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Investigation into the Electromechanical Properties of Dielectric Elastomers Subjected to Pre-Stressing

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A B S T R A C T

Dielectric elastomers (DEs) are being exploited for biological applications such as artificial blood pumps, biomimetic grippers and biomimetic robots. Generally, polyacrylate and silicone rubber (SR) are the most widely used materials for fabricating DEs in terms of mixing with other polymers or compounding them with highly dielectric particles. Furthermore, pre-stretch offers an effective approach to increasing actuated strain and dielectric strength and eliminating 'pull-in' instability. In the work described here, a comparison in electromechanical properties was made between SR/10% barium titanate ($BaTiO_3$) and commercial VHB 4910. Trends in these dielectric parameters are shown graphically for variation in pre-stretch ratio (λ_{pre}). It was found that permittivity of SR/10% $BaTiO_3$ was independent of frequency, whereas permittivity was frequency-independent due to the polarization of polymer chains. The maximum deformation and the coupling efficiency for SR/10% $BaTiO_3$ can be achieved at a pre-stretch ratio between 1.6 and 1.9. For VHB 4910, they can be obtained in the pre-stretch ratio range from 2.6 to 3.0. A maximum energy density of 0.05 MJ/m³ was achieved by SR/10% $BaTiO_3$ ($\lambda_{pre} = 1.6$) and VHB 4910 ($\lambda_{pre} = 3.4$). The findings provide an insight into critical pre-stretch ratios required for a range of applications of DEs based on silicone and the commercially available polyacrylate VHB 4910.

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1. Introduction

Dielectric elastomers (DEs) are also termed dielectric electroactive polymers which are a subset of electroactive polymers (EAPs). The high dielectric properties of a DE can be used to induce a large actuated strain by the application of an electric field. DEs were discovered by Pelrine [1]. In his experiments, area strains of up to 30%, corresponding to actuation pressures as high as 1.9 MPa and high specific energy densities up to 0.1 J/g, were obtained. DEs have gained a deserved reputation as "artificial muscles" [2–4] because they behave similarly to biological muscles in terms of actuation pressure, energy conversion efficiency and response speed to stimulus. Hence DEs are used for intelligent applications such as tactile displays [5–7], bionic actuators [8–12] and other applications [13–16].

The working principle of DEs is illustrated in Fig. 1. When a high voltage is applied to the compliant electrodes which are coated onto the surfaces of a DE, the DE can quickly change its shape and when the electric stimulus is removed, the DE rapidly recovers its original shape. The pressure produced by the electrostatic force induced by the electric stimulus compresses the film and transfers electrical energy into mechanical energy [17,18].

DEs can be considered as isochoric (volume retaining materials) [4]. When a high voltage is applied, the relation between the area strain s_a and the thickness strain s_z (the compressive ratio in thickness), can be easily obtained from Eq. (1).

$$(1 + s_a)(1 - s_z) = 1 \tag{1}$$

The pressure, p which originates from the Maxwell stress, created between the positive charges on the top surface of the DE and the negative charges on the bottom surface of the DE, complies with Eq. (2) [19]:

$$p = \varepsilon' \varepsilon_0 \left(\frac{\Phi}{H}\right)^2 = \varepsilon' \varepsilon_0 \varphi^2 \tag{2}$$

where ε' is the relative permittivity (dielectric constant) of the DE material, ε_0 is the permittivity of the free space (8.85×10^{-12} F/m), φ is the electric field which equals the applied high voltage (Φ) divided by the initial thickness of the DE (H).

However, for a dielectric elastomer, the voltage-actuated strain is often limited by pull-in instability (electromechanical instability) [20] and electrical breakdown. As the applied voltage is increased, the thickness of the polymer decreases, so that this same applied voltage induces an even higher electric field. In this case, the pull-in instability may
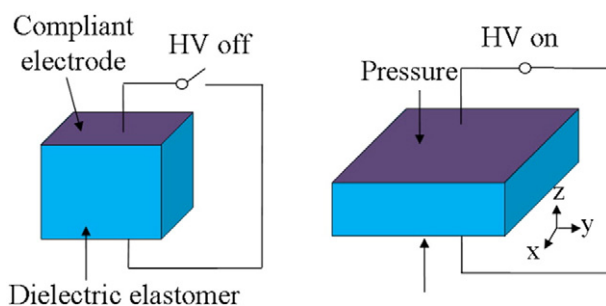
Fig. 1. Schematic representation of a DE's working principle.

cause the DE to reduce in thickness drastically and this can lead to electrical breakdown. Zhao [21] determined conditions for pull-in instability by analyzing voltage–stretch curves $\Phi(\lambda)$ deduced from stress–stretch curves $\sigma(\lambda)$ (Eq. (6)).

$$\Phi = H\lambda^{-2}\sqrt{\sigma(\lambda)/\varepsilon} \tag{6}$$

where $\varepsilon \approx \varepsilon' \varepsilon_0$ is the permittivity of elastomers.

Zhao assumed the material conformed to a neo-Hookean model and the application of the current induces a uniaxial compression kinematically equivalent to equi-biaxial stretching. As illustrated in Fig. 2, the critical stretch ratio of 1.26 was obtained by maximizing the voltage, which corresponds to a reduction in the thickness of 37%. Generally, the electric field will become unstable when the stretch ratio is above this value and pull-in instability may be induced.

The output properties of DE actuators can be improved by mechanically inducing uniaxial or equi-biaxial pre-stretch which has a significant effect on the material's performance, including improving breakdown strength [22,23] and reducing the effective compressive modulus which can enhance lateral actuation [14].

VHB 4910 is a polyacrylate, available as a commercial product from the 3 M Company. A VHB 4910 sample can exhibit a large deformation of about 100% [4] when a large voltage is applied. Kofod [23] proved that dielectric strength and electrostatic force were greatly increased by modelling cuboid DE actuators using VHB 4910 which has a relative permittivity of about 4.7. It was also found that the pull-in instability, which significantly affects the electrical performance of DEs, can be eliminated by applying a pre-stretch [24–27]. An investigation of the influence of pre-stretch on the electromechanical properties of DEs by
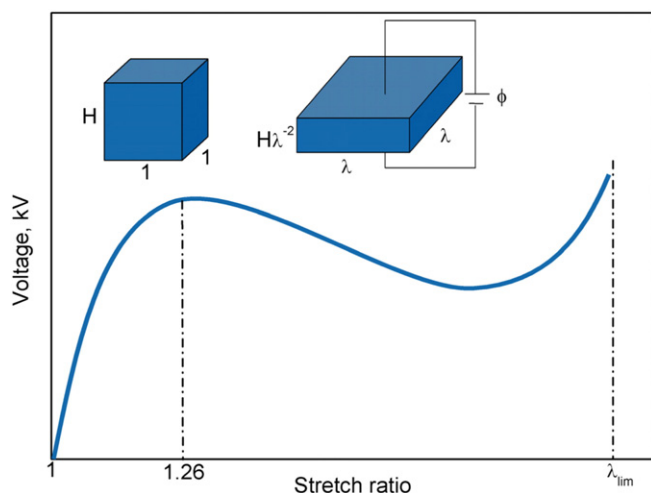
using VHB 4910 and DEs based on silicone rubber containing $BaTiO_3$ is presented here.

2. Materials

Two kinds of hyperelastic DE base materials were chosen for this research. A commercial silicone polymer, dimethylsiloxane (LSR 4305 DEV, Bluestar Ltd., U.S.A.), consisting of two parts (part A and part B) was used to fabricate DE samples and a commercial DE, VHB 4910, a polyacrylate from the UK 3 M company, having a thickness of 1 mm, was also employed. NYOGEL 756G (Nye Lubricants, Inc., USA) was chosen as the conductive carbon grease to be used as the compliant electrode for the DEs. $BaTiO_3$ was chosen as the filler material and added to the silicone to improve its permittivity. The particle size of the filler was below 3 µm and the density was 6.08 g/ml at 25 °C associated with a relative permittivity of 1200 at 1 kHz.

3. Experimental

3.1. Sample preparation

The silicone rubber used as the DE matrix contained two parts which were mixed at a ratio of 1:1. Barium titanate ($BaTiO_3$) was subsequently added to the silicone composite at a volume fraction of 10%. The mixture was stirred manually for 10 minutes and then poured into two moulds for making samples about 0.3 mm in thickness for different uses: one for mechanical tests and one for dielectric and electromechanical tests. After degassing for 1.5 hours, the samples were solidified in a heating room at a temperature of 60 °C for 12 hours.

3.2. Dielectric tests

Wideband dielectric spectroscopy was carried out on samples at 20 °C in the frequency range from 0.1 Hz to 10 MHz using a Turnkey broadband dielectric spectrometer. The cell used was a disposable gold-plated flat electrode with a diameter of 20 mm and thickness of 2 mm. Samples were placed in the gap between the electrode and the sample holder.

3.3. Electromechanical tests

The electromechanical test system consisted of a camera, a biaxial clamp for pre-stretching and a high voltage power supply ranging from 0 to 10 kV. Samples of 0.3 mm thickness were coated on both faces with the compliant electrode. The samples were clamped at the edges to give various pre-stretch ratios. A camera recorded changes in the area of the compliant electrode when an electric field was incrementally applied in voltage steps of 0.5 kV. Triplicate samples were tested.

4. Results and discussion

4.1. Dielectric tests

Generally, dielectric constant increases with the enhancement of polarizability of molecules in materials. Polar groups are reoriented under the electric field to promote the elongation of molecular chains. Therefore, the voltage-induced deformation resulted from not only the external Maxwell stress but also the internal polarization of DEs.

Fig. 3 shows the plot of dielectric constant related with frequency for SR, SR with 10% $BaTiO_3$ and VHB 4910. The dielectric constant of VHB 4910 fell dramatically from approximately 4.9 to 3.5 at a frequency of 1 MHz indicating that the polarization group had an obviously hysteretic response to the applied electric field. The dielectric constant of the silicone composite was measured at around 5 and changed only slightly
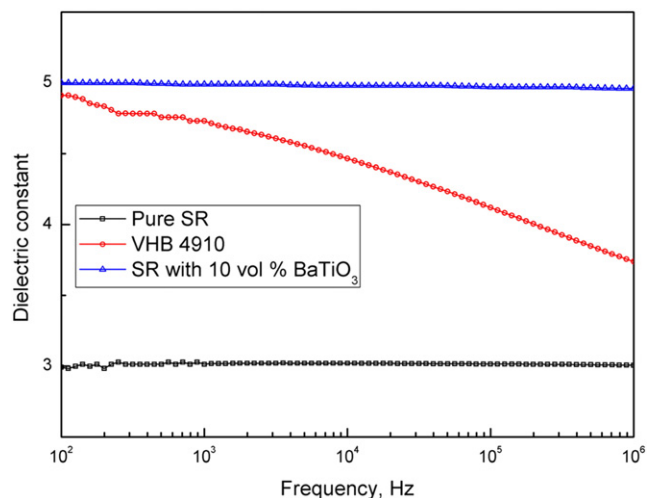

Fig. 2. A membrane of a dielectric elastomer subjected to a voltage reduces in thickness and expands in surface area. The voltage–stretch curve is typically not monotonic.

Fig. 3. Dielectric constant of silicone rubber, SR with 10% BaTiO$_3$ and VHB 4910 dependent on frequency.

over the range of test frequencies. When compared, the permittivity of silicone rubber LSR 4305 ($\varepsilon' \approx 2.9$) was lower than that of VHB 4910. This was probably due to the larger polarity of the ester group in VHB 4910 by contrast with that of the non-polar structure of the molecule of silicone rubber. When the high dielectric particles BaTiO$_3$ of 10 vol % were added in the silicone matrix, ε' increased to 5 and maintained this value in the wide frequency range from 100 Hz to 10 MHz. The appearance of dielectric enhancement is related to the Maxwell Wagner effect in the thin DE film [28]. This indicated that SR based DEs were more stable than VHB 4910 in respect of dielectric properties when subject to changes in frequency.

4.2. Electromechanical tests

The 3 cm square samples of silicone rubber with 10% barium titanate were stretched from their initial size in steps of 1 cm and to a final size of 6.5 cm square which was achieved just prior to tearing. The 2.5 cm square VHB 4910 samples were stretched, also in steps of 1 cm, from their original size to a final size of 9.5 cm × 9.5 cm before experiencing tear.

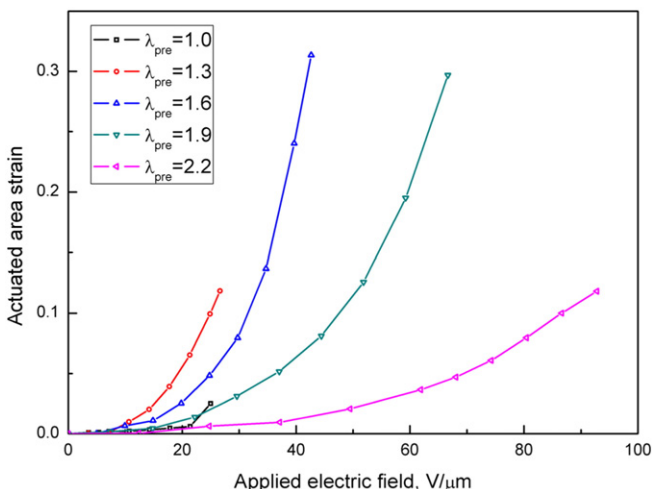#### 4.2.1. The behaviour of fabricated silicone rubber DEs with added BaTiO$_3$ particles

Fig. 4 shows samples of fabricated DEs with increased deformations depending not only on the applied electric field but also on the pre-stretch ratio (λ_{pre}). In the absence of a mechanical pre-stretch, small strains of no more than 0.3% were obtained; while pre-stretching the samples to a ratio of 1.6 achieved an increase in area of approximately 32%. As can be observed from the plots, the voltage-induced area strain for each samples used in the experiment increased with the increase in electric field. However, the electric stimulus was negligible, below 10 V/μm and this was possibly because the input electrical energy was not enough to drive the molecular chains.

Fig. 5 shows changes in the area strain achieved by varying the pre-stretch ratio. For the fabricated DEs, the actuated area strain increased steadily for pre-stretch ratios up to $\lambda_{pre} = 1.6$. After this threshold value, the area strain went down to 10% at a pre-stretch ratio of 2.2. Thereafter, the samples fractured at higher pre-stretch ratios. Pre-stretching can induce the molecular chains to rearrange to have approximate linearity and thus promote larger strains, but there is a material dependent limit on the pre-stretch that induces maximum strain.

Pre-stretch has a beneficial effect on the enhancement of Dielectric strength [29,30]. Dielectric strength increased with increasing pre-stretch ratio, mainly due to a decrease in thickness. As can be seen from Fig. 6, dielectric strength changed little under an equi-biaxial pre-stretch ratio of 1.4, whereas the dielectric strength increased linearly up to 90 V/μm beyond this ratio. As is well-known, polymer molecular chains are rearranged by the application of mechanical stress. Moreover, the mechanical stretch can generate free volume by driving the segmental motions of chains, which is propitious to increase the dielectric strength [31]. Furthermore, as a result of the thickness of the material being reduced when strained, the heat in DEs produced by the applied electric field can readily dissipate, which can also enhance dielectric strength [32].

The maximum pressure caused by the electrostatic force was calculated according to the dielectric strength and the relative permittivity of the composites. The maximum pressure increased to nearly 0.4 MPa which was 10 times larger than the pressure produced without pre-stretch (refer to Fig. 7). Notably, pre-stretch can greatly influence the pressure produced in DE materials.

The elastic energy density (e_e) is the amount of work generated in one actuation cycle per unit volume of the actuator. It is minimally affected by the compliant electrodes, power supplies, counter electrodes
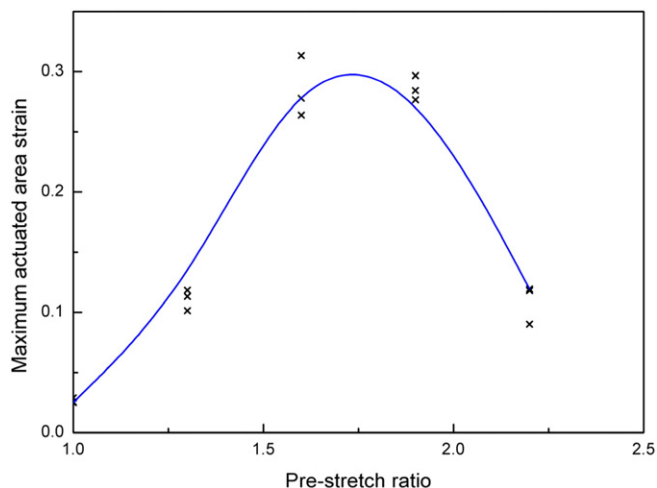

Fig. 4. Strain in area versus electric field applied to SR containing 10% BaTiO$_3$.

Fig. 5. Actuated area strain of SR with 10% BaTiO$_3$ at break related to equi-biaxial pre-stretch ratio.
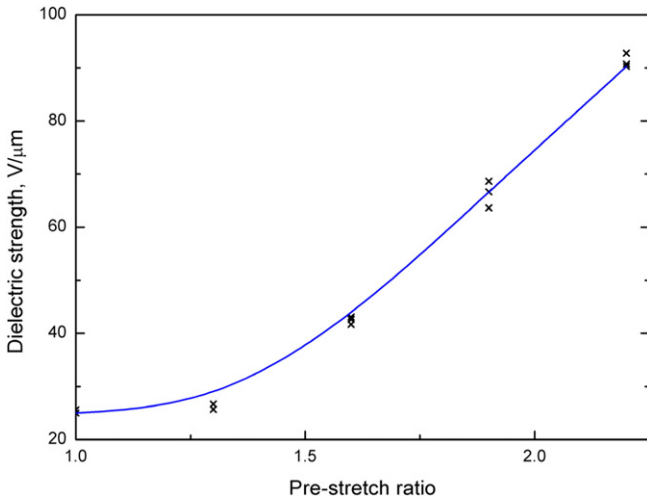
Fig. 6. Dielectric strength of SR with 10% BaTiO$_3$ at break, related to equi-biaxial pre-stretch ratio.
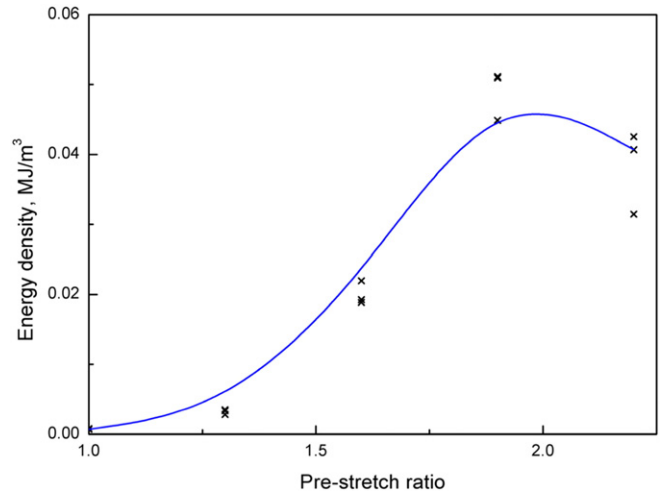

Fig. 8. Energy density of SR with 10% BaTiO$_3$ related to equi-biaxial pre-stretch ratio.

and packaging and can be calculated directly from experimentally measured quantities including the electrostatic stress and thickness strain. The equations representing these relations are as follows [33]:

$$\text{If } s_z < 10\%, \quad e_e = Y s_z^2/2 \tag{3}$$

where Y is the modulus of elasticity for the DE material,

$$\text{If } s_z \geq 10\%, \quad e_e = -p \ln(1 - s_z). \tag{4}$$

The elastic energy density increased with increasing pre-stretch ratio. The maximum energy density achieved was 0.05 MJ/m^3 (Fig. 8). However, the energy density appeared to decline for pre-stretching beyond ratios of about 2.

As well as the elastic energy density, another important parameter for characterizing a DE's performance is the efficiency (K^2), which is the electrical energy, converted into mechanical work per cycle relative to the electrical energy applied per cycle. This can alternatively be thought of as the ratio of stored mechanical energy to input electrical energy. Presuming that the elastomers are virtually isochoric materials,

the change in electrical energy upon actuation is approximately equal to the work output; in which case K^2 can be calculated using Eq. (5) [33]:

$$K^2 = 2s_z - s_z^2. \tag{5}$$

The electromechanical coupling efficiency exhibited a parabolic distribution for pre-stretched samples. As can be observed in Fig. 9, the peak of the curve (0.42) corresponded to a ratio somewhere between 1.6 and 1.9. The efficiency was directly dependent on the thickness strain induced by the electric field. K^2 increased monotonically up to about 0.42 which corresponded to a deformation of 32% as can be seen from Fig. 5. It can be assumed that the maximum strain would be experienced at a ratio between 1.6 and 1.9.

4.2.2. The influence of pre-stretch on electromechanical properties of VHB 4910

Fig. 10 shows that VHB 4910 samples obtained a maximum actuated strain at breakdown of approximately 75% for an equi-biaxial pre-stretch ratio between $\lambda_{pre} = 2.6$ and $\lambda_{pre} = 3.0$. However, the change in shape of the samples was minimal in the absence of pre-stretch. At other values for pre-stretched samples, slight changes occurred when
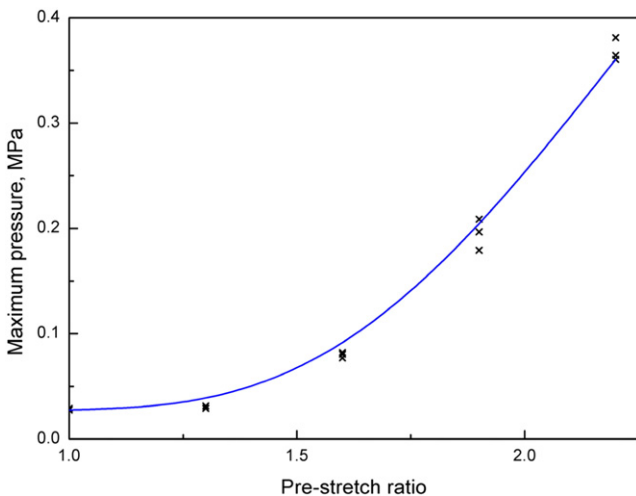

Fig. 7. Maximum pressure of SR with 10% BaTiO$_3$ related to equi-biaxial pre-stretch ratio.
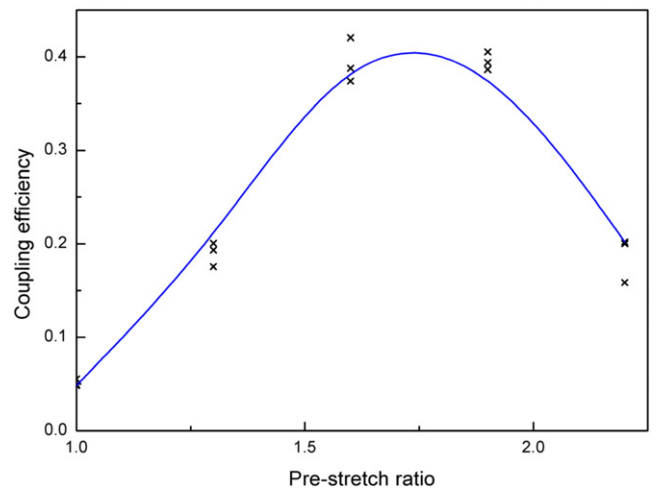

Fig. 9. Electromechanical coupling efficiency of SR with 10% BaTiO$_3$ related to equi-biaxial pre-stretch ratio.
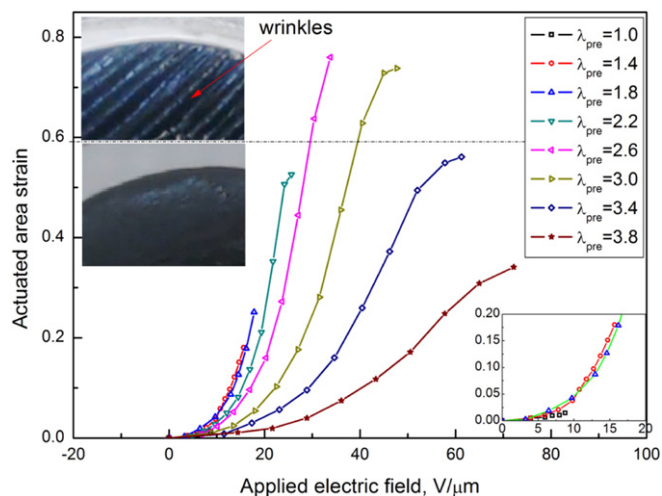
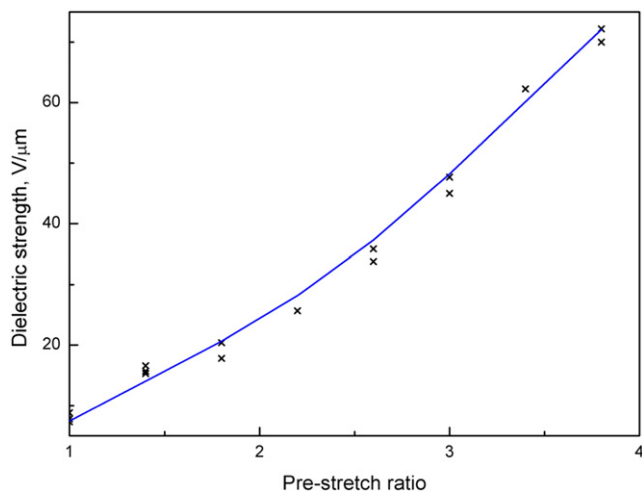**Fig. 10.** Area strain versus electric field strength for VHB 4910.

Fig. 12. Dielectric strength of VHB 4910 versus pre-stretch ratio.

applying electric fields under approximately 8 V/μm, while a steep increase occurred when the electric field was raised above this threshold value. However, as the transverse strain reached 37%, corresponding to an area strain of 59%, the films became wrinkled, indicating the onset of the pull-in effect in samples. As can be seen from Fig. 10, for the samples with pre-stretch ratios of 2.2, 2.8, 3.0 and 3.4, a plateau caused by the pull-in effect was reached before the samples punctured, while below the threshold, wrinkles were not found. The pull-in instability could be eliminated under these pre-stretch ratios for DEs if it was not essential to constrain DE test samples at their perimeters [24,34]. However, it should be considered that the maximum deformation of the samples was constrained by the frame employed in the test and this contributed to the pull-in instability in these experiments.

Pre-stretch plays an important part in electro-active deformation when applying high electric fields to DEs. Using this mechanical approach, the maximum actuated area strains at break were monotonically increased from almost zero without pre-stretch to 75% at stretch ratios of about 3. The area strain diminished to 34% at a pre-stretch ratio of 3.8 (Fig. 11). Considering that the molecular chains were completely extended when the DE samples were stretched nearly to tear, there was virtually no additional elongation stimulated by the application of an electric field.

Fig. 12 shows that dielectric strength can be enhanced by the equi-biaxial stretching of DE samples. The dielectric strength was

above 70 V/μm for VHB 4910 at the largest pre-stretch ratio of 3.8. This was because, firstly the pre-stretch reduced the thickness of the DE samples and secondly, pre-stretch promoted molecular alignment. However, the stretched samples suffered from relaxation in the elastomer and mechanical failure during the test process. This resulted in slight variations in electromechanical properties for all samples at different constant pre-stretch ratios.

An electrostatic pressure is created from the high electric field that is generated between the top and bottom surfaces of a DE material. Fig. 13 shows the relation between the electrostatic pressure and the equi-biaxial pre-stretch ratio. Stress increased with increasing pre-stretch ratio and reached a maximum value above 0.16 at a pre-stretch ratio of 3.8. However, stress increased only slightly, by approximately 0.02 MPa, up to a ratio of 2.2, before increasing markedly to a peak after this value.

The energy density e_e increased dramatically from 0 to a maximum value of above 0.05 MJ/m^3 (Fig. 14). e_e mainly depends on the electrostatic force and the strain in the thickness direction. Below a stretch ratio of around 3.4, the electrostatic force contributed more to the energy density output than to the change in strain. Thereafter, the strain decreased as a result of the energy density reducing.

The relation between electromechanical coupling efficiency and equi-biaxial pre-stretch ratio was observed to be in the form of a parabolic curve (Fig. 15). As can be seen from the figure, a maximum value
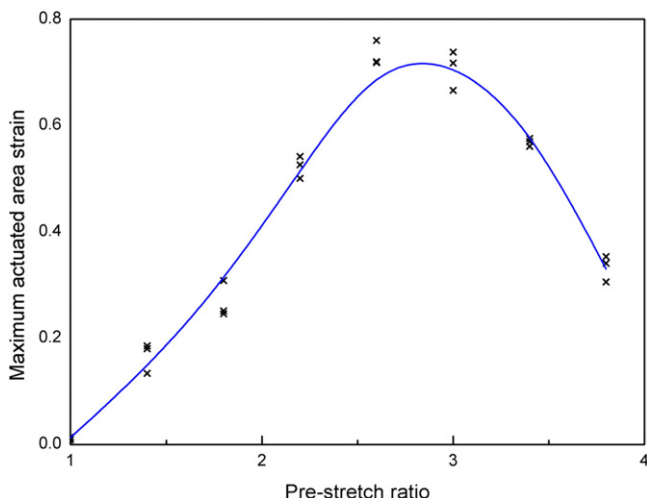

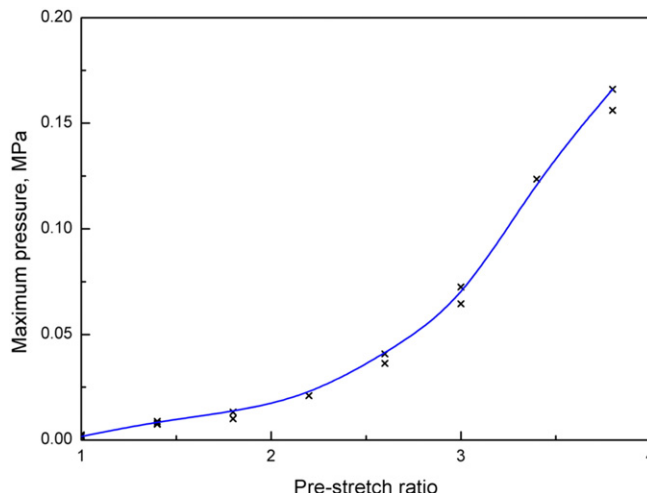**Fig. 11.** The actuated area strain of VHB 4910 at break related to pre-stretch ratio.

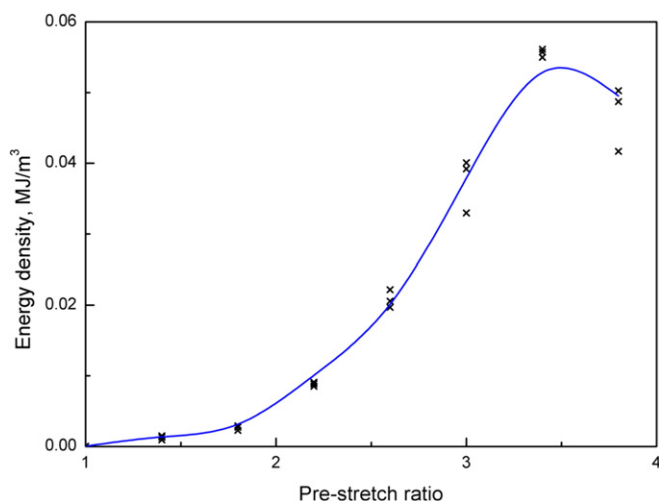**Fig. 13.** Maximum pressure of VHB 4910 versus pre-stretch ratio.

Fig. 14. Energy density of VHB 4910 related to pre-stretch ratio.

of about 70% was achieved at a peak pre-stretch ratio between 2.6 and 3. Also, K^2 increased with increased actuated area strain at fracture in the range of 0 to 75%. Hence, the form of the trend-line for K^2 related to λ_{pre} is similar to that for s_z related to λ_{pre}.

5. Conclusions

Pre-stretch greatly influences the electromechanical properties of DEs including those for DEs based on silicone and polyacrylate.

For silicone rubber based DEs with the inclusion of $BaTiO_3$ particles, a maximum actuated area strain of 32% and maximum coupling efficiency of 42% were obtained for an equi-biaxial pre-stretch ratio of 1.6. The highest dielectric strength of 90 V/μm and maximum pressure of about 0.4 MPa were each achieved for a DE with an equi-biaxial pre-stretch ratio of 2.2. The DE with an equi-biaxial pre-stretch ratio of 1.9 exhibited a maximum energy density of 0.05 MJ/m^3.

For VHB 4910 samples, both the maximum strain in area of 75% and the maximum electromechanical coupling efficiency of 70% occurred at an equi-biaxial pre-stretch ratio of 3.0. The highest dielectric strength of 70 V/μm and the maximum pressure of 0.16 MPa were obtained by applying a driving voltage to the VHB 4910 at an equi-biaxial pre-stretch ratio of 3.8. A maximum energy density of 0.05 MJ/m^3 was achieved for VHB 4910 at an equi-biaxial pre-stretch ratio of 3.4.
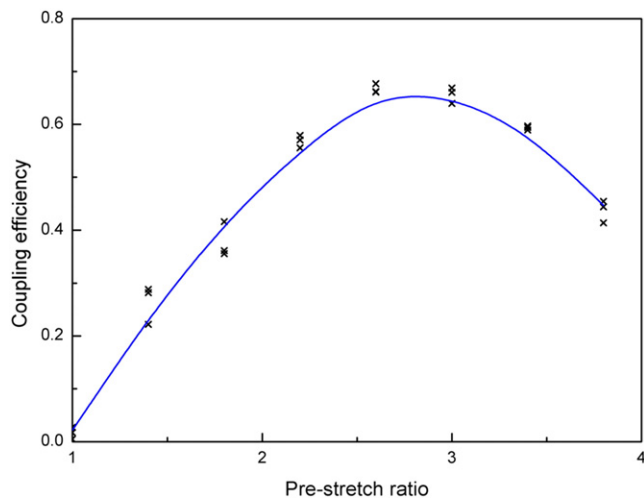

Fig. 15. Electromechanical coupling efficiency of VHB 4910 related to pre-stretch ratio.

Comparing the fabricated silicone DE with VHB 4910 showed that, though the maximum strain achieved for the silicone was slightly smaller, the maximum energy density output of each was roughly equal. Also, the maximum pressure and the maximum dielectric strength were much higher for the fabricated silicone DE samples containing $BaTiO_3$ particles than for the commercially available VHB 4910 samples and importantly they did not exhibit a pronounced pull-in effect. However, both silicone and polyacrylate are considered promising materials for DE applications.

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