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 (BaTiO₃) 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% BaTiO₃ 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% BaTiO₃ 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% BaTiO₃ ($\lambda_{pre} = 1.6$) 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
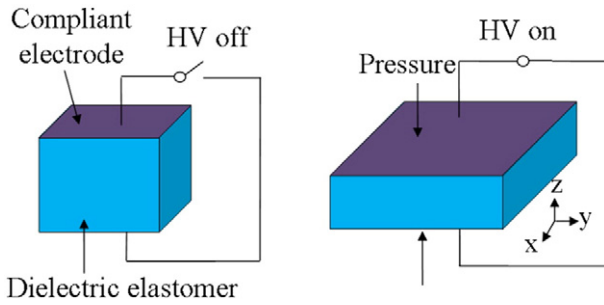
Fig. 1. Schematic representation of a DE's working principle.

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)).

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

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
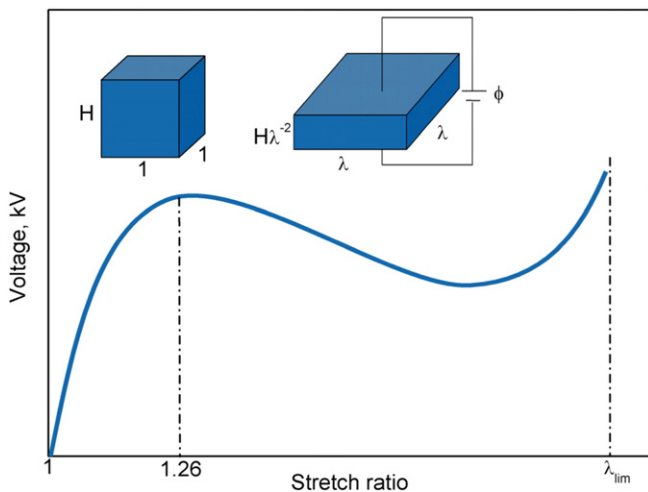

Fig. 2. A membrane of a dielectric elastomer subjected to a voltage reduces in thickness and expands in surface area. The voltage–stretch curve is typically not monotonic.

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