micro-cracked ELD silver thin films appear to have a lower stiffening effect on the elastomer-electrode composite as compared to a sputtered silver films of the same thickness. As a result, DEAs using ELD silver thin film electrodes managed to attain actuated area strains of up to 50%. Such actuated strains can be considered as high when compared to DEAs using sputtered gold and silver films that generally have maximum actuated area strains of less than 10%.

The biaxial stretchability and compliance of metal thin films were further enhanced by introducing biaxial compressive axial stresses, in order to make them buckle to form crumpled metal thin film electrodes. The stretchability of crumpled ELD silver thin films improved to 400% uniaxial strain and 110% radial strain. Also, the apparent stiffness of the thin film was reduced, which results in a lowered stiffening effect on the electrode-elastomer composite, comparable to that of graphite powder. Due to this combination of improved

stretchability and a lowered stiffening effect, in all in-plane directions, high actuated area strains of 128% was attained by DEAs using crumpled ELD silver thin film electrodes. This method of crumpling metal thin films to improve actuated strains is not exclusive to ELD silver thin films: DEAs using gold leaf electrodes have also been found to have greater actuated area strains when its electrodes were crumpled (73%) as compared to when its electrodes were flat (6%).

There are a couple of added benefits that make metal thin film electrodes particularly attractive for us in DEAs. Firstly, metal thin film electrodes are able to self-clear, which makes its DEAs less susceptible to premature failures. This applies to both the micro-cracked and the crumpled versions of ELD silver thin films. Secondly, the sheet resistances of metal thin films are much lower than that of carbon-based electrodes, which tend to be more than $10\text{k}\Omega/\square$. This is especially important when the elastomer starts to become conductive at high fields, as the low sheet resistance of metal thin films can help to reduce the voltage drop to the dielectric elastomer. Crumpled ELD silver thin film electrodes have managed to retain a sheet resistance of under $60\Omega/\square$ when uniaxially strained by 100% for one hundred cycles.

We have found that it is possible to model a DEA's electromechanical response based purely on the stiffness of its electrodes and dielectric elastomer. However, this is on the condition that the dielectric elastomer remains a perfect dielectric. In practice, the elastomer becomes conductive at high electric fields, resulting in a closed circuit. Due to the increase in leakage current, high resistance electrodes would cause a large voltage drop across it, resulting in a lower voltage across the elastomer, thereby decreasing the DEA's actuated strains. This shows the importance of a DEA's electrodes having to be both highly compliant and stretchable.

The metal thin film electrodes presented here are both biaxially stretchable and compliant, to strains comparable to that of conventional carbon-based electrodes. Also, they have the additional benefits of low conductivity and the ability to self-clear. As such, this makes them highly suitable for use as electrodes in high-strain DEA applications.

8.1 Future work

As the focus of this thesis is on the development of compliant metal thin film electrodes, little effort was spent in the development of the dielectric elastomer. Instead, a VHB elastomer was

used, as it is the most commonly used dielectric elastomer in research works regarding DEAs. The highly deformable VHB elastomer has enabled us to demonstrate the high stretchability of crumpled metal thin film electrodes. However, VHB elastomers are not perfect for use in DEAs due to time-dependent effects, such as viscoelasticity, and dissipative effects, such as a varying permittivity with changes in stretch and temperature. Silicone elastomers have been a popular alternative to VHB elastomers in the field of DEAs as it is more elastic and can therefore have a faster response speed. Although we have been able to deposit ELD silver thin film electrodes on silicone elastomers [109], the electrodes were not well adhered. This is because electroless deposition is a wet process and silicones are hydrophobic in nature. In order for ELD silver to be used with silicones, the silicone surface has to be made hydrophilic, by means of methods such as dry etching in oxygen plasma, followed by the its salinization using 3-aminopropyl-triethoxysilanes (APTES) [175]. In order for widespread use of electroless deposition of metals, further study will have to be aimed at either making the silicone surface hydrophilic or to develop a suitable dielectric elastomer that is naturally hydrophilic.

As the metal thin film electrodes studied here are biaxially compliant, they would be most suited for use in actuator configurations that make use of biaxial deformations. For example, biaxial planar expansions can be translated into linear strokes by making use of the “spider” configuration [90]. Apart from its use in DEAs, crumpled ELD silver thin film electrodes appear to be suitable for use in the dielectric elastomer generators (DEGs) as well. This is because DEGs make use of the change in its capacitance with stretch in order to generate electricity. As capacitance is directly proportional to changes in its area, it is essential that its electrodes are able to maintain a full coverage of the elastomer as it deforms. Crumpled ELD silver thin film electrodes have been shown to be able to maintain a full coverage of the elastomer as it deforms, as inferred from its capacitance being proportional to the forth power of stretch. In addition, DEGs used for applications such as wave harvesting are large-scale setups wherein the resistance of the electrodes would affect resistive losses due to Joule heating [26]. Also, low resistances can help to improve the RC time constant of the capacitor. As crumpled ELD silver thin film electrodes have a sheet resistance that is much lower than that of carbon-based electrodes, they appear to be competitive alternatives as electrodes for large-scale DEGs.

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Appendix

A1. MATLAB code for studying the effects of prestretch

This is the supporting data for section 3.3.

```
% Effect of prestretch on biaxial actuation
% assumption of dead weights supplying prestress
% electrodes are assumed to have been applied after prestretch
clear all; close all; clc;

%% Define parameters
permit = 4.5*8.854e-12; % permittivity
Z = 250e-6; % original elastomer thickness (in the reference
state)
H = 2e-6; % original electrode thickness (in the
prestretched state)
lambdaPre = 1:0.5:5; % prestretch
lambdaAct = 1:0.01:2; % actuated stretch
factor = 1; % shear modulus of electrode wrt elastomer

%% Input of material parameters
% Gent VHB (Bozlar's parameters)
VHBmuG = 50e3; VHBJlim = 125;

stressElec = cell(size(lambdaPre));
stressVHB = cell(size(lambdaPre));
shearMod = zeros(size(lambdaPre));
V = cell(size(lambdaPre));
Stretch = cell(size(lambdaPre));
ActAreaStretch = cell(size(lambdaPre));

for ii=1:length(lambdaPre)
    lambdaA = lambdaAct.*lambdaPre(ii); % stretch in active
region
    % determining force per unit length based on prestretch
ratio
    syms lambda1
    prestress = Gent(lambdaPre(ii),VHBmuG,VHBJlim);
    Nx = double(Z/lambdaPre(ii)*prestress); % force per unit
length is determined by the initial prestretch

    stressElec{ii} =
Gent(lambdaA./lambdaPre(ii),factor*VHBmuG,VHBJlim);
    stressVHB{ii} = Gent(lambdaA,VHBmuG,VHBJlim);

    % Calculation of the input voltage
    V{ii} = real(sqrt( ( H.*stressElec{ii}) + ...
```

```

        (Z./lambdaPre(ii).^2.*stressVHB{ii}) - ...
        (Nx.*lambdaA./lambdaPre(ii).^2)) ./ ...
        (permit./Z./lambdaPre(ii).^2.*lambdaA.^4) ));
    Stretch{ii} = lambdaA;
    ActAreaStretch{ii} = (lambdaA./lambdaPre(ii)).^2;
end

%% Breakdown field
Ebreak = [100e6 150e6]; %breakdown field
Lambdabreak = 1:0.1:max(lambdaA);
Vbreak{1} = Ebreak(1).*Z./Lambdabreak.^2;
Vbreak{2} = Ebreak(2).*Z./Lambdabreak.^2;

%% Plotting
figure(1)
for ii=1:length(lambdaPre)
    plot(Stretch{ii},V{ii},'LineWidth',2)
    hold all
end
plot(Lambdabreak,Vbreak{1},'c--
',Lambdabreak,Vbreak{2},'g:', 'LineWidth',2)
grid on
axis([1 6 0 9000])
xlabel('stretch in elastomer','FontSize',12)
ylabel('voltage (V)','FontSize',12)
set(gca,'FontSize',12)

%% plotting actuated area strains
figure(2)
plotPre = [1 1.5 2 2.5 3 4];
breakVolt1 = cell(size(plotPre));
breakVolt2 = cell(size(plotPre));
for ii=1:length(plotPre)
    breakVolt1{ii} =
Ebreak(1).*Z./plotPre(ii).^2./ActAreaStretch{ii};
    breakVolt2{ii} =
Ebreak(2).*Z./plotPre(ii).^2./ActAreaStretch{ii};
    for jj=1:length(ActAreaStretch{ii})
        index1(ii) = jj;
        if V{ii}(jj) > breakVolt1{ii}(jj)
            index1(ii) = jj-1;
            break
        end
    end
end
for jj=1:length(ActAreaStretch{ii})
    index2(ii) = jj;
    if V{ii}(jj) > breakVolt2{ii}(jj)
        index2(ii) = jj-1;
        break
    end
end
end
end

```

```

plot(ActAreaStretch{ii}(1:index1(ii)),V{ii}(1:index1(ii)),'-
','LineWidth',2)
    hold all
end
hold on
for ii=1:length(plotPre)

plot(ActAreaStretch{ii}(index1(ii):index2(ii)),V{ii}(index1(ii)
):index2(ii)),':', 'LineWidth',2)
    hold all
end
grid on
% axis([1 4 0 4000])
xlabel('actuated area stretch','FontSize',12)
ylabel('voltage (V)','FontSize',12)
set(gca,'FontSize',12)

```

A2. MATLAB code for studying the effects of electrode stiffness

This is the supporting data for section 3.3.

```

% Effect of electrode stiffness on biaxial actuation
% assumption of dead weights supplying prestress
% electrodes are assumed to have been applied after prestretch
clear all; close all; clc;

%% Define parameters
permit = 4.5*8.854e-12; % permittivity
Z = 250e-6; % original elastomer thickness (in the reference
state)
H = 20e-6; % original electrode thickness (in the
prestretched state)
lambdaPre = 2.5; % prestretch
lambdaAct = 1:0.01:2; % actuated stretch
lambdaA = lambdaAct.*lambdaPre; % stretch in active region

%% Input of material parameters
% Gent VHB (Bozlar's parameters)
VHBmuG = 50e3; VHBJlim = 125;

factor = [0 10 100]; % shear modulus of electrodes wrt
elastomer
stressElec = cell(size(factor));
stressVHB = cell(size(factor));
shearMod = zeros(size(factor));
V = cell(size(factor));

```



```

Stretch = cell(size(factor));
ActAreaStretch = cell(size(factor));

for ii=1:length(factor)

    % determining force per unit length based on prestretch
    ratio
    syms lambda1
    prestress = Gent(lambdaPre,VHBmuG,VHBJlim);
    Nx = double(Z/lambdaPre*prestress); % force per unit
    length is determined by the initial prestretch

    stressElec{ii} =
    Gent(lambdaA./lambdaPre,factor(ii)*VHBmuG,VHBJlim);
    stressVHB{ii} = Gent(lambdaA,VHBmuG,VHBJlim);

    shearMod(ii) = factor(ii)*VHBmuG;
    %% Calculation of the input voltage
    V{ii} = real(sqrt( ( H.*stressElec{ii}) + ...
        (Z./lambdaPre.^2.*stressVHB{ii}) - ...
        (Nx.*lambdaA./lambdaPre.^2)) ./ ...
        (permit./Z./lambdaPre.^2.*lambdaA.^4) ));
    Stretch{ii} = lambdaA;
    ActAreaStretch{ii} = lambdaA/lambdaPre;
end

%% Plotting

figure(1)
for ii=1:length(factor)
    plot(ActAreaStretch{ii},V{ii},'LineWidth',2)
    hold all
end
grid on
xlabel('actuated area stretch','FontSize',12)
ylabel('voltage (V)','FontSize',12)
set(gca,'FontSize',12)

```

A3. MATLAB code for studying DEAs prestretched by a rigid frame

This is the supporting data for section 3.4.

```

% Model of a circular membrane DEA secured onto a fixed frame
clear all; close all; clc;
%% input parameters
% DEA geometry

```

```

H = 2e-6;    % original electrode thickness (in the prestretched
state)
InnerRadius=10e-3; % inner radius of passive region (prestretched
state)
OuterRadius=50e-3; % outer radius of passive region (prestretched
state)
lambdaPre=2.5;
Z = 250e-6; % [m]
rel= 4.5; % relative permittivity
permit = 8.854e-12 * rel; % permittivity [Farads/m]
RA = InnerRadius/lambdaPre; % inner radius in ref. state
RB = OuterRadius/lambdaPre; % outer radius in ref. state

% Gent constitutive model
mu = 50e3;
Jlim = 125;
factor = 1; % number of times that of the elastomer
mu(1,2)=factor*50e3; % shear modulus of the electrode

MaxAct = 2; % maximum actuated stretch
StepSize = 0.01; % actuated stretch step size
step = ((MaxAct-1)/StepSize)+1; % number of steps for actuated
stretch
mesh = 10; % number of mesh points between RA and RB
lambdaAct = ones(1,step);
stressPassive = cell(1,step);

%% boundary value problem in passive region
syms lambdaR lambdaTheta
[DWDlambdaRVHB, DWDlambdaThetaVHB, D2WDlambdaR2VHB] =
Gent(mu(1),Jlim);

for ii=1:step
lambdaAct(ii) = 1+(ii-1)*StepSize;
lambdaA(ii) = lambdaAct(ii)*lambdaPre;

% obtaining stretches in inhomogeneous passive region
[x,y]=PassiveRegion(RA,RB,mesh,lambdaA(ii),lambdaPre,mu(1),Jlim);
Rstretch(ii)=y(1,1);
% true stress (passive region)
stressPassive{ii} =
y(1,1).*double(subs(subs(DWDlambdaRVHB,lambdaR,y(1,1)),lambdaTheta,y
(2,1)));
end

[DWDlambdaR, DWDlambdaTheta, D2WDlambdaR2] = Gent(mu(2),Jlim);
for ii=1:step
% obtaining stretches in homogeneous actuated region
% true stress in elastomer
stressElastomer =
lambdaA(ii).*double(subs(subs(DWDlambdaRVHB,lambdaR,lambdaA(ii)),lam
bdaTheta,lambdaA(ii)));
% true stress in electrodes

```

```

stressElectrodes =
lambdaAct(ii).*double(subs(subs(DWDlambdaR,lambdaR,lambdaAct(ii)),la
mbdaTheta,lambdaAct(ii)));

% finding the required voltage for that actuated stretch
voltage(ii) = real(sqrt((stressElastomer +
H.*lambdaPre.^2./Z.*stressElectrodes - stressPassive{ii}) * Z^2 /
(permit*lambdaA(ii)^4)));
EPressure(ii) = double(stressElastomer +
H.*lambdaPre.^2./Z.*stressElectrodes - stressPassive{ii});
EField(ii) = sqrt(double((stressElastomer +
H.*lambdaPre.^2./Z.*stressElectrodes - stressPassive{ii}) /
permit));
Rstress(ii)=double(stressPassive{ii});
Astretch(ii)=lambdaA(ii);
end

%% plot
ActStrain=(lambdaAct.^2-1)*100;
figure(1)
subplot(2,1,1)
plot(lambdaAct,voltage,'x-','MarkerSize',8)
grid on
title('Gent model')
xlabel('Actuated Stretch')
ylabel('Voltage (V)')
subplot(2,1,2)
plot(voltage,ActStrain,'x-','MarkerSize',10)
grid on
xlabel('Voltage (V)')
ylabel('Actuated area strain (%)')
%%%%%%%%%%%%%%%%%%%%%%%%%%%%%%%%%%%%%%%%%%%%%%%%%%%%%%%%%%%%%%%%%%%%%%%%

function [x,y]=PassiveRegion(IR,OR,N,lambdaA,PS,m,Jm)
options = bvpset('Stats','on');
solinit = bvpinit(linspace(IR,OR,N),@Initial);
sol = bvp4c(@ODEfun,@BCfun,solinit,options,PS,lambdaA,m,Jm);
x=sol.x; % radius in R
y=sol.y; % [radial stretch along R mesh ; circumferential stretch
along R mesh]
%%%%%%%%%%%%%%%%%%%%%%%%%%%%%%%%%%%%%%%%%%%%%%%%%%%%%%%%%%%%%%%%%%%%%%%%

function v = Initial(x)
v=[x./4
x./4];
%%%%%%%%%%%%%%%%%%%%%%%%%%%%%%%%%%%%%%%%%%%%%%%%%%%%%%%%%%%%%%%%%%%%%%%%

function dydx = ODEfun(x,y,PS,lambdaA,m,Jm)
% where x corresponds to the original variable r (radius)
% where y corresponds to the original variables
% y(1) = lambda1, y(2) = lambda2
syms R lambdaR lambdaTheta
[DWDlambdaR, DWDlambdaTheta, D2WDlambdaR2] = Gent(m,Jm);
DlambdaRDR = (DWDlambdaTheta-DWDlambdaR)/(R*D2WDlambdaR2);
DlambdaRDR = subs(DlambdaRDR,R,x);

```

```

DlambdaRDR = subs(DlambdaRDR, lambdaR, y(1));
DlambdaRDR = subs(DlambdaRDR, lambdaTheta, y(2));
DlambdaThetaDR= y(1)/x-y(2)/x;
dydx=[DlambdaRDR ; DlambdaThetaDR];
%%%%%%%%%%%%%%%%%%%%%%%%%%%%%%%%%%%%%%%%%%%%%%%%%%%%%%%%%%%%%%%%%%%%%%%%

function res = BCfun(ya,yb,PS,lambdaA,m1,a1,m2,a2,m3,a3,m4,a4)
% y(1) = lambda1, y(2) = lambda2
res=[ya(2)-lambdaA
yb(2)-PS];
%%%%%%%%%%%%%%%%%%%%%%%%%%%%%%%%%%%%%%%%%%%%%%%%%%%%%%%%%%%%%%%%%%%%%%%%

function [DWDlambdaR, DWDlambdaTheta, D2WDlambdaR2] = Gent(mu,Jlim)
syms lambda1 lambda2 lambda3 ...
lambdaR lambdaTheta
W=-0.5*mu*Jlim*log(1-(lambda1^2+lambda2^2+lambda3^2-3)/Jlim);
W=subs(W,lambda3,(lambdaR*lambdaTheta)^-1);
W=subs(subs(W,lambda1,lambdaR),lambda2,lambdaTheta);
% Obtaining dWdLambda for R and Theta
DWDlambdaR = diff(W,lambdaR);
D2WDlambdaR2 = diff(diff(W,lambdaR));
DWDlambdaTheta = diff(W,lambdaTheta);

```

A4. Simple 1D model describing the larger decrease in stress for elastomers with higher prestretch in a fixed-frame DEA setup

This is the supporting data for section 3.4.2.

In the passive region of a DEA prestretched by a fixed-frame, the drop in stress, with increasing actuated stretch, is larger for elastomers with a higher prestretch. This is because, for a given actuated stretch, although the absolute reduction in the passive area is the same, the relative reduction is larger for an elastomer with a higher prestretch, due to its smaller original dimensions. To show this, we shall use a simple one dimensional model to show that the reduction in stretch in the passive region is larger for a higher prestretched elastomer than a lower prestretched elastomer.

Assuming that the ratio of the active region's outer diameter to the passive region's outer diameter is the same regardless of the initial prestretch λ_{pre} , the absolute length of the passive region is fixed at l_0 , as shown in Figure 0.1. This means that for an elastomer with a higher prestretch, its original undeformed length L would be smaller than that for an elastomer with a lower prestretch. When the active region expands in area due to electromechanical

activation, it causes the passive region to decrease in length by d . Like l_o , d is absolute and remains the same regardless of the initial prestretch in the elastomer. Due to this decrease in length, the stretch in the passive region also decreases to a residual stretch of λ_{res} .

Figure 0.1. A one dimensional model showing the change in the passive region's length due to a stroke of magnitude d .

The undeformed length of the passive region is given by

$$L = \frac{l_o}{\lambda_{pre}}$$

Let us define the decrease in stretch in the passive region by $\lambda_{pre} - \lambda_{res}$, which is given by

$$\lambda_{pre} - \lambda_{res} = \frac{d}{l_o} \lambda_{pre}$$

Therefore, given that d and l_o are fixed, it can be seen that for DEAs with a higher prestretch, the reduction in stretch and therefore stress is larger.

A5. MATLAB code for determining the change in electrode area from photographs

This is the supporting data for section 4.1.

```

clear all; close all; clc;
% Get file names
filenames = uigetfile('*.jpg','multiselect','on');

%Loop over files
area = zeros(size(filenames));
for ii = 1:length(filenames)
    % for fixed threshold
    if ii==1
        temp = imread(filenames{ii});
        threshval = thresh_tool(temp(:,:,1));
    end
    % Call algorithm
    [area(ii)] = AreaMeasurementFcn(filenames{ii},threshval);
end

%% Save to excel with .csv
pixelNum = num2cell(area');
cell2csv('pixels.csv',pixelNum,'');
%%%%%%%%%%%%%%%%%%%%%%%%%%%%%%%%%%%%%%%%%%%%%%%%%%%%%%%%%%%%%%%%%%%%%%%%

function [pixels] = AreaMeasurementFcn(imgName,threshval)
%% Area Measurement Function
rgbImg = imread(imgName);

%% Segment the image
rlayer = rgbImg(:,:,1);
rlayer = imadjust(rlayer);

%% Manual Thresholding
%threshval = thresh_tool(rlayer);
electrode = ~im2bw(rlayer,threshval/255);

electrode = imclearborder(electrode);

L = bwlabel(electrode);
stats = regionprops(L,'Area');
A = [stats.Area];
[tmp,biggest] = max(A);
electrode(L~=biggest) = 0;

%% Fill up holes
electrode = imfill(electrode,'holes');

%% Calculate the number of pixels
pixels = nnz(electrode);

```

A6. Approximation of the sheet resistance of circular electrodes undergoing radial stretch

The sheet resistance for a radially-stretched circular electrode is just slightly less than the measured resistance. As such, the measured resistances given in the radial-stretch experiments can be approximated to be equal to the sheet resistance. Here, the method of determining the sheet resistance of a radially-stretched circular electrode is discussed.

The determination of the sheet resistance of the circular electrodes was carried out by sectioning the electrode into m segments, each of length Δd , as shown in Figure A.1. The width of the n^{th} segment w_n can be determined by finding the chord length at a distance of $n\Delta d$ from the centre of the circle.

Figure A.1. The circular electrode is divided into many segments and the segments are modelled to be in a series connection.

The measured resistance R is taken to be equal to the sum of the resistance of all the segments, as they are in series, wherein

$$R = R_1 + R_2 + R_3 + \cdots + R_m$$

Resistance is related to sheet resistance ρ/t , length l and width w by $R = \left(\frac{\rho}{t}\right)\frac{l}{w}$. Assuming homogeneity of the electrode, all the segments have the same sheet resistance. As such, the measured resistance is approximately equal to

$$R = \left(\frac{\rho}{t}\right)\frac{\Delta d}{w_1} + \left(\frac{\rho}{t}\right)\frac{\Delta d}{w_2} + \left(\frac{\rho}{t}\right)\frac{\Delta d}{w_3} + \cdots + \left(\frac{\rho}{t}\right)\frac{\Delta d}{w_m}$$

As such, the sheet resistance can be determined from the measured resistance by:

$$\frac{\rho}{t} = \frac{R}{\Delta d} \left[\frac{1}{w_1} + \frac{1}{w_2} + \frac{1}{w_3} + \dots + \frac{1}{w_m} \right]^{-1}$$

By running the MATLAB script shown below, it was found that the sheet resistance of the circular electrodes is just slightly smaller than the measured resistance of the electrodes, as shown in Figure A.2. As such, the measured resistances given in the radial-stretch experiments can be approximated to be equal to the sheet resistance.

Figure A.2. Sheet resistance versus measured resistance of circular electrodes undergoing radial stretch.

MATLAB script:

```
% Determining the sheet resistance of a circular thin flat electrode
load radialstretchR

strain{1} = crumpled(:,1)*100;
R{1} = crumpled(:,2);
strain{2} = graphite(:,1)*100;
R{2} = graphite(:,2);
strain{3} = sputtered(:,1)/2*100;
R{3} = sputtered(:,2);

for jj=1:3
    radius = 4e-2;
    n = 100;
    l = radius/n;
```



```

x = 0:1:radius;
w = 2*sqrt(radius^2-x.^2);
Wadd = 0;
for ii = 1:length(w)-1
    Wadd = Wadd + 1./w(ii);
end
Rs{jj} = R{jj}./2./1./Wadd;
end
semilogy(strain{1},Rs{1},'b-',strain{1},R{1},'b--',...
    strain{2},Rs{2},'k-',strain{2},R{2},'k--',...
    strain{3},Rs{3},'r-',strain{3},R{3},'r--',...
    'LineWidth',2)
xlabel('strain (%)','FontSize',12)
ylabel('resistance or sheet resistance (ohm)','FontSize',12)
set(gca,'FontSize',12)
grid on

```

A7. Comparison with crumpled gold leaf electrodes

The introduction of crumples or wrinkles in ELD silver thin film electrodes has helped to improve actuated strains of its DEAs. Here, using the same fabrication method outlined in section 7.1, we aim to determine if crumpling gold leaf electrodes would also be able to improve the actuated strains of its DEAs.

A7.1. Morphology of crumpled gold leaves

Like ELD silver thin films, the buckling in beaten gold thin films also becomes more severe with increasing compressive strains, as shown in Figure A.3. However, the buckles appear to be more like folds, and possibly delaminations, rather than wrinkles. This type of folding in thin films tends to occur when the substrate is very soft, such as water or gels [138, 51]. In this case, it is possible that the gold leaf was not well-adhered to the dielectric elastomer, thereby resulting in its delamination and therefore large folds. This is because with delamination, the thin film's substrate is air, therefore resulting in folds similar to that seen in crumpled sheet of paper. As shown in Figure A.4, there is a stark contrast between the morphology of the crumpled gold leaf and the crumpled ELD silver thin films. Although similarly compressed by 40% strain, the buckles in the gold leaf are sharp and spaced out, while the buckles in the ELD silver thin film are more rounded and finely spaced. This suggests that unlike the beaten gold thin films, the ELD silver thin films are well adhered to the substrate.

Figure A.3. Change in morphology as the beaten gold thin film was subjected to varying amounts of compressive strain. A lower magnification is shown on the left column while a higher magnification is shown on the right column.

Figure A.4. Difference in morphology between (left) crumpled gold leaf thin films and (right) crumpled ELD silver thin films, at the same magnification.

A7.2. Electromechanical performance of crumpled gold leaf DEAs

The maximum actuated area strain obtained by membrane DEAs using crumpled gold leaf electrodes was 73%, as shown in Figure A. 5, at a driving voltage of 1700V, which corresponds to an electric field of $69\text{V}\mu\text{m}^{-1}$. This is in comparison to an area strain of 128% attained by DEAs using crumpled ELD silver thin film electrodes at a driving voltage of 1800V, as discussed in section 7.4.

Figure A. 5. Membrane DEAs using 40% crumpled gold leaf electrodes, with a biaxial prestretch of 2.5, had a maximum actuated area strain of 73%, at a driving voltage of 1700V ($69\text{V}\mu\text{m}^{-1}$).

As shown in Figure A. 6, the extent of compression brought about differing actuated strain behaviour in its DEAs. Similar to DEAs using crumpled ELD silver thin film electrodes, the

best actuated strain performance was attained by DEAs with gold leaf electrodes that had been crumpled by 40% compressive strain. This was followed by DEAs with gold leaf electrodes that had been crumpled by 30%, 50% and then 20% compressive strain, with a maximum actuated area strain of 48%, 45% and 26% respectively. In contrast, the maximum actuated area strain attained by DEAs using flat gold leaf electrodes was 6%. This shows that the crumpling of gold leaf electrodes helps to improve the actuated strains attained by its DEAs.

Figure A. 6. Actuated area strain, with respect driving voltage, of DEAs using gold leaf electrodes that had been crumpled by 0%, 20%, 30%, 40% and 50% compressive strains.

By comparing the actuated strains of DEAs using crumpled gold leaf electrodes with that of DEAs using crumpled ELD silver thin film electrodes, as shown in Figure A. 7, two main observations can be made. Firstly, the DEAs using crumpled gold leaf electrodes generally attain lower actuated strains for a given electric field. Secondly, the DEAs using gold leaf electrodes tend to break down or to start a lengthy self-clearing process at lower electric fields. The reason for this might lie in different ways in which the thin films wrinkled or folded as a result of the applied compressive strains. As was discussed in section A7.1, the folds in the gold leaf are much courser than the wrinkles in the ELD silver thin film. Given

the same compression, wherein the same amount of material has to be packed into the same area, one can deduce that the amplitudes of the folds in the gold leaf are larger than that in the ELD silver thin film. The folding of the metal thin films into the dielectric can result in charge concentrations, thereby leading to premature failures and extensive self-clearing activity.

Figure A. 7. Comparison between the actuated strains of DEAs using crumpled ELD silver thin film electrodes and DEAs using crumpled gold leaf electrodes. The percentages indicate the initial extent of compressive strain in the thin film electrodes.

A8. Deviation from predicted actuated strains based on stiffness properties

In section 7.4, it was found that the simulated actuated strains, based on stiffness properties of the elastomer and electrodes, were unable to match experimental results. Here, we look at two main possible causes for the deviations. The first is the change in permittivity of the

elastomer with stretch and the second is a voltage drop due to increasing conductivity of the elastomer and high resistances of the electrodes.

A8.1. Permittivity change with stretch

We have assumed that the permittivity of the dielectric elastomer stays constant throughout the actuations. However, studies have shown that the relative permittivity of VHB elastomers varies with prestretch, temperature, frequency and the type of electrode used [168, 86, 174, 115, 21, 45]. Due to the complexity of the analysis and the wide range of empirical values in literature, the relative permittivity will be modelled to decrease linearly with increasing stretch in order to qualitatively show how a changing permittivity would affect actuated strains: at a radial stretch of 2.5, a relative permittivity of 4.5 was used, and at a radial stretch of 4, a relative permittivity of 3.6 was used (i.e. $\epsilon_r = -0.6 \lambda + 6$). This is opposed to the constant relative permittivity of 4.5 that was used in the model.

As shown in Figure A. 8, by assuming that the permittivity decreases with increasing stretch, the modelled actuated strains taper off to qualitatively resemble the experimental data. However, an accurate model will not be attempted here the dielectric elastomer's relative permittivity is affected by the many parameters mentioned above. In addition, as we aim to use crumpled ELD silver electrodes on a variety of elastomer substrates, an accurate model of one type of elastomer will not serve when another type elastomer is used instead.

Figure A. 8. Effect of decreasing permittivity of the dielectric material with increasing stretch on the DEA's actuated strains.

A8.2. Imperfect dielectric and importance of electrode resistance

The elastomer was assumed to be a perfect dielectric, while the electrodes were assumed to be perfect conductors that have a full-coverage of the elastomer. If the dielectric elastomer was a perfect insulator, there would be no leakage current through the elastomer. However, in practice, leakage current exists, due to the presence of impurities and imperfections in the elastomer [44]. The dissipative effects of leakage current is exacerbated at high electric fields of more than $40\text{V}\mu\text{m}^{-1}$, wherein the amount of leakage current increases exponentially [49]. As shown in Figure A. 9, leakage current starts to increase from a driving voltage of about 1000V (or an electric field of about $30\text{-}40\text{V}\mu\text{m}^{-1}$). Due to the higher leakage current at higher actuated strains and therefore electric fields, the electrostatic force tends to be lower than what was predicted in the model, which assumes the dielectric elastomer to be a perfect insulator. This tapering of electrostatic forces at high electric fields has also been observed in electro-adhesion actuators [87].

Figure A. 9. Increase in leakage current at higher voltages and electric fields.

The combination of the high resistance of the graphite and flat metal thin film electrodes, as well as the increased conductivity of the dielectric elastomer is likely to have caused a loss of voltage across the capacitor, thereby requiring a higher driving voltage to make up for resistive losses. Such a problem wouldn't have surfaced if the elastomer remained insulating to keep an open circuit in the steady state. The circuit shown Figure A. 10 is equivalent to the DEA when its electrodes have high resistances and when its dielectric elastomer becomes conductive. This typically occurs at large strains [26], as resistance of the electrodes tend to increase with strain, while leakage current increases with increasing electric fields. The voltage across the dielectric V_{de} is related to the voltage from the power source V

$$V_{de} = \frac{R_{de}}{2R_e + R_{de}} V \quad (0.1)$$

wherein R_{de} is the resistance of the dielectric elastomer and R_e the resistance of the electrode.

Figure A. 10. Equivalent circuit for a dielectric elastomer actuator with electrodes of resistance R_e , a dielectric elastomer of resistance R_{de} and a capacitance of C_{de} .

As the actual voltage across the dielectric elastomer is inversely proportionate to the resistance of the electrodes, a higher electrode resistance would lead to a higher voltage source being required to attain the same actuation. Therefore, although the sheet resistance of the electrodes does not have a significant effect on the electromechanical behaviour of its DEA at low strains, it would at higher strains. This is apparent in the electromechanical behaviour of DEAs at higher strains, wherein DEAs using graphite and metal thin film electrodes required a higher source voltage in order to attain the same actuated strains as DEAs using highly conductive crumpled ELD silver thin film electrodes.

Going hand-in-hand with the electrodes' resistance is their ability to retain a full coverage of the elastomer as it expands in area. The inability of graphite powder and flat metal thin film electrodes to maintain a full area coverage is also likely to have contributed to the plateauing of their DEA's actuated strains. For graphite powder, the already high resistance across the graphite electrode increases at an increasing rate when radially stretched beyond 18% radial strain, or about 40% area strain, as shown in Figure 7.4. As for the ELD silver thin film, even though resistance starts off low, resistance increases at an increasing rate from about 5% radial strain (10% area strain) and loses its conductivity at 10% radial strain (20% area strain). This corresponds quite well to the deviations that start to occur at an area strain of 10%, indicating that the stretchability of the ELD silver thin films were the predominant limiting factor of actuated area strains beyond 10%. However, this explanation does not hold for crumpled ELD silver electrodes, as its resistance does not vary much until about 100% area strain, as shown in Figure 7.4. Also, the capacitance of biaxially stretched crumpled silver electrode shows that within the range of actuated strains, the electrodes were able to maintain a full coverage of the elastomer as it expanded in area.

Relevant publications

Journal publications

1. Low S H, Shiau L L and Lau G K 2012 Large actuation and high dielectric strength in metallized dielectric elastomer actuators *Applied Physics Letters* **100** 182901
2. Low S-H and Lau G-K 2014 Bi-axially crumpled silver thin-film electrodes for dielectric elastomer actuators *Smart Materials and Structures* **23** 125021

Conference publications

3. Low S H and Lau G K 2011 High actuation strain in silicone dielectric elastomer actuators with silver electrodes vol 7976: SPIE)
4. Low S H, Tan A W Y, Shiau L L and Lau G K 2012 Actuated strains in excess of 100 percent in dielectric elastomer actuators using silver film electrodes. In: SPIE Smart Structures and Materials + Nondestructive Evaluation and Health Monitoring: International Society for Optics and Photonics) pp 834014--9
5. Low S-H and Lau G-K 2013 The effect of folds in thin metal film electrodes used in dielectric elastomer actuators. In: SPIE Smart Structures and Materials+ Nondestructive Evaluation and Health Monitoring: International Society for Optics and Photonics) pp 86872P-P-9
6. Low S H and Lau G K 2013 Effect of Crack Formation on Stretchable Silver Electrode for Dielectric Elastomer Actuators *Advances in Science and Technology* **79** 26-31