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Effect of Pre–Strain on the Dielectric and Dynamic Mechanical Properties of HSIII Silicone

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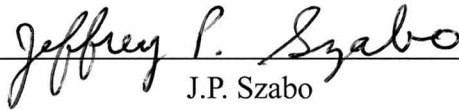
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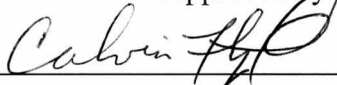
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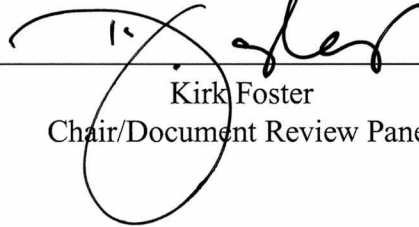
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Abstract

In this study, the mechanical and electrical properties of a silicone polymer were studied as a function of pre–strain in order to improve our understanding of dielectric breakdown phenomena in amorphous elastomeric materials. HSIII silicone (Dow Corning) films were prepared and studied by dynamic mechanical analysis, dielectric analysis, and dielectric breakdown experiments. It was found that the storage modulus increased significantly with uniaxial stretch, from 0.4 MPa in the unstretched state to 9.1 MPa at 250% uni-axial pre–strain. The mechanical loss factor was unaffected by pre–strain. The real and imaginary parts of the complex dielectric permittivity were also unaffected by the application of a biaxial pre–strain.

For HSIII films with no pre–strain applied, the dielectric strength increased with decreasing thickness. The dielectric strength was also found to be strongly dependent on pre–strain, with a near doubling of dielectric strength with a 200% uniaxial pre–strain applied. A series of experiments carried out over a range of film thicknesses and at two pre–strain levels (0% and 200% uniaxial) demonstrated that both pre–strain and thickness independently affected the observed breakdown strength, and that pre–strain was the more important factor over the thickness range studied (25–430 μm).

Résumé

Dans le cadre de cette étude, les propriétés mécaniques et électriques d'un polymère de silicone ont été étudiées en fonction d'une précontrainte appliquée, afin de mieux connaître le phénomène de claquage diélectrique dans les matériaux élastomères amorphes. Des pellicules de silicone HS III RTV (Dow Corning) ont été préparées et étudiées lors d'analyses mécaniques dynamiques, d'analyses diélectriques et d'expériences de claquage diélectrique. On a observé que le module de stockage augmentait considérablement en fonction de l'allongement uniaxial, de 0,4 MPa (non allongé) à 9,1 MPa avec une précontrainte uni-axiale de 250 %. Le facteur de perte mécanique n'a pas été affecté par la précontrainte. Les parties réelles et imaginaires de la permittivité diélectrique complexe n'ont pas été affectées non plus par l'application d'une précontrainte biaxiale. Dans le cas des pellicules HSIII sans précontrainte appliquée, la résistance mécanique diélectrique est inversement proportionnelle à l'épaisseur. La résistance mécanique diélectrique est également fonction de la précontrainte, et la résistance diélectrique double pratiquement lorsqu'une précontrainte uniaxiale de 200 % est appliquée. Une série d'expériences réalisées pour différentes épaisseurs de films et pour deux niveaux de précontraintes (unixiales de 0 % et de 200 %) ont démontré que la précontrainte et l'épaisseur affectaient de manière indépendante la résistance mécanique de claquage observée, et que la précontrainte était le facteur le plus important pour la plage d'épaisseurs étudiée (25 à 430 μm).

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Executive summary

Effect of Pre–Strain on the Dielectric and Dynamic Mechanical Properties of HSIII Silicone:

J.P. Szabo, R.S. Underhill, M. Rawji, I.A. Keough; DRDC Atlantic TM 2005-251; Defence R&D Canada – Atlantic; January 2006.

Background

One of the key technologies used in modern warships to reduce ship radiated noise is the use of resilient mounts to isolate the vibrations originating from marine engines and generators. Active vibration isolation shows great promise either as a replacement for, or in combination with passive isolation, which is currently used on Canadian Forces ships and submarines.

Dielectric actuators are promising candidates for active vibration isolation applications in military platforms where they can combine both passive as well as active isolation characteristics. Integration of passive and active aspects of vibration isolation into a single actuator offers several advantages over traditional approaches, including effectiveness at low frequencies, where passive isolation performance is inherently poor. For passive isolation, the dielectric elastomer may be formulated to have a Young's modulus near that of natural rubber ($\sim$ 2 MPa), the most commonly used material for vibration isolation. Other types of actuator materials, including shape memory alloys, piezoelectric polymers and ceramics, are generally too stiff to be useful for passive isolation, typically having Young's moduli hundreds or thousands of times larger than natural rubber.

Over the last few years, DRDC Atlantic has carried out research on dielectric polymer actuators under a Technology Investment Fund project, as well as a collaborative project with Victhom Human Bionics. The performance of these devices depends strongly (to the second power) on the maximum voltage that can be applied before dielectric breakdown occurs across the polymer film. Previous research had shown that for some materials, such as VHB acrylic elastomer (3M), the dielectric breakdown strength can be increased substantially (up to an order of magnitude) with uniaxial and biaxial stretching of the film. These studies suggested that the increased dielectric strength was caused by pre–strain, but they did not separate the effects of film thickness, which is also known to also influence the dielectric breakdown strength of thin films.

This report describes a set of experiments that were carried out to help improve the understanding of the effect of pre-strain on mechanical and electrical properties of polymeric films. HSIII from Dow Corning was selected for this study because films could be produced

in the laboratory with controlled thickness, as opposed to VHB acrylic which is factory made and available in only two nominal thicknesses.

Principal results

HSIII silicone (Dow Corning) films were prepared and studied by dynamic mechanical analysis, dielectric analysis, and dielectric breakdown experiments. It was found that the storage modulus increased significantly with uniaxial stretch, from 0.4 MPa in the unstretched state to 9.1 MPa at 250% pre–strain. The mechanical loss factor was unaffected by pre–strain. The real and imaginary parts of the complex dielectric permittivity were also unaffected by the application of a biaxial pre–strain.

For HSIII films with no pre–strain applied, the dielectric strength increased with decreasing thickness. The dielectric strength was also found to be strongly dependent on pre–strain, with a near doubling of dielectric strength with a 200% uniaxial pre–strain applied. A series of experiments carried out over a range of film thicknesses and at two pre–strain levels (0% and 200%) demonstrated that both pre–strain and thickness independently affected the observed breakdown strength, and that pre–strain was the more important factor over the thickness range studied (25–430 μm).

Significance of results

The results of this work improve our understanding of dielectric breakdown phenomena in thin films, which is critical to the performance of dielectric actuators. Dielectric actuators show enormous potential for use in future military platforms as actuators, sensors, and energy harvesting devices. The results are also relevant to the area of high energy density capacitors, which will likely find increasing use in the military as compact energy storage devices.

Future work

Follow on work will seek to further clarify the underlying mechanism by which pre–strain affects dielectric breakdown in polymeric films. Specifically, the effects of pre–strain on breakdown will be studied in the context of changes in polymer morphology (orientational effects) and modulus for a number of different materials. Future work will support the Supercapacitor TIF project beginning in April 2006.

Sommaire

Effect of Pre–Strain on the Dielectric and Dynamic Mechanical Properties of HSIII Silicone:

J.P. Szabo, R.S. Underhill, M. Rawji, I.A. Keough; DRDC Atlantic TM 2005-251; R & D pour la défense Canada – Atlantique; janvier 2006.

Contexte

L'une des technologies clés utilisées dans les navires de combat dans le but de réduire le bruit rayonné des navires est l'utilisation de supports élastiques ayant pour but d'isoler les vibrations produites par les moteurs et les génératrices des navires. L'isolation active des vibrations est prometteuse, soit comme remplacement de l'isolation passive qui est employée actuellement à bord des navires et des sousmarins des Forces canadiennes, ou conjuguée à celleci.

Les actionneurs diélectriques constituent des systèmes prometteurs qui permettraient d'assurer l'isolation active des vibrations sur des plates-formes militaires, où ils pourraient combiner des caractéristiques d'isolation passive et active. L'intégration de l'isolation passive et active des vibrations dans un actionneur unique offre plusieurs avantages par rapport aux méthodes classiques, y compris l'efficacité aux basses fréquences, lorsque le rendement de l'isolation passive est faible. Pour assurer l'isolation passive, l'élastomère diélectrique doit posséder un module d'élasticité voisin de celui du caoutchouc, qui est le matériau anti-vibration le plus couramment utilisé, soit un module d'environ 2 MPa. Les autres matériaux utilisés à cette fin, dont les alliages à mémoire de forme, les polymères piézoélectriques et les céramiques, sont généralement trop rigides pour assurer une isolation passive, car ils possèdent normalement un module d'élasticité qui est des centaines de fois, voire des milliers de fois plus élevé que le module d'élasticité du caoutchouc naturel.

Au cours des dernières années, RDDC Atlantique a réalisé des recherches sur les actionneurs diélectriques en polymère dans le cadre d'un projet réalisé grâce au Fonds d'investissement technologique, ainsi qu'un projet en collaboration avec Victhom Human Bionics. Le rendement de ces dispositifs dépend considérablement (un rapport carré) de la tension maximale pouvant être appliquée avant que survienne le claquage diélectrique de la pellicule en polymère. Des travaux de recherche réalisés précédemment avaient démontré que, pour certains matériaux, comme l'élastomère acrylique VHB (3M), la résistance au claquage peut être accrue de manière substantielle (jusqu'à un ordre de grandeur), avec étirement uniaxial et biaxial de la pellicule. Ces études laissent supposer que la résistance au claquage diélectrique accrue était causée par une précontrainte, mais elles n'ont pas distinguer les effets de l'épaisseur de la pellicule, qui est également reconnue pour avoir une incidence sur la résistance au claquage diélectrique des pellicules minces. Le présent

rapport décrit des expériences qui ont été réalisées dans le but d'aider à améliorer les connaissances des effets des précontraintes sur les propriétés mécaniques et électriques des pellicules polymériques. Des pellicules en silicone HSIII de Dow Corning ont été retenues pour cette étude en raison du fait qu'elles peuvent être produites dans le laboratoire et qu'on peut leur conférer une épaisseur contrôlée, contrairement à l'acrylique VHB qui est fabriqué en usine et qui n'est disponible que dans deux épaisseurs nominales.

Principaux résultats

Les pellicules en silicone HSIII de Dow Corning ont été préparées et étudiées lors d'analyses mécaniques dynamiques, d'analyses diélectriques et d'expériences de claquage diélectrique. On a observé que le module de stockage augmentait considérablement en fonction de l'allongement uniaxial, de 0,4 MPa (non allongé) à 9,1 MPa avec une précontrainte uniaxiale de 250 %. Le facteur de perte mécanique n'a pas été affecté par la précontrainte. Les parties réelles et imaginaires de la permittivité diélectrique complexe n'ont pas été affectées non plus par l'application d'une précontrainte biaxiale. Dans le cas des pellicules HSIII sans précontrainte appliquée, la résistance mécanique diélectrique est inversement proportionnelle à l'épaisseur. La résistance mécanique diélectrique est également fonction de la précontrainte, et la résistance diélectrique double pratiquement lorsqu'une précontrainte uniaxiale de 200 % est appliquée. Une série d'expériences réalisés pour différentes épaisseurs de films et pour deux niveaux de précontraintes (unixiales de 0 % et de 200 %) ont démontré que la précontrainte et l'épaisseur affectaient de manière indépendante la résistance mécanique de claquage observée, et que la précontrainte était le facteur le plus important pour la plage d'épaisseurs étudiée (25 à 430 μm).

Portée des résultats

Les résultats de ces travaux ont permis de mieux connaître le phénomène de claquage diélectrique dans les pellicules minces, qui constitue un aspect critique du rendement des actionneurs diélectriques. Les actionneurs diélectriques présentent de l'intérêt pour les plateformes militaires futures, comme les actionneurs, les capteurs et les dispositifs de stockage de l'énergie. Les résultats sont également pertinents pour ce qui est des condensateurs haute énergie, qui pourraient être de plus en plus utilisés dans le domaine militaire sous la forme de dispositifs de stockage d'énergie.

Recherches futures

Les travaux de suivi auront pour but d'éclaircir davantage le mécanisme sousjacent par lequel la précontrainte a une incidence sur le claquage diélectrique dans les pellicules polymériques. Plus particulièrement, les effets de la précontrainte sur le claquage seront

étudiés dans le contexte des changements qui surviennent dans la morphologie des polymères (effets d'orientation) et dans le module pour un certain nombre de matériaux différents. Les travaux à venir appuieront le projet de supercondensateur TIF qui débutera en avril 2006.

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1   Introduction

Dielectric polymer actuators are a promising new class of active materials. Under the influence of an external electric field, thin elastomeric films with complaint electrodes exhibit deformation due to the induced Maxwell stress. For unconstrained boundaries, the resulting actuation strain γ in the plane of a film of thickness d may be described by

$$\gamma = \frac{\varepsilon_\circ \varepsilon E^2}{2Y} \tag{1}$$

In Equation 1, $\varepsilon_\circ = 8.85 \times 10^{-12}$ F/m is the permittivity of free space, ε is the relative permittivity of the dielectric, E is the applied electric field, and Y is the Young's modulus. Equation 1 assumes that the Poisson's ratio of the elastomer is equal to 0.5. For small strains, the electric field is approximately equal to the applied voltage divided by the initial film thickness, $E = V/d$. According to Equation 1, the maximum achievable strain has a quadratic dependence on the maximum field that can be applied.

Kofod *et al.* have shown that uniaxial or biaxial pre-strain has a large effect on the measured dielectric breakdown strength of VHB polyacrylate films [1,2]. In fact, applying a 500% × 500% biaxial pre-strain to VHB films increases its dielectric breakdown strength $E_b = V_b/d$ from 18 MV/m to 218 MV/m [2]. As a result of the very high dielectric strength of pre-strained VHB films, it has been possible to fabricate dielectric polymer actuators with linear actuation strains exceeding 70% [3].

Application of a pre-strain to a polymer film results in alignment of polymer chains in the plane of the film as well as a decreased film thickness. For example, in the course of applying 500% × 500% pre-strain to VHB films, the thickness is decreased by a factor of ≈ 36 (assuming conservation of volume). Both polymer alignment and decreasing film thickness may contribute to increasing the dielectric strength of stretched VHB films, but the relative importance of each cannot be determined from data currently published in the literature.

This report describes a set of experiments that were carried out to help increase the understanding of the effect of pre-strain on mechanical and electrical properties of polymeric films. HSIII from Dow Corning was selected for this study because films could be produced in the laboratory with controlled thickness, as opposed to VHB acrylic which is factory made and available in only two nominal thicknesses: 1.0 mm and 0.5 mm. In addition, HSIII based actuators have shown an increased actuation strain when a pre-strain is applied in a manner similar to VHB [4].

2 Experimental

2.1 Film Preparation

HS-III silicone resin and clear catalyst (Dow Chemical, MI) were added in a 10:1 w/w ratio to a double planetary mixer (Charles Ross & Son, NY) and stirred under vacuum for 10 minutes at 20 RPM.

The mixture was then poured onto aluminum plates that had been previously cleaned with acetone. Drawdown blades (Gardco, FL) were used to cast films ranging from 5 to 25 mil. The films were allowed to cure for 48 hours in a closed plastic box to protect from dust.

After curing, the films were marked with gridlines spaced 1 cm apart using an indelible marker. They were then transferred to a cutting surface (to avoid marring the casting plate backing) by gently releasing most of the film from the backing and then sliding the cutting plate underneath it, laying down the film as carefully as possible.

To yield thinner films, a 3×3 cm section was cut out, and for thicker films a 8×8 cm portion was removed.

2.2 Breakdown Measurements

For breakdown measurements, films were tested with either no pre–strain or 200 % uniaxial pre–strain. Pre–strain was applied using a custom–made stretch apparatus (Martec Limited [5]), and determined by measuring the distance between the grid lines before and after stretching. The uniaxial pre–strain is defined here in terms of the initial length L_o and the final length L as

$$\gamma = \frac{L - L_o}{L_o} \tag{2}$$

Film thicknesses were measured using an eddy current probe (Gardco, Fl). To avoid the uncertainty associated with the pressure exerted by the probe, a microscope cover slip of known thickness was placed between the probe and the samples. This spread the force over a wider area.

Breakdown measurements were carried out using a method that involved a point contact electrode and a large area aluminum backing plate [6], as illustrated in Figure 1. For stretched films, a smaller aluminum plate was gently held against the bottom side of the film using a paper accordion.

Custom software controlling a NI-6036E data acquisition board (National Instruments) was used to drive a high voltage amplifier (Trek model 20/20C) through a slow voltage
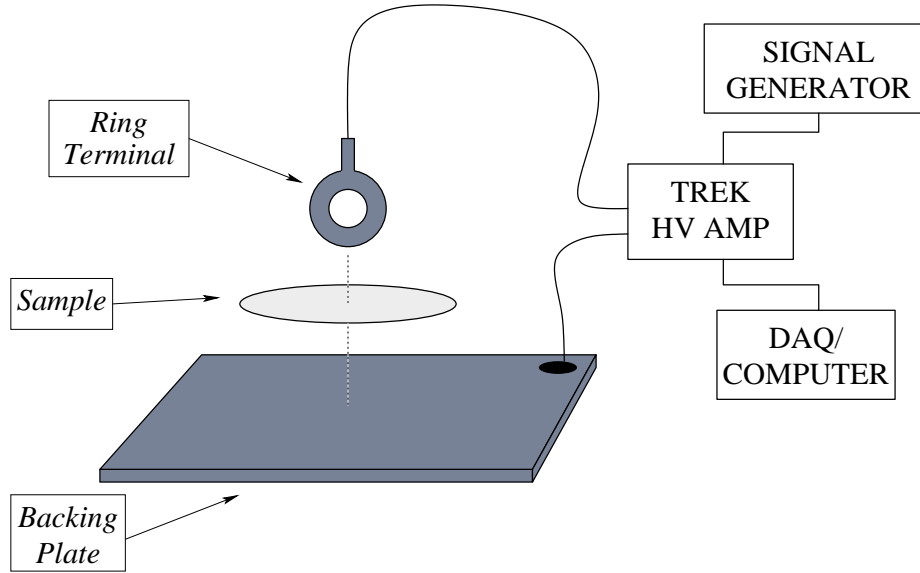
Figure 1: *Illustration of method used to carry out point breakdown experiments. Adapted with permission from Reference [6].*

ramp. Film breakdown caused the amplifier to enter a trip status, which was detected by the software. The breakdown field E_b was determined using Equation 3:

$$E_b = V_b/d \tag{3}$$

where V_b is the breakdown voltage and d is the film thickness.

2.3   Permittivity Measurements

Complex permittivity measurements were carried out using TA Instruments DEA 2970 Dielectric Analyzer with parallel plate sensors. Experiments were carried out at $20°C$ and over the frequency range 10^{-1} Hz to 10^5 Hz. The real and imaginary parts of the complex permittivity were determined for an HSIII film with no pre–strain applied as well as for an HSIII film with that was biaxially stretched. The thickness of the sample before stretching L_o, and after stretching L, was determined by the LVDT sensor located in the base of the DEA. Using the assumption of constant volume (see Section 3 and Equation (5)), the area strain was calculated from the change in sample thickness:

$$\text{area strain} = \frac{L_o - L}{L} \tag{4}$$

2.4 DMTA Measurements

The samples were analyzed in a TA Instruments Q800 Dynamic Mechanical Analyzer using the thin film tension clamps. The temperature was maintained at 20°C using a Gas Cooling Apparatus. The samples were excited at 0.1 Hz and at 1% dynamic strain amplitude. The samples were exposed to increasing pre–load (0.02 N to 2.50 N) values to obtain uniaxial pre–strains up to 150%. The sample length, before and after exposure to increasing pre–loads, was determined by the displacement sensor located in the base of the instrument. The complex modulus was determined from experimental force and displacement data using equations presented in Section 3.

3   Dynamic Mechanical Properties

For samples under large uniaxial pre–strains, the deformed length and cross-sectional area are significantly different from the undeformed values, and this must be accounted for in the calculation of complex modulus. While the length was directly measured during the experiment, the cross-sectional area was not. Rather, it was inferred from the initial dimensions and the measured length L, as described below.

For the purposes of this work, it was assumed that the Poisson's ratio $= 0.5$, and the volume was constant:

$$V_o = L_o A_o = L_o w_o d_o = V = LA = Lwd \tag{5}$$

where V_o, L_o, A_o, w_o, d_o are the initial volume, length, cross–sectional area, width and thickness, and V, L, A, w, d are the deformed volume, length, width and thickness of the sample. Constant volume is a good assumption for soft viscoelastic materials [7]. It follows from Equation 5 that the deformed cross-sectional area is given by $A = V_o/L$, and the dynamic stress amplitude may be expressed as:

$$\widetilde{\sigma} = \frac{\widetilde{F}L}{V_o} \tag{6}$$

where $\widetilde{F}$ is the dynamic force amplitude. The tilde symbol is used here to represent the amplitude of a complex variable, *i.e.*, $\widetilde{F} = |F^*|$. The dynamic strain amplitude is defined as:

$$\widetilde{\gamma} = \frac{\widetilde{L}}{L} \tag{7}$$

where the $\widetilde{L}$ is the dynamic displacement amplitude.

The Young's modulus Y is defined by the following equations:

$$Y^* = \frac{\sigma^*}{\gamma^*} = \frac{\widetilde{\sigma}}{\widetilde{\gamma}}\exp(i\varphi) \qquad (8)$$

where φ is the phase angle between stress and strain. It follows that the magnitude of the Young's modulus is given by

$$\widetilde{Y} = |Y^*| = \frac{\widetilde{\sigma}}{\widetilde{\gamma}} \qquad (9)$$

The complex Young's modulus may also be expressed in terms of its real and imaginary components, referred to as the storage Y' and loss modulus Y'', respectively. The following equations summarize the relationships among Y^*, Y', Y'', the loss factor η, and the phase angle φ:

$$Y^* = Y' + iY'' \qquad (10)$$

$$\eta = \frac{Y''}{Y'} = \tan\varphi \qquad (11)$$
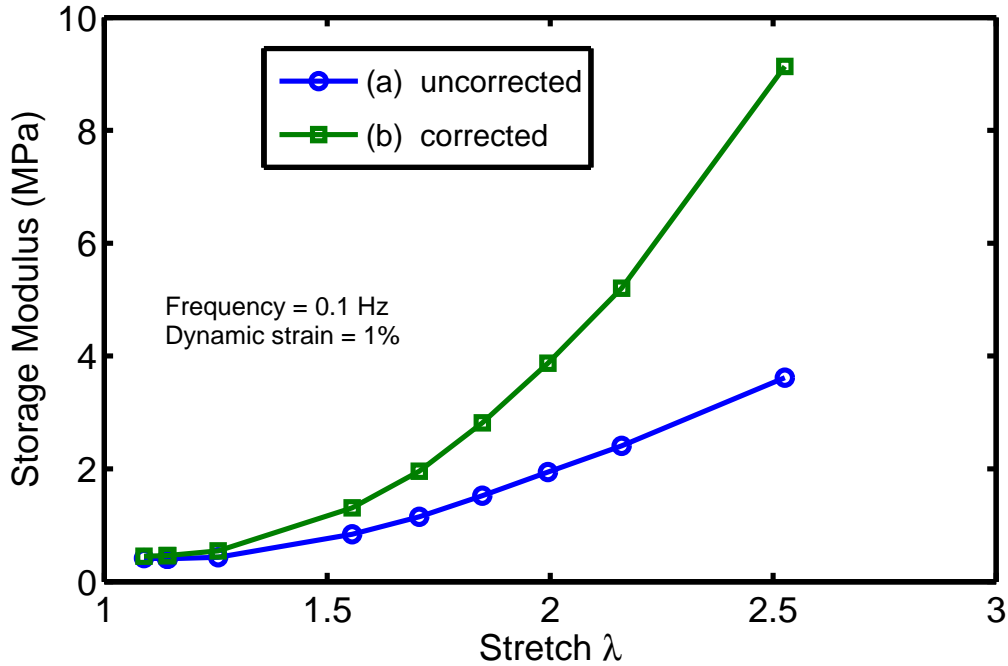
$$Y' = \widetilde{Y}\cos\varphi \qquad (12)$$

$$Y'' = \widetilde{Y}\sin\varphi \qquad (13)$$

It follows from Equations 6,7,9,and 12, that the storage modulus corrected for changes in cross-sectional area may be expressed as

$$Y' = \frac{\widetilde{F}L^2}{V_o\widetilde{L}}\cos\varphi \qquad (14)$$

Figure 2(b) shows the storage modulus of HSIII at 20 °C and 0.1 Hz calculated according to Equation 14, plotted as a function of the stretch λ, defined as

$$\lambda = \frac{L}{L_o} \qquad (15)$$

Figure 2: *Storage modulus of HSIII silicone at 20 °C and 0.1 Hz as a function of uniaxial stretch λ. (a) Uncorrected modulus, Equation 16. (b) Corrected modulus, Equation 14.*

For comparison, the uncorrected storage modulus (assuming cross-sectional area is constant, $A = A_o$) is presented in Figure 2(a). It was calculated by the TA Instrument DMA software according to the equation:

$$Y' = \frac{\widetilde{F}L}{A_o\widetilde{L}}\cos\varphi \qquad (16)$$

No corrections were necessary for the loss factor, which was computed according to Equation 11 and presented in Figure 3.

It is clear from Figures 2 and 3 that while the loss factor is essentially independent of stretch, the storage modulus has a strong dependence. In fact a stretch of $\lambda = 2.5$ is associated with a storage modulus of $Y' = 9.1$ MPa, compared with $Y' = 0.4$ MPa for the unstrained state ($\lambda = 1$). The increase in modulus with pre-strain in HSIII (a filled polydimethylsiloxane) is most likely due to strain induced crystallization, which is commonly observed in elastomers with a regular backbone structure such as *cis*–1,4–polybutadiene, butyl rubber, and natural rubber [8]. Other elastomers such as styrene butadiene rubber (SBR) show no effect of pre–strain on dynamic modulus [9].

It is interesting to note that the pre-strain had no significant effect on the mechanical loss
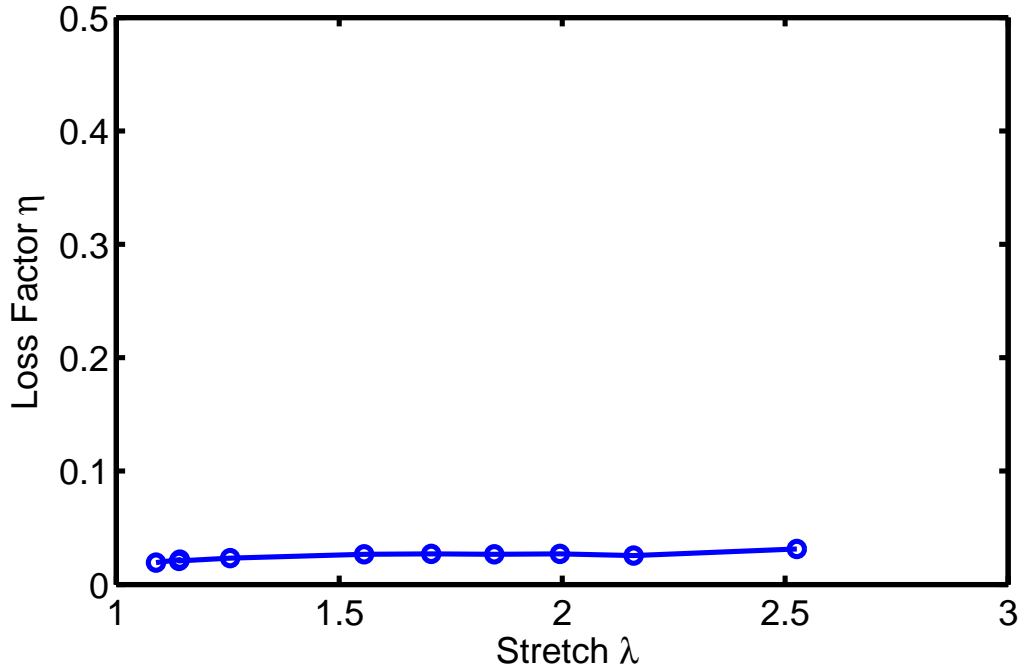
Figure 3: *Mechanical loss factor* η *of HSIII silicone at 20 °C and 0.1 Hz as a function of uniaxial stretch* λ.

factor (Figure 3).

4   Dielectric Permittivity

The dielectric properties of HSIII films are presented in Figures 4 and 5. Figure 4 shows the real part of the complex permittivity as a function of frequency for unstretched films, and for films stretched biaxially such that the area increase was approximately 38%, *i.e.* $A/A_o \sim$ 1.38. Figure 5 displays the frequency dependence of the imaginary part of the complex dielectric permittivity for stretched and unstretched HSIII films. The data suggests that there is little or no influence of pre-strain on the dielectric properties of HSIII. A change in dielectric properties with pre-strain is associated with electrostriction. The electrostrictive stress coefficient Q is defined by [10]

$$Q \equiv \frac{1}{2}\frac{\partial \varepsilon}{\partial \gamma} \tag{17}$$

For certain semi-crystalline polymers such as polyvinylidene difluoride (PVDF) and its co-polymers with trifuoroethylene or tetrafluoroethylene, electrostriction may contribute
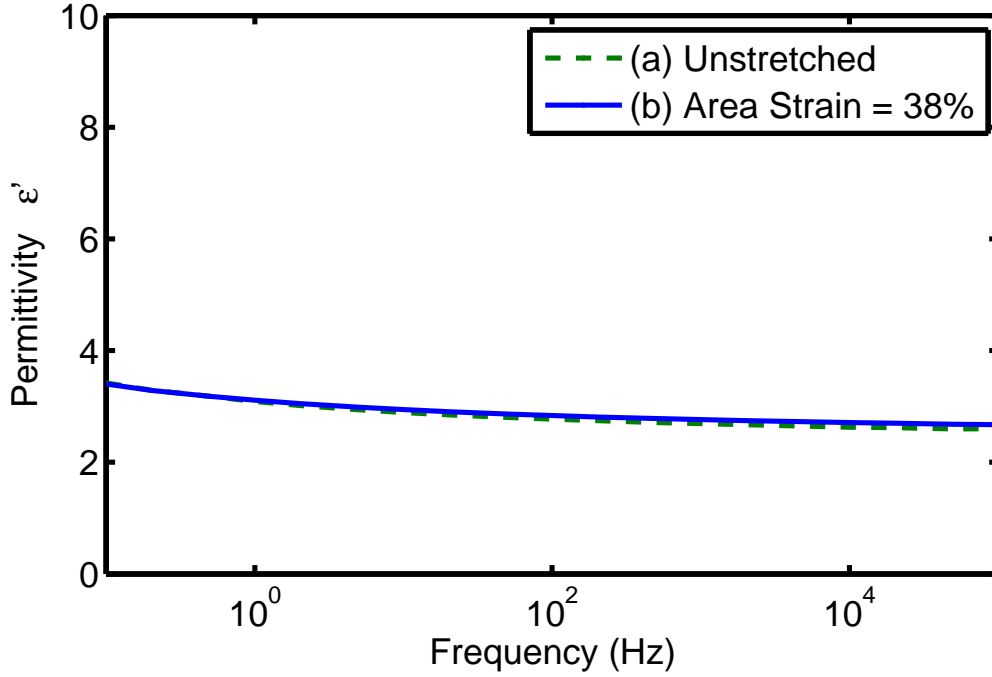
Figure 4: *Real part of complex dielectric permittivity of HSIII silicone at 20 °C for (a) unstretched film and (b) biaxially stretched film with area strain = 38%.*

up to 90% of the actuation strain in dielectric actuators. For soft amorphous polymers above their glass transition temperature, electrostriction is thought to make only a minor contributution to the total actuation strain [11]. Beslin [12] has explained the low electrostriction coefficients of soft elastomers in terms of low volume strain associated with the incompressibile rubbers (Poisson's ratio $\simeq 0.5$). The observed dielectric behaviour of HSIII silicone reported here is consistent with other soft elastomers such as VHB 4910 acrylic [2].

5   Dielectric Strength

In this section the dielectric strength of HSIII films is presented for various thicknesses and pre–strains. For uniaxial pre–strain, the stretches along the width and thickness axes λ_w and λ_d are equal, *i.e.*

$$\lambda_w = \lambda_d \tag{18}$$

$$\frac{w}{w_o} = \frac{d}{d_o} \tag{19}$$
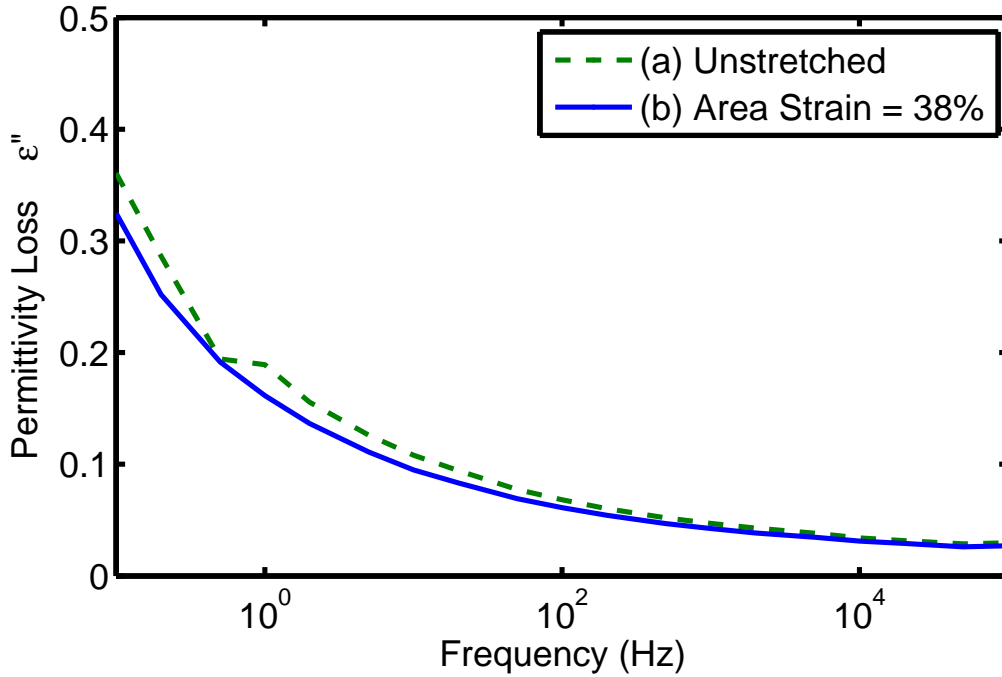
Figure 5: *Imaginary part of complex dielectric permittivity of HSIII silicone at 20 °C for (a) unstretched film and (b) biaxially stretched film with area strain = 38%.*

For constant volume conditions, it follows from Equations 5 and 19 that the final thickness d is related to the stretch along the length axis $\lambda_L = L/L_o$ by Equation 20:

$$d = \frac{d_o}{\sqrt{\lambda_L}} \qquad (20)$$

5.1 Constant Initial Thickness

A series of HSIII films were prepared with a fairly narrow range of initial thicknesses ranging from 130–153 μm. These films were stretched various amounts up to 200% (λ_L=3) along the length axis. The measured final thicknesses after stretching ranged from 65–130 μm. Figure 6 shows the calculated final thickness plotted as a function of the measured final thickness. As one can see from the figure, Equation 20 slightly overestimates the final thickness. Part of the explanation for this may lie in the fact that the deformation imposed on the films was not pure uniaxial stretch, since the width of the samples was a significant fraction of the length. Under such conditions, the thickness would be expected to vary over the area of the film, and deviate from Equation 20 to some extent.

Dielectric breakdown experiments were conducted on the stretched HSIII films described
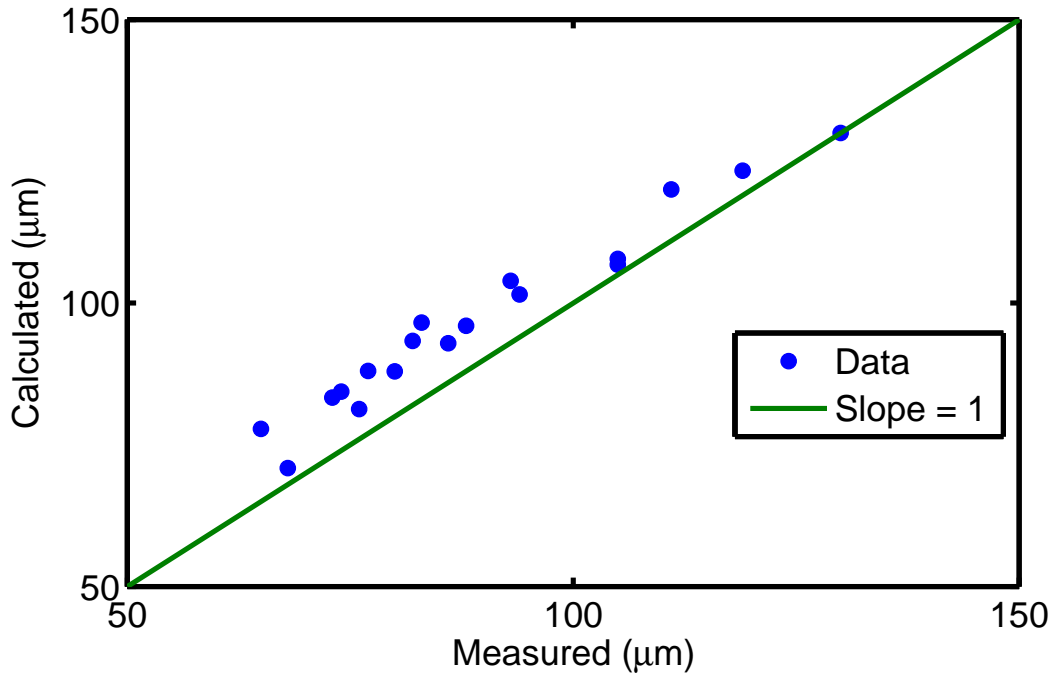
Figure 6: *Thickness of uniaxially stretched HSIII films calculated according to Equation 20 (ordinate) and measured thickness (abscissa). Solid line corresponds to a slope = 1.*

above. The breakdown results are presented as a function of strain in Figure 7. The error bars in breakdown fields in Figure 7 correspond to an uncertainty in thickness measurement of $\pm$ 5 μm. The dielectric strength increases with pre–strain, nearly doubling from 63 MV/m at no pre–strain to 108 MV/m at 200% pre–strain.

The dielectric breakdown data in Figure 7 are plotted as a function of final thickness (after stretching) in Figure 8. As the figure shows, the breakdown strength decreases with final thickness. However, since the final thickness and pre–strain are strongly correlated in this set of experiments, it is not possible to say if this decrease is caused by thickness or pre–strain effects.

5.2   Constant Pre–Strain

In order to improve our understanding of dielectric breakdown phenomena in HSIII silicone, a series of films were prepared over a wide range of initial thicknesses ranging from 25–430 μm. The dielectric breakdown strength of each film was determined in the unstretched state (λ_L=1) and at 200% strain (λ_L=3). The results of these experiments are presented in Figure 9. For unstretched films, the dielectric strength decreases monotonically with film thickness. For films stretched to 200%, the dielectric strength does not
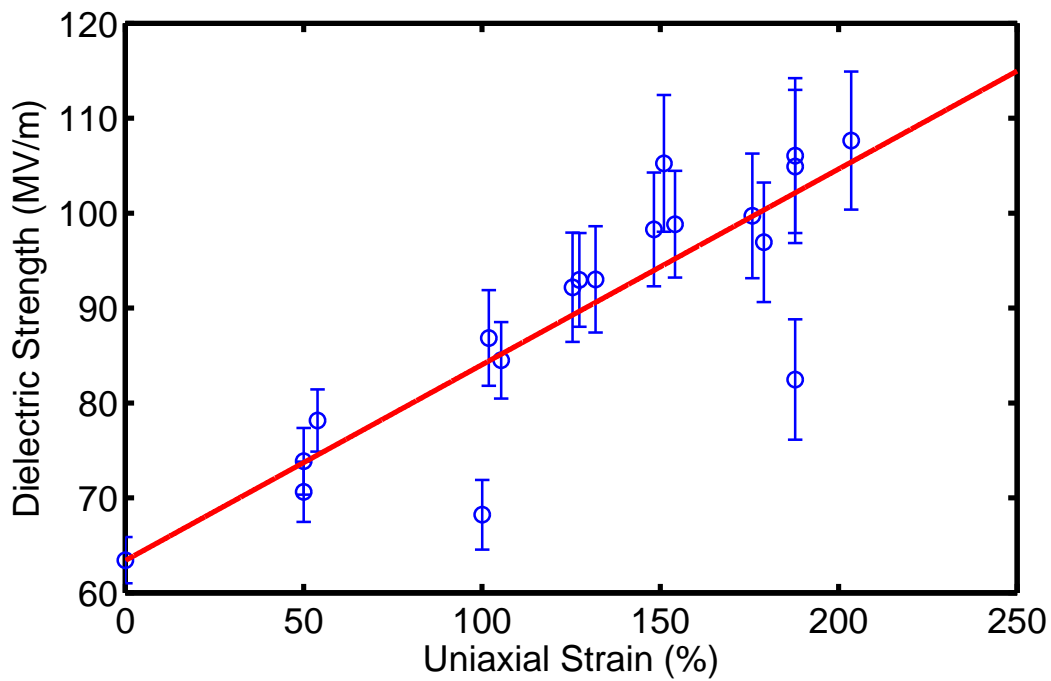
Figure 7: *Dielectric breakdown strength of HSIII films as a function of uniaxial pre–strain. Films were all approximately the same initial thickness (130-153 μm thick). Solid line is the linear least squares regression line. Error bars in dielectric strength correspond to ± 5 μm error in thickness measurement.*
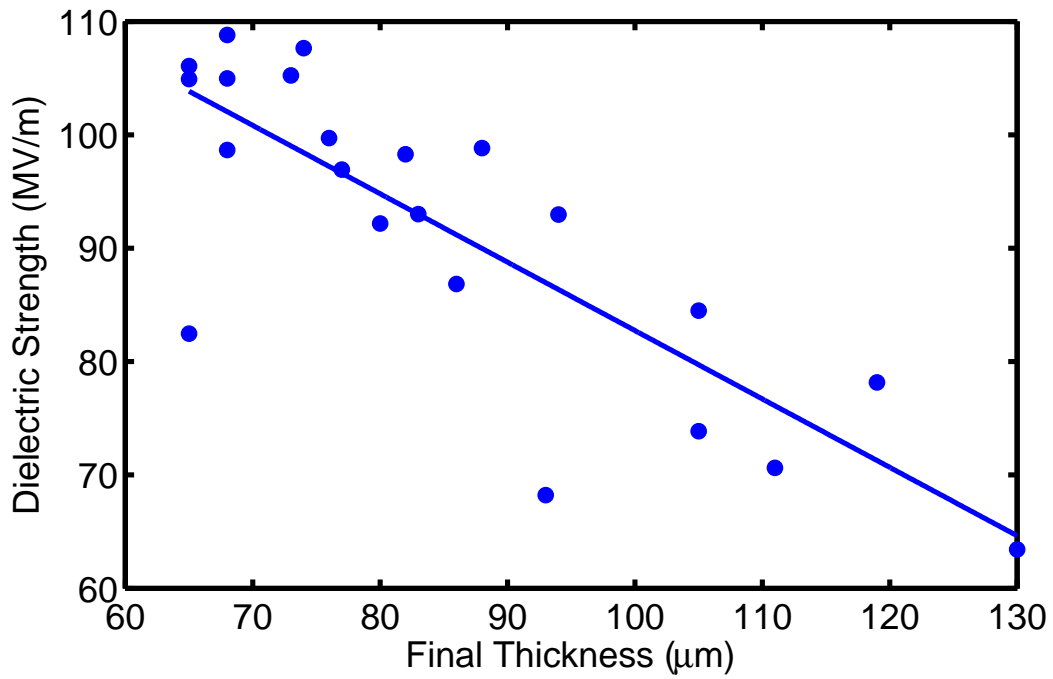
Figure 8: *Dielectric breakdown strength of stretched HSIII films as a function of final thickness. Films were all approximately the same initial thickness (130-153 μm thick). Solid line is the linear least squares regression line.*

decrease with increasing thickness, and in fact may increase slightly with increasing film thickness. Because of the scatter in the data at low thicknesses, the statistical significance of the slope in Figure 9(b) is marginal (slope $= 0.13 \pm 0.09 \times 10^{12}$ V/m).

The data in Figure 9 clearly shows that for the same final thickness, stretched HSIII films have a significantly higher dielectric breakdown strength than unstretched HSIII films. For example, at 177 μm film thickness, E_b =60 MV/m for unstretched films *versus* E_b =114 MV/m for 200 % strain. The effect of stretch is less clear at thicknesses below 50 μm, in part because of higher scatter in the data and larger error bars at low thickness.

It was proposed in Kofod *et al.* that the increase in the dielectric strength with pre–strain in VHB acrylic films is due to polymer chain alignment [13]. It was hypothesized that the collision cross-section between avalanching electrons and polymer atoms would be higher along the (aligned) polymer chains than between chains. While this is a reasonable hypothesis, there are alternative mechanisms that would explain the data in Reference 13 equally as well, including the effects of pre–strain on film thickness, modulus, defect distribution, and/or shape of air void defects. The results presented here suggest that for HSIII silicone films, pre–strain increases the dielectric strength, and this increase is not due to film thickness effects. Further study is required to test the hypothesis that chain alignment is the predominant mechanism for pre–strain effects in amorphous elastomeric films.

6   Conclusions

The effects of pre–strain on the properties of HSIII silicone films have been examined in this study. It was found that the storage modulus increased significantly with uniaxial stretch, from 0.4 MPa in the unstretched state to 9.1 MPa at 250% pre–strain. The mechanical loss factor was unaffected by pre–strain. The real and imaginary parts of the complex dielectric permittivity were also unaffected by the application of a biaxial pre–strain.

For HSIII silicone films with no pre–strain applied, the dielectric strength increased with decreasing thickness. The dielectric strength was also found to be strongly dependent on pre–strain, with a near doubling of dielectric strength with a 200% pre–strain applied. A series of experiments carried out over a range of film thicknesses and at two pre–strain levels (0% and 200%) demonstrated that both pre–strain and thickness independently affected the observed breakdown strength, and that pre–strain was the more important factor over the thickness range studied (25–430 μm). Future experiments are planned to elucidate the mechanism by which pre–strain affects dielectric breakdown strength in polymeric films.
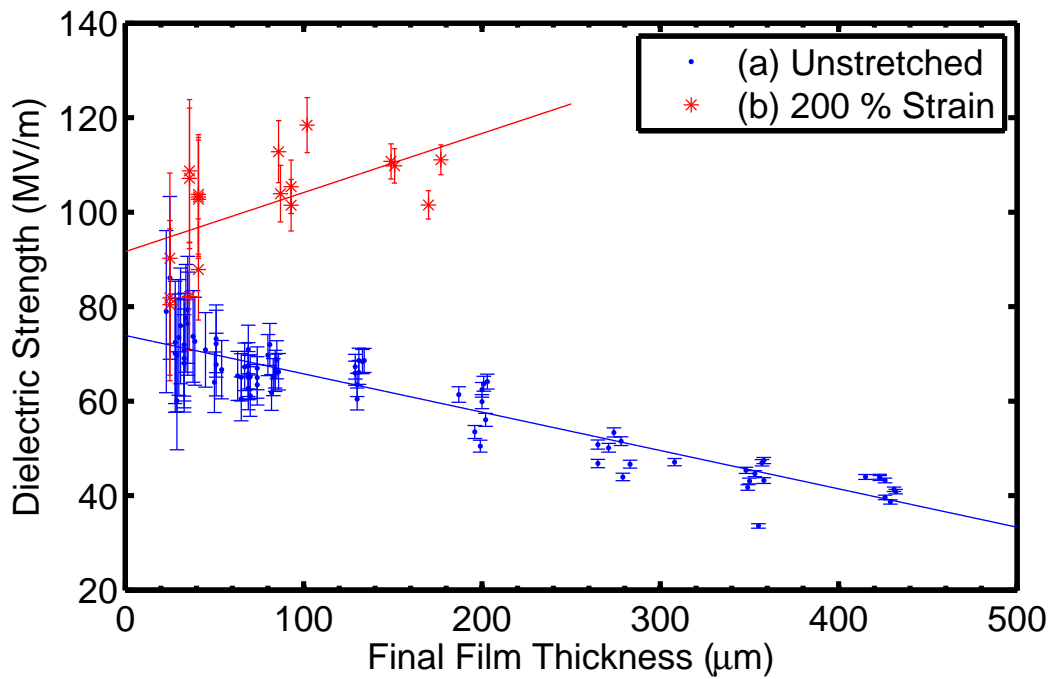
Figure 9: *Dielectric breakdown strength of HSIII films at two different pre–strain levels and over a range of final thicknesses. (a) Unstretched films. (b) Films with 200% pre–strain. Solid lines correspond to linear least squares regression lines. Error bars in dielectric strength correspond to ± 5 μm error in thickness measurement.*

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In this study, the mechanical and electrical properties of a silicone polymer were studied as a function of pre–strain in order to improve our understanding of dielectric breakdown phenomena in amorphous elastomeric materials. HSIII silicone (Dow Corning) films were prepared and studied by dynamic mechanical analysis, dielectric analysis, and dielectric breakdown experiments. It was found that the storage modulus increased significantly with uniaxial stretch, from 0.4 MPa in the unstretched state to 9.1 MPa at 250% uni-axial pre–strain. The mechanical loss factor was unaffected by pre–strain. The real and imaginary parts of the complex dielectric permittivity were also unaffected by the application of a biaxial pre–strain.

For HSIII films with no pre–strain applied, the dielectric strength increased with decreasing thickness. The dielectric strength was also found to be strongly dependent on pre–strain, with a near doubling of dielectric strength with a 200% uniaxial pre–strain applied. A series of experiments carried out over a range of film thicknesses and at two pre–strain levels (0% and 200% uniaxial) demonstrated that both pre–strain and thickness independently affected the observed breakdown strength, and that pre–strain was the more important factor over the thickness range studied (25–430 μm).

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