Mechanical Constraints Enhance Electrical Energy Densities of Soft Dielectrics

by

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Thesis submitted in partial fulfillment of
the requirements for the degree of Master of Science in the Department of
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ABSTRACT

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Abstract

In this thesis, a new method---mechanical constraint to increase the energy density of soft dielectrics is described. Electromechanical breakdown is one of the main factors limiting the energy density and it is induced by instabilities. Pull-in instability has been widely investigated and a theoretical model has been developed. A new type of instability—creasing to cratering instability is observed in soft dielectrics. A theoretical model is developed and shows perfect consistent with the experimental results. Further research has been done on creasing to cratering instabilities in deformed soft dielectrics. By suppressing these two kinds of instabilities, the electrical energy density could be enhanced nearly 9 times.

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Acknowledgement

First of all, I would like to express my gratitude to my advisor Professor Xuanhe Zhao, who led me to the field of electromechanics of solids with interesting projects. The many discussions during my research period kept me going with my study and research work. I would also like to thank Professor Piotr E Marszalek and Professor Teh Y Tan not only for the classes I have taken, but also the suggestion for my future study. I would like to thank all the SAM's lab members, especially Qiming Wang, Jianfeng Zang, Qing Tu for thoughtful discussion and cooperation. I would like to thank all staff in SMIF for helping me develop experimental skills. Finally, I would like to thank my friends and my family for always supporting me during the two years of study.

1. Introduction

1.1 Brief review of soft dielectrics

Dielectric, which was named by William Whewell, is an insulator that can be easily polarized under electric fields. Because of outstanding electric properties, dielectric materials are always used to control and stored electric energy for electric device and electric system. Among all kinds of dielectric materials, soft dielectrics attract more attention because of their merits contrasting with other dielectric materials, such as relative high energy density, high electric breakdown field, low dielectric loss, low cost, and grace failure^{1,2}. Numerous scientific and technological applications have been developed using soft dielectrics such as polymer capacitors, surface patternings³, polymer actuators^{4,5}, tunable optics⁶, electrocaloric refrigerators⁸, energy harvesters⁷. For these applications, the electrical energy densities of soft dielectrics are the key technical qualification. When an electric field is applied to soft dielectrics, electric breakdown will happen as the electric field goes beyond a critical value. So, electrical energy densities are strongly dependent on electric breakdown.

1.2 Electromechanical breakdown and Stark-Garton Model

When a high electric field is applied to dielectric materials, electric field will exert an electrostatic compressive force, which could change according to the strength of the electric field. When the electrostatic compressive force goes beyond a critical value

which cannot be balanced by the dielectric's elasticity, electromechanical breakdown occurs. In order to give a theoretical explanation to this phenomenon, K.H. Stark and C. G. Garton developed a model to analysis electric breakdown⁹. The main assumption is that on the equilibrium condition, the value of electrostatic compressive force equals to the mechanical compressive strength⁹⁻¹³. The electrostatic compressive force could be expressed as

$$\sigma_e = \frac{1}{2} \epsilon_r \epsilon_0 E^2 \quad (1.1)$$

Where σ_e is electrostatic compressive force, ϵ_r is relative dielectric constant, ϵ_0 is the dielectric constant of vacuum ($\epsilon_0 = 8.85 \times 10^{-12}$ F/m) and E is the electric field. Mechanical strength could be calculated by

$$\sigma = \mu \ln\left(\frac{d_0}{d}\right) \quad (1.2)$$

Where μ is the Young's modulus for materials, d_0 is the thickness of the dielectric polymers before applying an electric field, d is the reduced thickness due to compression and is a function of applied voltage. According to the previous assumption, at equilibrium state,

$$\frac{1}{2} \epsilon_r \epsilon_0 E^2 = \mu \ln\left(\frac{d_0}{d}\right) \quad (1.3)$$

When voltage exceeds critical voltage, the thickness of the film will not change with voltage. Mathematically this can be expressed

$$\frac{dd}{d\phi} = -\infty \text{ or } \frac{d\phi}{dd} = 0 \quad (1.4)$$

Where ϕ is the voltage ($\phi = \frac{E}{d}$). Using the equation (1.3) and (1.4), we could get critical true strain e_c and electromechanical breakdown strength E_c

$$e_c = \ln\left(\frac{d_0}{d}\right) = \frac{1}{2} \quad (1.5)$$

$$E_c = \left(\frac{\mu}{\epsilon_r \epsilon_0}\right) \quad (1.6)$$

Or using the initial thickness d_0 ,

$$E_a = E_c \left(\frac{d_c}{d_0}\right) = E_c \exp\left(-\frac{1}{2}\right) \quad (1.7)$$

1.3 Electric energy density of dielectrics

When a conductor is placed in an electric field, electric charges can easily flow in the materials. However, as an insulator, electric charges in dielectrics could only shift from equilibrium positions to cause dielectric polarization when applying electric field. In order to measure how easily dielectric materials polarizes due to electric fields, electric susceptibility X_e is used. Polarization density P could be expressed in response to an electric field E as

$$P = \epsilon_0 X_e E \quad (1.8)$$

The electric susceptibility of a material is related to relative electric dielectric ϵ_r by

$$X_e = \epsilon_r - 1 \quad (1.9)$$

The electric displacement D can be expressed by polarization density P and electric field E by,

$$D = \epsilon_0 E + P \quad (1.10)$$

Combining equation (1.8),(1.9) and (1.10),

$$D = \epsilon_r \epsilon_0 E \quad (1.11)$$

The electric energy density of a dielectric can be expressed as

$$u = \int D dE \quad (1.12)$$

Electrical breakdown of the dielectric limits the maximum electrical energy density it can achieve, so

$$u_{max} = \int_0^{E_B} D dE \quad (1.13)$$

where E_B is the breakdown electric field. Soft dielectric usually behave as ideal dielectrics, as shown in equation (1.11), where E and D are linearly related by a dielectric constant $\epsilon_r \epsilon_0$. So, for soft dielectrics, the maximum electrical energy density can be calculated as

$$u_{max} = \epsilon_r \epsilon_0 \frac{E_B^2}{2} \quad (1.14)$$

1.4 Failure of Stark-Garton Model and Power Law Model

Stark-Garton model could successfully predict the critical electric field for kinds of materials. They used the Young's modulus data provided by A. Charlesby and W. H. Hancock to calculate the critical electric field for polyethylene and shows perfect numerical agreement with experimental data⁹. However, the electric breakdown fields for some materials were found inconsistent with Stark-Garton Model. For example,

using Stark-Garton model to calculate the electric breakdown field overestimate the breakdown strength for poly(vinylidene fluoride-hexafluoropropylene) [P(VDF-HFP)] copolymer, as shown in figure 1¹⁴.

Figure 1:(a) Electric energy density of P(VDF-HFP) at room temperature. (b)The Weibull breakdown field E_b as a function of temperature¹⁴

The failure of Stark-Garton model is due to the erroneous evaluation the mechanical properties of P(VDF-HFP), which is a semicrystalline polymer. In practice, numerous soft dielectrics do not follow the elastic stress-strain relationship as shown in equation (1.2). Moreover, plastic deformation always occurs with elastic deformation before critical mechanical stress.

In order to give a more precise prediction to characterize the deformation for soft dielectric, X.H. Zhao et al. developed power law model¹⁵,

$$\frac{1}{2} \varepsilon_r \varepsilon_0 E^2 = K \left(\ln \frac{d_0}{d} \right)^N \quad (1.15)$$

Where K is dependent on the yield strength and N usually changes between 0.1 and 0.6 due to the plastic deformation. It is obviously that if N=1, equation (1.15) is equivalent to equation (1.3) to describe the pure linear elasticity. If N=0, equation (1.15) could be utilized to describe the ideal plastic deformation. Equation (1.15) gives,

$$\ln \left(\frac{d}{d_0} \right) = -\frac{N}{2} \quad (1.16)$$

$$E_c = \sqrt{\frac{2K}{\varepsilon_r \varepsilon_0}} \left(\frac{N}{2} \right)^{\frac{N}{2}} \quad (1.17)$$

Or using the initial film thickness,

$$E_a = E_c \frac{d_c}{d_0} = E_c \exp \left(-\frac{N}{2} \right) \quad (1.18)$$

Using $N=1$, equation (1.18) is the same as equation (1.7). However, in order to using equation (1.16)-(1.18), the stress-strain behavior should be measured to get the value for parameters K and N . Xin. et al got the value for K and N for P(VDF-HFP) at room temperature and plugged them into equation (1. 17) to predict the breakdown strength¹⁴. As shown in figure 1, the breakdown strength calculated by the new power-law is perfect consistent with experimental data.

Stark and Garton assumed the dielectrics deform homogeneous under the electric fields. Actually, deforming the large area of the soft dielectric under the electric field is impossible. As a voltage is applied on a layer of a soft dielectric, the thickness of the layer of soft dielectrics trend to decrease when the electric field increases, so that the same voltage induces a higher electric field. This positive feedback may cause some region of the dielectric to thin down quickly, leading to the electrical breakdown. This kind of instability causing by the positive feedback is termed by pull-in instability. Just because of the pull-in instability, the actually electric breakdown strength is much less than the prediction by Stark-Garton model, which is obviously shown in figure 1.

1.5 Pull-in instability

Actually, electromechanical instability has long been recognized as the reason for electric insulators failure in industry^{9,12}. For better prediction of the critical electric field

due to pull-in instability, X.H. Zhao et al.¹⁶ formulate a general method¹⁷ according to non-linear field theory of deformable dielectrics^{17,18,19,20}.

As shown in figure 2, a cuboid soft dielectrics with dimension $L_1 L_2 L_3$ before deformation. Mechanical forces P_1 and P_2 are applied to the $L_2 L_3$ and $L_1 L_3$ plane respectively. Electric voltage ϕ is applied to the $L_1 L_2$ plane. The deformation of the soft dielectrics is taken to be homogeneous and incompressible. Using $\lambda_1, \lambda_2, \lambda_3$ to denote the stretches in the L_1, L_2, L_3 direction respectively. So $\lambda_3 = \frac{1}{\lambda_1 \lambda_2}$. $\tilde{E}$ is used to denote the nominal electric field. In this case, $\tilde{E}$ equals to the voltage in the deformed state divided by the previous thickness of the soft dielectrics, $\tilde{E} = \frac{\phi}{L_3}$. On the contrary, the true electric field E equals to the voltage in the deformed state divided by the thickness of the soft dielectrics in the current state, $E = \frac{\phi}{\lambda_3 L_3}$.

Similarly, we could define the nominal electric displacement and true electric displacement, i.e. $\tilde{D} = \frac{Q}{L_1 L_2}$ and $D = \frac{Q}{\lambda_1 L_1 \lambda_2 L_2}$. Where, Q is the charge on an electrode in the deformed state. The nominal stresses for the $L_2 L_3$ and $L_1 L_3$ plane are $s_1 = \frac{P_1}{L_2 L_3}$ and

$$s_2 = \frac{P_2}{L_1 L_3}.$$

Figure 2: Schematic experimental setup for analyzing pull-in instability²⁰.

In this case, soft dielectric could be treated as an elastic dielectric with free-energy function $W(\lambda_1, \lambda_2, \tilde{D})$. The soft dielectrics with the weights and the battery constitute a thermodynamic system, the free energy of the system is

$$G = L_1 L_2 L_3 W(\lambda_1, \lambda_2, \tilde{D}) - P_1 \lambda_1 L_1 - P_2 \lambda_2 L_2 - \Phi Q \quad (1.19)$$

When small amounts $\delta\lambda_1, \delta\lambda_2, \delta\tilde{D}$ changes, the variation of the free energy of the system is,

$$\begin{aligned} \frac{\delta G}{L_1 L_2 L_3} = & \left(\frac{\partial W}{\partial \lambda_1} - s_1 \right) \delta \lambda_1 + \left(\frac{\partial W}{\partial \lambda_2} - s_2 \right) \delta \lambda_2 + \left(\frac{\partial W}{\partial \tilde{D}} - \tilde{E} \right) \delta \tilde{D} + \frac{1}{2} \frac{\partial^2 W}{\partial \lambda_1^2} \delta \lambda_1^2 + \frac{1}{2} \frac{\partial^2 W}{\partial \lambda_2^2} \delta \lambda_2^2 + \\ & \frac{1}{2} \frac{\partial^2 W}{\partial \tilde{D}^2} \delta \tilde{D}^2 + \frac{\partial^2 W}{\partial \lambda_1 \partial \lambda_2} \delta \lambda_1 \delta \lambda_2 + \frac{\partial^2 W}{\partial \lambda_1 \partial \tilde{D}} \delta \lambda_1 \delta \tilde{D} + \frac{\partial^2 W}{\partial \lambda_2 \partial \tilde{D}} \delta \lambda_2 \delta \tilde{D} \end{aligned} \quad (1.20)$$

According to thermodynamics, at equilibrium state, G should be minimized. For equation (1.20), at equilibrium, the coefficients of the first –order variations vanish, which means,

$$s_1 = \frac{\partial W}{\partial \lambda_1}, \quad s_2 = \frac{\partial W}{\partial \lambda_2}, \quad \tilde{E} = \frac{\partial W}{\partial \tilde{D}} \quad (1.21)$$

For ideal dielectric elastomer, the free-energy function could be denoted as

$$W(\lambda_1, \lambda_2, \tilde{D}) = \frac{\mu}{2} (\lambda_1^2 + \lambda_2^2 + \lambda_1^{-2} \lambda_2^{-2} - 3) + \frac{\tilde{D}^2}{2} \lambda_1^{-2} \lambda_2^{-2} \quad (1.22)$$

On the right hand side of this equation, the first term is elastic energy and the second term is the dielectric energy. Plug equation (1.22) into equation (1.21), the equilibrium equations will become

$$s_1 = \mu(\lambda_1 - \lambda_1^{-3} \lambda_2^{-2}) - \frac{\tilde{D}^2}{\varepsilon} \lambda_1^{-3} \lambda_2^{-2} \quad (1.23a)$$

$$s_2 = \mu(\lambda_2 - \lambda_2^{-3} \lambda_1^{-2}) - \frac{\tilde{D}^2}{\varepsilon} \lambda_2^{-3} \lambda_1^{-2} \quad (1.23b)$$

$$\tilde{E} = \frac{\tilde{D}}{\varepsilon} \lambda_1^{-2} \lambda_2^{-2} \quad (1.23c)$$

Considering a more general and special case, the elastomer is under equal biaxial stresses, which means $s_1=s_2=s$ and $\lambda_1=\lambda_2=\lambda$. Then equation (1.23) is simplified to

$$\frac{\tilde{D}}{\sqrt{\varepsilon \mu}} = \sqrt{\lambda^6 - 1 - \frac{s}{\mu} \lambda^5} \quad (1.24a)$$

$$\frac{\tilde{E}}{\sqrt{\frac{\mu}{\varepsilon}}} = \sqrt{\lambda^{-2} - \lambda^{-8} - \frac{s}{\mu} \lambda^{-3}} \quad (1.24b)$$

In the absence of the prestress ($s/\mu=0$), according to equation (1.24), the critical stretch $\lambda^c \approx 1.26$ to maximize $\tilde{E}$. This ratio corresponds to reduction in the thickness by around 37%. Plugging $\lambda_c = 1.26$ into equation (1.24b), we get

$$\tilde{E}^c \approx 0.69 \sqrt{\frac{\mu}{\varepsilon}} \quad (1.25)$$

The critical nominal electric field is dependent on the Young's modulus and dielectric field. Using the representative values $\mu=10^6$ N/m² and $\varepsilon=4*10^{-11}$ F/m, the critical nominal electric field is around 10⁸V/m, which is consistent with the reported breakdown fields²¹.

2. Creasing to Cratering Instability in Soft Dielectrics

2.1 Introduction to creasing to cratering instability

Pull-in instability happens on the soft dielectrics which are clamped with two compliant electrode under an electric field. If soft dielectrics are bonded on a rigid substrate on one side, there will be another kind of instability. Previous experiment setup usually applies an electric field to soft dielectrics bonded on a rigid substrate directly through an air gap^{3,22-25}. The common patterns for this electric-field –induced instability are pillars and strips^{3,22-25}. V. Shenoy et al. study the mechanism for this type of instability. The driving force for this instability process is the permeability difference between air and polymers, which competing with surface tension and elasticity of the polymers²⁶. They also prove the critical field for the instability is proportional to the square root of the polymer's modulus²⁶. But the main limiting for this experiment is the involving of air gap. The electrical breakdown for the air gap is around 10^6 V/m, so the maximum applied electric field could not go beyond this scale. In order to avoid this problems, we designed a new experimental setup, which could applied an electric field directly to polymers bonded on a substrate²⁷ without air gap. By using this experimental setup, a new type of instability in a elastic polymer bonded on substrate were observed under an ultrahigh electric field up to 10^8 V/m²⁸.

2.2 Experimental process

As shown in figure 3, Kapton, (DuPont, USA) a 125 μ m thick rigid polymer with Young's modulus of 2.5 GPa was used as the substrate. A polydimethyl siloxane (PDMS) based elastomer, Sylgard 184 (Dow corning, USA) was spin coated on the Kapton film. The PDMS films were cured at 65°C for 12 hours in the oven. Four kinds of PDMS with different crosslinker percentage changing from 2% to 5% were made as samples. Their shear modulus ranges from 6.7 kPa to 155 kPa, as shown in figure 4(a). The thickness of PDMS films was dependent on the spin coating speed. As shown in figure 4(b), the spin coating speed changes from 500rpm to 3000 rpm, then the thickness of PDMS films change from 40 μ m to 212 μ m. Micro-Strain Analyzer (TA Instruments, USA) was used to measure the shear modulus of PDMS films under a loading rate of $2.5 \times 10^{-4} \text{ s}^{-1}$. The experimental stress-stretch data were obtained and fitted to the neo-Hookean model to calculate the shear modulus of the PDMS films. Dektak 150 Stylus Profiler (Bruker AXS, USA) was used to measure the thickness of the films. A transparent conductive solution (20% wt NaCl solution) was used as conformal electrode contacting with the PDMS film. Because of the perfect transparency of conductive solution, the film deformation could be observed directly from a microscope (Nikon, Japan) above the solution. Voltage was applied between the metal substrate and the conductive solution by a high voltage supply (Matsusada, Japan). Ramping rate could be controllable by this equipment. The

modulus of Kapton is 1000 times higher than those of PDMS films, the deformation of the Kapton film could be neglected comparing with PDMS's deformation under applied voltages. So, the electric field in the PDMS film could not become dramatically high. So this new experimental setup could work under ultrahigh electric field to observe the instability patterns.

Figure 3: The experimental setup for observing the electrocreasing to cratering instability²⁸.

Figure 4: (a) The shear modulus of the PDMS elastomer as a function of the crosslinker concentration (b) The PDMS-film thickness as a function of the spin speed for PDMS with various crosslinker concentrations²⁸.

2.3 Experimental results

Evolution process of a new type of instability structures in PDMS film under an electric field was shown in figure 5. The ramping rate for the supply voltage is 10V/s. Evolution process could be divided into 6 procedures depending on the increasing of voltage. Initially, as shown in figure 5(a), the surface of the PDMS film keeps a flat and smooth state without obviously deformation. Secondly, some regions of the surface of PDMS films fold upon themselves suddenly to form a pattern of creases, shown in figure 5(b) and 5(h) when the voltage goes beyond critical value. Thirdly, the pattern of creases become larger and thicker and the density of creases decreases, on figure 5(c). Then, the center regions of some creases open dramatically changing to crater, so a pattern of

coexistent creases and craters form in the PDMS films shown in figure 5(d). As the voltage further rises, as shown in Figure 5(e), all creases change into craters eventually. Finally, the diameters of the craters increase, shown in figure 5(f). Because of the large deformation causing by craters, PDMS film would be fractured catastrophically at the craters.

Figure 5: Instability evolution in a substrate-bonded PDMS film under a ramping voltage: (a) flat state at 7 kV, (b) creased state at 8.8 kV, (c) crease coarsening at 9.7 kV, (d) coexistent states of creases and craters at 10.5 kV, (e) crater state at 13.3 kV, (f) craters with larger diameters at 15.6 kV. Schematic illustrations of the electric fields in a region at (g) flat, (h) creased, and (i) crater states²⁸.

2.4 Theoretical analysis for creasing to cratering instability

In order to further study this new instability process, driving force of this instability should be explained first. Actually, creasing instability has been widely observed on surfaces of soft materials under compression, such as bending an elastomer bar²⁹⁻³¹, condensing a polymer foam³², leavening a bread dough³³, and swelling a polymer gel bonded on a rigid substrate³⁴⁻³⁶. Creasing to cratering instability occurs in the polymer which is bonded on a rigid substrate under electric fields is due to the electric-field-induced stress in the film. When soft dielectrics are subject to an electric field, an electric-field-induced stress, known as Maxwell stress, is generated³⁷. Maxwell stress includes two parts: a tensile stress along the field and a biaxial compressive stress normal to the field. In the initial flat state, as shown on figure 5(g), the applied electric field is normal to the film is

$$E = \frac{\phi}{H+H_s\epsilon/\epsilon_s} \quad (2.1)$$

where ϕ is the applied voltage, the permittivity of Kapton is ϵ_s , and the thicknesses of the PDMS film and Kapton substrate are H and H_s , respectively.

The biaxial compressive stress induced by the electric field as shown in figure 5(g) depend on the electric field. As the increasing of voltage, the compressive stress will reach a critical value, which will lead the surface of PDMS to form creases. The further increase of the compressive stress will force the crease coarsening. As for our

experiment, the surface of the PDMS films could be treated as electrodes. Initially, the electric field is paralleled to the thickness of PDMS films [Figure 5(g)]. When the crease appears, the electric field in the will induced a tensile stress [Figure 5(h)], which would lead the creases to form creators [Figure 5(i)].

During the instability process, there are three main types of energies---electrostatic energy, elastic energy and surface energy per unit thickness. The depth of the crease is L , the electrostatic energy could be termed as $\sim \epsilon E^2 L^2$. The elastic energy per unit thickness of the crease region is $\sim \mu L^2$. The surface energy per unit thickness is $\sim \gamma L$, where γ is the surface tension of PDMS in water. In the instability process, the electrostatic energy decreases while the elastic energy and surface energy increase. The electrostatic energy competes with the increase of elastic energy and surface energy.

For PDMS films, $\gamma \approx 4 \times 10^{-2} \text{Nm}^{-1}$ ¹⁷, $\mu \approx 10 - 200 \text{kPa}$, $H \approx 40 - 200 \mu\text{m}$ and $L \approx 10 \mu\text{m}$. So elastic energy is on the order of $\sim 10^{-7}$ N, and surface energy is on the order of $\sim 10^{-8}$ N. It is obviously, the elastic energy dominates over the surface energy, balancing the elastic and electrostatic energies, gives

$$E_c \sim \sqrt{\mu/\epsilon} \quad (2.2)$$

The critical electric field could be calculated by equation (2.1). In equation (2.1), the critical voltages for the electro-creasing instability, the thickness of PDMS and Young's modulus could be measured through the experiment. The relationship between

critical voltage and the thickness of PDMS films of different Young's modulus was shown in figure 6(a). For the same modulus, the critical voltage is linearly dependent on the film thickness. Using equation (2.1) to calculate the critical electric fields E_c , using $H_s = 125 \mu m$, $\varepsilon_s = 3.5\varepsilon_0$, $\varepsilon_0 = 8.85 \times 10^{-12} Fm^{-1}$ and $\varepsilon = 2.65 \varepsilon_0$ ³⁸ and shown the relationship between E_c and $\sqrt{\mu/\varepsilon}$ in figure 6(b). It is clearly that E_c is linear with $\sqrt{\mu/\varepsilon}$, which is consistent with the previous conclusion in equation (2.2).

Figure 6: (a) The critical voltages and (b) Electric fields for the electrocreasing instability, and (c) the wavelengths of the craters in films with various thicknesses and modulus²⁸.

The crater and crease have the same energy as the electric field reaches certain value higher than E_c , as shown in figure 5(d), crease and crater will exist at the same time. The craters become more favorable as the electric field further rises. Craters locally arrange into polygons, the length of the polygons λ is nearly the same throughout the film as shown in Figures 5(e-f). During the instability process, the decrease of the electrostatic energy would reduce λ while the increase of elastic energy would increase λ . Finally, it will become equilibrium. As shown in figure 6(c), $\lambda \approx 1.5H$, independent of the modulus of the films.

More exactly calculation for the instability process was done according to the potential energy difference in different states. In this calculation, the effect of the substrate was neglect because it does not affect the critical field. The surface energy of the film is also neglected according to our previous discussion. So the potential energy in a unit thickness of the region only includes two parts, the electrostatic potential energy of the region and elastic energy. The potential energy in a unit thickness of the region can be expressed as

$$\Pi = \int_A W(\mathbf{F})dA + \int_A \frac{1}{2}\varepsilon|\mathbf{E}|^2dA - \int_s \phi\omega dS \quad (2.3)$$

Where $W(\mathbf{F})$ is the elastic energy density, $\mathbf{F}$ is the deformation gradient, ϕ is the applied voltage on the surface, A is the area of the region, ω is the surface charge density, and S is the contour of the region. The first term on the right hand side gives the elastic energy,

and the second and third terms represent the electrostatic potential energy of the region. The charge density on the surface of the film could be calculated by Gaussian law, $\omega = -\epsilon E \cdot \mathbf{n}$, where $\mathbf{n}$ is the surface normal. Substituting ω into equation (2.3), and using divergence theorem and Gaussian law, equation (2.3) change to

$$\Pi = \int_A (W(F) - \frac{1}{2} \epsilon |E|^2) dA \quad (2.4)$$

Figure 7: (a) Schematic illustrations of creased state in the film (b) The potential energy difference between the creased and flat states as a function of the applied electric field²⁸.

In a flat state, the electric field is uniform and the elastic energy is zero. So for flat state, the potential energy per unit thickness is equal to $\Pi_{flat} = -A\epsilon E^2/2$, E could be calculated by equation. (2.1). For the crease state, as shown in figure 7(a)⁴⁰, the thickness for the crease is L and the electric field is non-uniform in this region. ABAQUS 6.10.1 with a user subroutine UMAT were used to analyze the coupled deformation²⁸. Using equation (2.4), the potential energy of the creased state Π_{crease} could be calculated. The

region is treated as deform under plain-strain conditions. Moreover it obeys the neo-Hookean model with the elastic energy density is $\mu(F_{iK}F_{iK} - 3)/2$. Due to symmetry, only half of the region is calculated. In figure 7(a), only a small portion of the calculation domain is shown. The actual size of the calculation region is 100 times larger than the size of the crease. The potential energy difference has a form ³⁹

$$\Delta\Pi = \Pi_{crease} - \Pi_{flat} = \mu L^2 f(E\sqrt{\varepsilon/\mu}) \quad (2.5)$$

In equation (2.5), $f(E\sqrt{\varepsilon/\mu})$ is a dimensionless function of the applied electric field, which could be calculated by equation (2.1). Figure 7(b) show the relationship for $\Delta\Pi/(\mu L^2)$ and $E\sqrt{\varepsilon/\mu}$.

In absence of the applied field, the potential energy for flat state is $\Pi_{flat} = -\frac{A\varepsilon E^2}{2} = 0$, while the potential energy for the crease state gives a positive value because of the elastic energy. Thus the potential energy difference is positive, which indicates that the flat state is energetically preferable. As the electric field increase, because the creased state has a lower electrostatic potential energy than the flat state, so the potential energy difference decreases. When the electric field reaches to the critical value, the potential difference is 0, which indicates the creased state has the same potential energy as the flat state. So the crease begin to appears in the surface^{30,33,39}. As demonstrated in figure 7(b), when $\Delta\Pi = 0$,

$$E_c \approx 1.03\sqrt{\mu/\varepsilon}. \quad (2.6)$$

is consistent with the experimental data shown in figure 7(b)

$$E_c \approx 0.86\sqrt{\mu/\varepsilon} \quad (2.7)$$

The deviation for the theoretical and experimental results may be due to the ideal dielectric and elastic assumption. The actual behavior may be deviated from the neo-Hookean model.

2.5 Conclusion

We observe the creasing to cratering instability in the soft dielectrics film bonded on a rigid substrate. The driving force for crease is the in-plane biaxial compressive stress induced by the electric field. After a crease is formed, the electric field induces a tensile stress normal to the crease surfaces, tending to pull the creases open into craters. We developed a model by comparing the potential energies in the creased and flat states to predict the critical electric field. The prediction based on this model is consistent with the experimental results.

3. Creasing to Cratering Instability in Deformed Soft Dielectrics

3.1 Introduction to creasing to cratering instability in deformed soft dielectrics

In chapter 2, we focused on the creasing to crater instability in undeformed soft dielectrics. How about the electro-creasing in deformed soft dielectrics?

When electric field is subject to the sandwich structure shown in figure 3, compressive stress perpendicular to the electric field is induced by the electric field. When the compressive stresses reach critical values, some regions of the surfaces of the materials suddenly fold upon themselves to deform into patterns of creases. As discussed in Chapter 2, the driving force for the electro-creasing instability is the compressive stress parallel to the film which is induced by the electric field. The dielectric behavior of the soft dielectric is liquidlike and unaffected by deformation, the stress which is induced by electric field can be expressed as^{17,19,20}

$$\sigma^E = -\frac{1}{2}\epsilon E^2 \quad (3.1)$$

$$\sigma_{\perp}^E = \frac{1}{2}\epsilon E^2 \quad (3.1)$$

There are two kinds of electric-induced stresses, one is the biaxial compressive stress σ^E parallel to the film, which is perpendicular to the electric field, and the other is the tensile stress $\sigma_{\perp}^E$ normal to the film, which is along the electric field as shown in figure 8(a). Because the soft dielectric is regarded as incompressible, if a hydrostatic

stress is superimposed to the dielectrics, the deformation of the film will not be affected. As demonstrated in figure 8(a), a hydrostatic stress of $-1/2\epsilon E^2$ is superimposed onto the film, so the total stress normal to the film become zero. This operation results in an effective electrical stress¹⁵,

$$\sigma^E = -\epsilon E^2 \quad (3.2)$$

The effective electrical stress is compressive and along the biaxial directions parallel to the film as shown on figure 4(b). Moreover, the effective electrical stress increases with the applied electric field. When the electric field reaches a critical value, the electro-creasing instability occurs.

Figure 8: (a) The electrical stress in the film is superimposed by a hydrostatic stress to give an effective electrical stress. (b) The effective electrical stress plus the mechanical stress from the pre-stretch (c) gives the total stress in the film⁴¹.

If the soft dielectrics are pre-stretched before applying the electric field, what will happen?

3.2 Experimental process

Ecoflex 00-10 (Smooth-On, USA), which is a two-part silicone elastomer, has high deformability⁴². The two parts were mixed and spin-coated on a glass slide, which is covered by Scotch tape (3M, USA). Then the Ecoflex film is cured at room temperature for 12 hours. Different shear modulus changing from 5.4 kPa to 10.4 kPa was obtained by varying the volume ratio between crosslinker and base from 0.2:1 to 1:1. The thickness of Ecoflex changing from 227 μm to 724 μm was obtained by changing the spin-coating speed from 500 rpm to 150 rpm. Micro-Strain Analyzer (TA Instruments, USA) was used to measure the shear modulus of Ecoflex films under a loading rate of $2.5 \times 10^{-4} \text{ s}^{-1}$. The experimental stress-stretch data were obtained and fitted to the neo-Hookean model to calculate the shear modulus of the Ecoflex films. Dektak 150 Stylus Profiler (Bruker AXS, USA) was used to measure the thickness of the un-stretched films.

Figure 9(a) illustrates the experimental setup to study the electro-creasing instability of deformed polymers bonded on substrates. After the Ecoflex films cured, the films were peeled from the glass slide. Then, films were equally stretched in two orthogonal directions by a factor of λ_p as shown in figure 9(a). The stretched Ecoflex films were adhered to Kapton (Dupont, USA) substrate, which is a rigid polymer film, to

preserve the pre-stretch. The Kapton was bonded on a copper electrode with conductive epoxy (SPI, USA). A transparent conductive solution (20% wt NaCl solution) was used as conformal electrode contacting with the Ecoflex film. Because of the perfect transparency of conductive solution, the film deformation could be observed directly from a microscope (Nikon, Japan) above the solution. Voltage was applied between the metal substrate and the conductive solution by a high voltage supply (Matsusada, Japan). Ramping rate could be controllable by this equipment. When the electro-creasing instability was observed through the microscope, the voltage was recorded as the critical voltage.

Figure 9: Schematic illustrations of the current experimental setup for studying the (a) electro-creasing instability, (b) a pattern of electro-creases, (c) the electric fields in the polymers (d) a cross section of the creased film⁴¹.

Using the experimental setup shown in figure 10, the breakdown fields of pre-stretched VHB films could be measured. A 500 μm VHB film (3M, USA) was equally stretched in two orthogonal directions by a factor of λ_p and then bonded on a metal substrate. Silicone oil was used to immerse the top surface of the VHB film and a copper rod, immersed in the silicone oil, was placed vertically contacting with the VHB films. The diameter of the spherical tip of the copper rod is 8 mm. In order to avoid any movement during the tests, the rod was also mechanically constrained. The metal rod and substrate was used as electrodes. A ramping voltage was applied to this structure and the breakdown voltages were recorded until electrical breakdown occurred.

Figure 10: Schematic illustrations of the current experimental setup for measuring the electrical breakdown fields in pre-stretched VHB film⁴³.

3.3 Theoretical analysis for creasing to cratering instability in deformed soft dielectrics

The relationship for critical electric field and pre-stretch ratio λ_p are plotted in figure 11(a). Four groups of data are shown in figure 11(a) according to different film thickness and shear modulus. It is obviously that the critical voltages strongly depend on the pre-stretch ratio. But the relationship between voltage and pre-stretch is not monotonic. The relationship may be simple if the only variable is modulus. So we use critical electric field to take place of voltage in figure 11(b). Because Ecoflex is incompressible, the relationship for voltage and electric fields is $\phi = EH\lambda_p^{-2} + \frac{\varepsilon E}{\varepsilon_s}H_s$. So the electric field in the Ecoflex film can be calculated as

$$E = \frac{\phi}{H\lambda_p^{-2} + H_s\varepsilon/\varepsilon_s} \quad (3.3)$$

Here we use $H_s = 125 \text{ um}$, $\varepsilon_s = 3.5\varepsilon_0$, $\varepsilon_0 = 8.85 * 10^{-12} \text{ Fm}^{-1}$, $\varepsilon = 2.5 \varepsilon_0$ and $\varepsilon_s = 3.5 \varepsilon_0$ ^{42,44} to calculate the electric field. The critical fields are plotted as functions of the pre-stretch ratio in figure 11(b). It is clearly that for Ecoflex with same modulus, the critical fields are nearly overlapped at the same pre-stretch despite of the difference of thickness. Moreover, the critical electric fields increase along with the increasing of pre-stretch ratio. As we shown in Chapter 2, critical fields has a close relationship with $\sqrt{\mu/\varepsilon}$. We normalize critical fields by dividing $\sqrt{\mu/\varepsilon}$ and plot it in figure 11(c). For different thickness and modulus, the normalized critical electric fields are nearly the

same. From Figure 11(b) and (c), it is evident that biaxial pre-stretch greatly enhances the critical field for the electro-creasing instability.

Figure 11: The critical voltages (a) and electric fields (b, c) for the electro-creasing instability in polymers with different thicknesses, modulus, and pre-stretch ratios⁴¹.

In order to explain this phenomenon, we first go back to figure 8 to do the qualitative analysis. When the Ecoflex films are pre-stretched, a biaxial mechanical stress σ_p is induced. When an electric field is applied to pre-stretched films as shown in figure 8(b), the total stress in the film is the summation of the effective electrical stress and the mechanical stress, as shown in figure 8(c), that is

$$\sigma = -\varepsilon E^2 + \sigma_p \quad (3.4)$$

Because the mechanical stress is tensile, so pre-stretch counteracts the effect of the applied electric field. A higher electric field is needed than in the unpre-stretched in order to cause the electro-creasing instability. This is consistent with the experimental results given in figure 11(b) and (c).

We use the method discussed in Chapter 2 to quantitatively analysis the effect for pre-stretched. The potential energy in a unit thickness of the region in either flat or creased state can be expressed as

$$\Pi = \int_A (W - \frac{1}{2}\varepsilon|\mathbf{E}|^2)dA \quad (3.5)$$

where W is the elastic energy density, $\mathbf{E}$ is the electric field and A is the area of the region. The first term on the right hand of equation (3.5) gives the elastic energy, and the second term on the right hand gives the electrostatic potential energy of the region. The surface energy of the film can be neglected, because it is much smaller than the elastic energy.

Ecoflex films obey the neo-Hookean model within a stretch of 3 shown by the mechanical tests. So the elastic energy density is equal to $W = \mu(I - 3)/2$, where $I = \lambda_1^2 + \lambda_2^2 + \lambda_3^2$. λ_1 , λ_2 , and λ_3 are the principal stretches. Because of the incompressibility of silicone elastomer, $\lambda_1^2 \lambda_2^2 \lambda_3^2 = 1$.

At the flat state, the electric field is uniform and the elastic energy is zero. So for flat state, the potential energy per unit thickness is equal to $\Pi_{flat} = -A\varepsilon E^2/2$. At the crease state, as shown in figure 12(b)⁴⁰, the thickness for the crease is L and the electric field is non-uniform in this region. ABAQUS 6.10.1 with a user subroutine UMAT were used to analyze the coupled deformation²⁸. Using equation (3.5), the potential energy of the creased state Π_{crease} could be calculated. The region is treated as deform under plain-strain conditions. Due to symmetry, only half of the region is calculated. In figure 12(b), only a small portion of the calculation domain is shown. The actual size of the calculation region is 100 times larger than the size of the crease. The potential energy difference has a similar form with equation (2.5)³⁹

$$\Delta\Pi = \Pi_{crease} - \Pi_{flat} = uL^2 f(E\sqrt{\varepsilon/u}, \lambda_p) \quad (3.6)$$

where $f(E\sqrt{\varepsilon/u}, \lambda_p)$ is also a dimensionless function of the electric field and the pre-stretch ratio. In figure 12(c), potential energy difference for various pre-stretches is plotted as a function of the electric field. From figure 12(c), a higher pre-stretch ratio gives a higher elastic energy to form a crease in the film. Therefore, to balance the elastic

energy at the higher pre-stretch ratio, a higher electrostatic energy is required. As a result, the increasing of pre-stretch will increase the critical electric field, as shown on figures 11(c) and 12(c).

Figure 12: (a) Schematic illustrations of flat state in the film (b) Schematic illustrations of creased state in the film (c) The potential energy difference between the creased and pre-stretched states with various pre-stretch ratios⁴¹.

According to equation (3.4), $\sigma_c = -\varepsilon E^2 + \sigma_p$. For unpre-stretched films, i.e $\sigma_p = 0$ the critical electric field could be calculated by $E_c \approx 1.03\sqrt{\mu/\varepsilon}$ according to equation (3.6). For pre-stretched films, $\sigma_p \neq 0$, the critical electric field could be calculated as,

$$E_c \approx \sqrt{\frac{1.06\mu + \sigma_p}{\varepsilon}} \quad (3.7)$$

For soft dielectric films that obey the no-Hookean model, biaxial tensile stress is

$$\sigma_p = \mu(\lambda_p^2 - \lambda_p^{-4}). \quad (3.8)$$

Using equation (3.7) to calculate the electric field, electric field is plotted as functions of the pre-stretch ratio in figure 11(c). The theoretical prediction agrees well with the experimental and numerical modeling results as shown in figure 11(c).

3.4 Outreach

Breakdown electric field of a biaxial pre-stretched films dielectric elastomer could largely increase comparing with not pre-stretched films^{4,45}. G. Kofod et al. measured the breakdown field of VHB, one of the most widely used soft dielectric, as shown in figure 14(a)⁴⁵. One of explanations for this enhancement is the reduction of defect density^{4,45}. Here, we show the measured breakdown field of soft dielectric is extremely possible to be the electro-creasing critical field, which could be enhanced by pre-stretch.

The most common used experimental setup for measuring the breakdown fields of soft dielectrics is shown in figure 13(a)⁴⁵. Soft dielectrics were clamped by a pair of

metal electrodes, which can mechanically constrain the deformation of the film. The soft dielectrics are subject to a ramping voltage. The voltage was recorded to calculate the breakdown field, when observing the electrical breakdown.

Here we use VHB as the soft dielectric, brass as electrodes and observe the electrode-polymer interface under a microscope. As shown in figure 13(b), air bubbles appear on the interface. The breakdown field for air bubble is much lower than soft dielectric, so at a low voltage, air bubbles will be discharged and then work as a flexible electrode as illustrated in figure 13(c). Moreover, the electro-creasing instability will be induced in regions below the air bubbles when the electric field in the soft dielectrics reaches a critical value.

In the experimental setup shown in figure 13(a), there is no rigid substrate. So the instability will induce electrical breakdown of the dielectric elastomer, which means the breakdown field is equal to the critical field for the instability^{9,12,14}. It is common that the air bubble traps in the interface when measuring the breakdown fields of dielectric elastomers⁴⁵ and no comment on avoiding air bubble has been reported. Therefore, the measured breakdown fields are probably the critical fields for the electro-creasing instability in soft dielectric films.

Figure 13: (a) The previous experimental setup for measuring the breakdown electric fields of dielectric elastomers⁴⁵, (b) Air bubbles trapped on the polymer-electrode interface. (c) The discharged air bubbles may induce the electro-creasing instability in the dielectric elastomer⁴¹.

By pre-stretching the soft dielectric films, the critical fields of the electro-creasing instability could be enhanced, so the measured breakdown fields will increase as shown in figure 13(a).

We predict the critical fields (i.e. breakdown fields) by equation (3.7) with the dielectric and mechanical properties of the dielectric elastomer. The dielectric constant of VHB is $4.5\epsilon_0$. The nominal stress-stretch relation of VHB measured from a uniaxial tensile test with a stretch rate of 0.094 s^{-1} is shown in figure 14(b)²¹. Given the stiffening of the polymer at high stretches, Arruda-Boyce model is used to fit the nominal stress-

stretch data, in the equation s and λ are the nominal stress and stretch for uniaxial tension, and $I = \lambda^2 + 2\lambda^{-1}$, and n is a parameter resulting from the stiffening effect⁴⁶. As shown in figure 14(b), $\mu = 4.834 \times 10^4 \text{ Pa}$ and $n = 7.1$. The true stress in the biaxially stretched film equal to

$$s = \mu(\lambda_p^2 - \lambda_p^{-4})(1 + \frac{I}{5n} + \frac{11I^2}{175n^2} + \dots) \quad (3.9)$$

where $I = 2\lambda_p^2 + \lambda_p^{-4}$. By substituting equation (3.9) into equation (3.7), the breakdown fields for pre-stretched VHB films which measured with the method illustrated on figure 13(a) could be calculated. As shown figure 14(a), it is clearly that the theoretical prediction matches consistently with the experimental results.

According to D.B. Watson⁴³, the breakdown fields of pre-stretched VHB films could be measured with the experimental setup shown in figure 10. The spherical tip of the top electrode is very small. When the electric field is applied, the breakdown occurs under the spherical tip, the effect of air bubbles could be eliminated⁴³. For each pre-stretched ration, at least 10 samples were tested. In figure 14(a), the mean and standard deviation of the breakdown fields is shown. For the same pre-stretch ratios, using current method, the measured breakdown fields are much higher than the values measured from the previous method. When the pre-stretch ratio increases, the current measured breakdown fields increase milder than the previous measuring. These results

strongly support the hypothesis that the breakdown field measured using previous method is the critical electric field of the electro-creasing instability.

Figure 14: (a) The breakdown fields of VHB measured with the previous and current methods and the theoretical prediction of results from the previous method⁴⁵ (b) The nominal stress-stretch curve²¹ of VHB fitted to the Arruda-Boyce model⁴¹.

3.5 Conclusion

We show pre-stretching a soft dielectrics film could largely increase the critical field for the electro-creasing. Moreover, we develop a theoretical model to predict the critical field using the same method discussed in Chapter 2. The theoretical prediction matches well with the experimental results. We further show that the measured breakdown fields of dielectric elastomers are very possible to be the electro-creasing critical fields. Therefore, biaxial pre-stretches can greatly enhance the measured breakdown fields of dielectric elastomers. The theoretically predicted critical fields of VHB match well with experimentally measured breakdown fields under different pre-stretches.

4. Mechanical Constraints Enhance Electrical Energy Densities of Soft Dielectrics

4.1 Introduction to electric energy density

Recent years, high electric energy densities for soft dielectrics have been studied^{5,14,47,48}. Traditional methods to increase the electric energy density include developing new materials with high energy density and increasing the surface volume ration. For example, B.J. Chu et al. developed a defect-modified new soft dielectrics---poly(vinylidene fluoride) with ultrahigh energy density due to the high dipole density⁴⁸. Nanoporous materials, which have extremely surface volume ration, are always used to improve the electric energy density. In this thesis, a totally different method to achieve high-energy-density soft dielectrics is present. The electric energy density could be enhanced by 9 times through proper mechanical constraints of the dielectrics.

In chapter one, we has already shown the relationship between electrical breakdown and the electric energy density in equation (1.14), i.e. $\mu_{max} = \epsilon_r \epsilon_0 \frac{E_B^2}{2}$. It is obviously the electric energy density could be improved by improving the electrical breakdown. Previous discussion has shown two main electromechanical instabilities, which are the major cause for electrical breakdowns in soft dielectrics. If these two electromechanical instabilities could be suppressed, the electrical breakdown could be improved largely, leading to the improvement of the electric energy density.

4.2 Experimental setup

Figure 15: A film of a soft dielectric is under four types of mechanical constraints from electrodes: (a) no constraint, (b) one-side constraint, (c) two-side constraint with a defect, (d) two-side constraint⁵³.

As shown in figure 15 (a), soft dielectrics were clamped by two compliant electrodes. In our experiment, carbon grease was used as compliant electrodes, which

deforms freely with the dielectric. The compliant electrodes would not limit the deformation of the soft dielectrics. When this structure is subject to an electric field, the soft dielectric reduces its thickness and expands its area, so the same voltage induced a higher electric field. This positive feedback causes some regions of the soft dielectric to thin down drastically, which is known as pull-in instability. In figure 15(b), the soft dielectric is clamped by compliant electrode on one side and rigid electrode on the other side. Carbon grease is used as compliant electrode a thick layer (>1mm) of carbon epoxy is used as rigid electrode which can mechanically constrain the surface of the film. So when an electric field is applied to this structure, the compressive stresses induced by the electric field would force the some regions of surface folding themselves to form creases. As the electric field increase further, creases change to craters, this type of electromechanical instability is known as electro-creasing instability²⁸. In figure 15(c), the soft dielectric is clamped by rigid electrode on both sides with defects in one of the rigid electrodes. Defects can lead to the electro-creasing instability when voltage is subject to this structure. The diameter of the electrodes is 8mm, and the diameter of the defect is ~1.5 mm. In figure 15(d), the soft dielectric is clamped by rigid electrodes on both sides. As illustrated on figure 15(a) and (b), the electromechanical instabilities requires deformation of the soft dielectrics. So if both sides of the soft dielectric are constraint, no instabilities happen when an electric field is applied to this structure. Based on this fact,

we hypothesize the electrical breakdown fields and the energy densities of soft dielectrics could be extremely enhanced. In our experiment, three types of widely used soft dielectrics: VHB tapes (3M Inc.), Sylgard 527 silicone dielectric gel (Dow Corning Inc.), and Ecoflex silicone rubber (Smooth-on Inc.) were used. The Sylgard 527 and Ecoflex were fabricated into thin films using the spin-coating method^{28,41}, the thickness of the films are around 200 μm , and VHB tapes of 500 μm were used as received. The films of soft dielectrics follow the neo-Hookean law at moderate deformation^{21,28,49}. Micro-Strain Analyzer (TA Instruments, USA) was used to measure the shear modulus of Ecoflex films under a loading rate of $2.5 \times 10^{-4} \text{ s}^{-1}$. The dielectric constants of soft dielectrics were taken from their data sheets⁵⁰.

A high voltage supply (Matsusada, Japan) with a ramping rate of 500 V/s is used to apply a direct-current voltage between the electrodes on the dielectric film. The voltage was recorded as ϕ_B when the electric breakdown occurs. The thickness of the film at the undeformed state is H . So the breakdown field was calculated as $\frac{\phi_B}{H}$. The exact thickness of the soft dielectrics H was measured by Dektak 150 Stylus Profiler (Bruker AXS, USA). In our experiment, the changing of thickness of the film is less than 10%, so it is reasonable to use the undeformed thickness to calculate the breakdown field. This assumption underestimates the breakdown fields of no-constraint films in Figure 15(a)¹⁶, but this assumption is widely used in previous research^{9,14,51}. For these

three kinds soft dielectrics in four different models shown in Figure 15, at least 20 groups of samples were tested. The breakdown fields were evaluated by using two-parameter Weibull analysis ⁵²,

$$P_f = 1 - \exp [-(E/E_B)^{-\beta}] \quad (4.1)$$

Figure 16: The electric breakdown fields of two-side constraint Ecoflex with a defect were evaluated by the twoparameter Weibull analysis⁵³

where P_f is the probability of the soft dielectric film to breakdown below an electric field E , E_B is the characteristic breakdown field of the soft dielectric film, and β is a shape parameter. On figure 16, probability density as a function of the measured breakdown field for two-side constraint Ecoflex with a defect is plotted. A characteristic breakdown field can be fitted to be 25.38MV/m. Following this method, the characteristic

breakdown fields for soft dielectric films under four types of mechanical constraints (no constraint [figure 15(a)], one-side constraint [figure 15(b)], two-side constraint [figure 15(d)], and two-side constraint with a defect on the dielectric-carbon epoxy interface [figure 15(c)]) were calculated, and the results were plotted on figure 17(a).

4.3 Results and discussion

As shown on figure 17(a), for all these three kinds of soft dielectrics, the breakdown fields for no-constraint and one-side-constraint films are much lower than those of two-side-constraint films, which verifies our previous assumption---pull-in instability set in no-constraint films as shown in figure 15(a) and electro-creasing instability set in one-side-constraint films as shown in figure 15(b). Moreover, electrical breakdown for two-side-constraint with defects is similar comparing with those of one-side-constraint due to the electro-creasing instability which were set in by the defects. Because of two-side-constraint model can suppress these two types of instabilities, so the breakdown field is much higher than that of no-constraint and one-side-constraint model, which is consistent with the experimental results shown in figure 17(a). For instance, the breakdown field for two-side-constraint VHB is 3 times of that for no-constraint VHB. We further imaged the two-side-constraint films with defect after electrical breakdown to identify the breakdown locations, shown in figure 18. Electrical breakdown mostly occurs around the defects, regardless of the location of the defects.

This is because the defect allows deformation of the film beneath it and thus inducing the electro-creasing instability.

Figure 17: (a) The breakdown fields of soft dielectrics under various types of mechanical constraints (b) Electric energy densities of soft dielectrics under various types of mechanical constraints⁵³.

From previous discussion in chapter 1 and chapter 2, the critical electric field for the electromechanical instabilities can be expressed as

$$E_c = Z \sqrt{\frac{\mu}{\varepsilon}} \quad (4.2)$$

where K is a measurement of the stiffness of the polymer, such as Young's modulus or shear modulus, and Z is a non-dimensional factor depending on the mode of the instability. For pull-in instability, we get $Z \approx 0.69$ as shown in Chapter 1 and for electrocreasing instability, $Z \approx 1.03$ as shown in Chapter 2. Because for soft dielectrics, the electrical breakdown is induced by electromechanical instabilities, the breakdown fields of the dielectrics are nearly the same as the critical fields for the instabilities,

$$E_B \approx E_c = Z \sqrt{\frac{\mu}{\varepsilon}} \quad (4.3)$$

Combining equation (4.3) and equation (1.14), the maximum electrical energy density of ideal soft dielectrics due to different types of electromechanical instabilities can be calculated as

$$u_{max} = \frac{Z^2 \mu}{2} \quad (4.4)$$

Figure 18: The surface of two-side constraint and a defect soft dielectrics film after electrical breakdown⁵³.

According to equation (4.4), the electrical energy density is linear with the rigidity, which is agreement with the experimental results^{9,14,28,41,51}. What is more, the electrical energy density could be largely improved by suppressing all the possible electromechanical instabilities, i.e. to make $u_{max} > \frac{Z^2 \mu}{2}$. Using equation (4.4), the critical electric fields for pull-in instability and electro-creasing instability could be calculated and plotted in figure 17(a). Comparing with the theoretical values with the experimental

date shown in figure 17(a), it is obviously that experimental results is agreement with the theoretical analysis, even though less deviation. The inconsistency may be due to random factors such as defects in the samples and experimental errors.

We further compare the reported breakdown fields of the soft dielectrics from data sheets⁵⁰ with those of two-side-constraint films in **Table I**. It is obvious that the breakdown fields for two-side-constraint films are much higher (at least 2 times) than the values from the data sheets⁵⁰. These experimental and theoretical results prove that by proper mechanical constraints, the breakdown fields of soft dielectrics can be significantly enhanced by suppressing electromechanical instabilities.

Table 1: Breakdown fields of two-side-constraint soft dielectrics and reported values from data sheets⁵³

Breakdown Field	VHB	Ecoflex	Sylgard 527
	(MV/m)	(MV/m)	(MV/m)
Reported from data sheets²⁰	18	13.78	15
Two-side constraint	38.94	42.35	31.18

The electrical energy densities of soft dielectrics under four types of mechanical constraints were calculated with equation (1.14) and plotted in figure 16(b). It is clear that the two-side constraints significantly enhance the electrical energy densities of soft

dielectrics. For example, the electrical energy density of VHB is increased by 9 times by the two-side constraint.

4.4 Summary

We show that electromechanical instabilities including pull-in and electro-creasing instabilities is a major cause for electrical breakdowns of soft dielectrics. Proper mechanical constraints can greatly enhance the electrical breakdown fields and energy densities of soft dielectrics by suppressing the instabilities. We expect that the breakdown fields and energy densities may be further increased by increasing the adhesion strength between the soft dielectrics and rigid electrodes.

5. Summary and Future Work

In this thesis my experimental and theoretical studies on increasing electric energy density for soft dielectrics is present. Mechanical constraint, a new method, is used to enhance the energy density 9 times.

Creasing to cratering instability is one of the most common instability occurs when soft dielectrics are subject to an electric field. My group observes this type of instability, and developed a theoretical model to predict this instability, which is consistent with the experimental results. Moreover, we further study the creasing to cratering instability in deformed soft dielectrics films. A similar model was developed and showed perfect agreement with the experimental results. The theory also explains why pre-stretch soft dielectrics films could largely increase the electric field of soft dielectrics films. By using mechanical constraint method, which could suppress the instability of the soft dielectrics films, the electric energy density could be largely increased.

Based on the work in this thesis, I suggest conducting further research on increasing the adhesion strength between the soft dielectrics and rigid electrode to increase the strength of mechanical constraint. By doing that, the breakdown fields and energy densities may be further increased.

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