Fault-tolerant silicone dielectric elastomers

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Soft silicone films have garnered a great deal of interest for use in dielectric elastomer transducers due to their excellent properties, including high elongation to rupture, low viscoelasticity, and broad application temperature range. However, silicone films generally have higher stiffness and lower dielectric strength than VHB acrylic elastomers, which limits the achievable actuation strain. Devices based on silicone dielectric elastomers always experience high rates of premature dielectric failure when operated at high strains. The premature failure is characterized by the loss of functionality or mechanical rupture of the material when operated below the material’s dielectric strength and elongation to rupture. The use is reported of ultrathin coatings of single-walled carbon nanotubes (SWNTs) as the compliant electrodes, which can overcome the issue of premature failure. The self-clearing of the SWNT electrodes in the event of localized dielectric breakdown improves the apparent dielectric strength of the material by isolating the regions of reduced dielectric strength. The actuators may be operated at higher than 50% area strain with reasonably long lifetimes. High strains were measured between −40 and 80°C and in a broad frequency range up to 100 Hz. The fault tolerance introduced by the SWNT electrodes should broaden the application scope of silicone dielectric elastomers.

Keywords: silicone elastomer; fault tolerance; single-walled carbon nanotube; dielectric elastomer

1. Introduction

Dielectric elastomers are a promising transducer technology capable of matching the performance of human muscles in terms of strain, stress, work density, and mechanical compliance [1–3]. The basic structure of a dielectric elastomer consists of an elastomer film sandwiched between two overlapping compliant electrodes. The electrodes conform to the surface of the elastomer film, and stretch/contract with the elastomer in response to changes in the applied electric field [4–6]. In the late 1990s and early 2000s, a large number of elastomer materials were tested, including silicones [7–9], polyurethanes [10], thermoplastics [11], and acrylics [2–4,8]. Of these materials, 3M’s VHB4910 family of acrylic elastomers, VHB-based interpenetrating polymer networks (IPN) [12,13], and soft silicones appear to be the most promising in terms of overall actuation performance. Area strains in excess of 300% have been reported with highly prestrained VHB4910 films and IPN films. The maximum calculated energy density is >3 MJ/m³. The acrylic and IPN films display reduced performance at moderate to high frequencies and at temperatures below 0°C.

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Using plasticizers can alleviate these problems to a certain degree but may reduce the mechanical robustness of the materials [17].

Soft silicone films are among the best elastomers in terms of low viscoelasticity and broad application temperature range (−80 to 300°C) [18–20]. However, silicone films have higher stiffness and lower dielectric strength compared to acrylic and IPN films. The reported area strains are usually lower than 50% [7,21,22]. Higher strains are possible, but result in extremely poor reliability and lifetime. Devices based on silicone dielectric elastomers always display a high rate of dielectric failure when operated at high strains [23].

We have shown previously that by employing ultrathin coatings of single-walled carbon nanotubes (SWNTs) as the compliant electrodes, actuators based on acrylic films can be made fault tolerant [24–26]. The SWNT coatings around a dielectric breakdown site can self-clear to electrically isolate the defect. The remaining active area retains its capacity for high-strain actuation. It is of interest to investigate whether similar fault tolerance can be achieved in silicone films, which are more prone to unexpected dielectric breakdown than the acrylic films. We report the successful demonstration of fault tolerance in silicone films and the improvement of the polymer’s apparent dielectric strength. The resulting silicone dielectric elastomers may be actuated reliably at >50% area strain.

2. Experimental

2.1. Materials

2.1.1. Silicone films

A two-component thermally cured silicone (LC-20-2004, Dow Corning) was used to fabricate the soft silicone films used in our study. The two components were dissolved separately in toluene at a 50% weight concentration. The resulting solutions were mixed in equal volume. A doctor blade coater as used to cast films out of the resulting viscous mixture. The films were allowed to dry in a fume hood overnight and were subsequently thermally cured in a convection oven at 80°C for 24 h. The resulting freestanding films were peeled from the substrate and prestrained to various extents for use as the dielectric film in the construction of the circular expansion actuators described below.

2.1.2. SWNT electrodes on silicone

Carbon nanotubes (Sun Innovations, Inc.) were dispersed in chloroform with a concentration of 0.2 mg/ml using a bath sonicator until a uniform dark dispersion was obtained. The dispersion was sprayed onto the cured prestrained silicone films using a contact mask to define an overlapping circular electrode area across the silicone film. The resulting actuators are called circular expansion actuators due to the circular shape of the active area. The electrode coating procedure has been described in detail elsewhere [24,25].

2.2. Measurement methods

2.2.1. Actuation test

The circular expansion actuators were subjected to a step-increase actuation test at voltages starting at 1 kV with 0.5 kV increments until the actuators failed. The actuation test set-up is described in [25,26]. Briefly, an actuator was connected in series with a Keithley 2000 multimeter, a high-voltage power supply, and a 20 MΩ current-limiting resistor. The
Keithley 2000 multimeter was used to measure the charging and leakage currents during actuation, which are typically in the range of a few microamperes (μA).

The circular active area of the actuators was monitored using a digital camera. The area strain was calculated using the equation: \((a_1 - a_0)/a_0 \times 100\%\), where \(a_0\) is the active electrode area before actuation and \(a_1\) is the active area during actuation.

2.2.2. High-frequency actuation

The actuation strain of dielectric elastomers usually decays with increasing actuation frequency due to the viscoelastic nature of the elastomer films. An optical apparatus, shown in Figure 1, was designed to characterize the strain degradation of silicone elastomers. The collimated light beam emitted from a laser passes through a sample and is focused by a convex lens onto a photodiode. The electrodes coated on the sample are made from carbon grease that blocks the light beam. The area change of the electrode (active area) during actuation causes the transmitted light intensity to change. The transmitted light intensity decreases linearly with the increase in electrode area. The ratio of the strain at frequency \(f\) to that at a low frequency (1 Hz) can be calculated by

\[
DS_f = \frac{a_{h_f} - a_{l_f}}{a_{1_f} - a_{1_l}} = \frac{I_{h_f} - I_{l_f}}{I_{1_f} - I_{1_l}},
\]

where \(DS_f\) is defined as the decay of actuation strain at frequency \(f\) compared with the strain at 1 Hz; \(a_{h_f}\) is the largest active area during expansion at frequency \(f\); \(a_{l_f}\) is the smallest active area during contraction at \(f\); \(a_{1_f}\) and \(a_{1_l}\) are, respectively, the largest and smallest active areas during actuation at 1 Hz. \(I_{h_f}\) and \(I_{l_f}\) are the measured highest and lowest transmitted light intensity, respectively, at frequency \(f\), and \(I_{1_f}\) and \(I_{1_l}\) are the lowest and highest transmitted light intensity, respectively, at 1 Hz. The strain at 1 Hz can be measured with a camcorder. Thus, based on the strain decay curve, the strain at frequency \(f\) is

\[
S_f = S_1 DS_f,
\]

where \(S_1\) is the measured strain at 1 Hz.

Figure 1. Set-up for strain decay test: (1) laser light source; (2) dielectric elastomer actuator sample; (3) convex lens; (4) photodiode; (5) signal process box.
Samples were tested with a square wave voltage signal at a 50% duty cycle, at 1, 5, 10, 20, 30, 40, 50, 60, 70, 80, 90, and 100 Hz. The peak voltage (3 kV) was supplied using a 30 W high voltage converter. A 1 MΩ drain resistor was used to discharge the actuators when the voltage was turned off. The circular active area was 6.35 mm in diameter and 44.4 μm in thickness in the rest state, and the dielectric constant of silicone is \( \varepsilon = 3 \). The cut-off frequency of this test system is \( f_c = \frac{1}{2\pi RC} = 8.43 \times 10^3 \) Hz, which is high enough to characterize the frequency response of our silicone test devices up to 100 Hz.

2.2.3. Thermal testing

The influence of temperature on the actuation performance of silicone-based dielectric elastomers was characterized using an environmental chamber (Thermotron). Circular expansion actuators with SWNT electrodes were tested at -40, 25, and 80°C. At each temperature, a sample was put in the chamber for 30 min before applying any voltage to ensure an even temperature distribution across the film. The actuation strains were measured from images taken by a camera.

3. Results

3.1. Dielectric strength improvement with carbon nanotube electrodes

In this study, 100 μm thick silicone films were biaxially prestrained by 25%, 50%, 75% and 100%. The corresponding thickness of prestrained films is 64, 44, 33, and 25 μm, respectively. Circular expansion actuators with SWNT electrodes and carbon grease electrodes were tested.

The maximum strains obtained at different levels of prestrain and for different electrode materials are shown in Figure 2. With SWNT electrodes, the maximum strains achieved were around 50%, 65%, 85%, and 50% in area with corresponding biaxial prestrains of 25%, 50%, 75%, and 100%, respectively. For carbon grease electrodes, the maximum strains were

![Figure 2. Maximum area strain distribution with prestrain for silicone-based actuators with SWNT or carbon grease electrodes. The listed prestrains indicate the percentage strain along both planar directions (i.e. 25 × 25%, 50 × 50%, etc).](attachment:figure2.png)
around 40%, 65%, 40% and 10% for films with prestrains of 25%, 50%, 75% and 100%. For prestrains up to 50%, the actuation strain for both electrode materials is comparable. For prestrains above 50% there is a marked difference between the maximum achievable strains using SWNT and carbon grease electrodes, with the SWNT electrodes outperforming the carbon grease by a large margin.

The higher strain obtained using SWNT electrodes is attributed to the self-clearable properties of the SWNT electrodes, which prevents the premature failure of the actuator [25]. Figure 3 shows the actuation current as a function of time during actuation testing for actuators with 75% prestrained films coated with SWNT or carbon grease electrodes. For the actuator with SWNT electrodes (Figure 3a), the actuation was normal for applied voltages ranging from 1 kV to 3 kV. At 4 kV, a spike can be observed in the current curve. This is indicative of a successful clearing event, wherein the actuator suffered a localized dielectric

Figure 3. Chronoamperic curves for actuators during step-increase actuation testing: (a) 75% prestrained silicone with SWNT electrodes; (b) 75% prestrained silicone with carbon grease electrodes.
breakdown but avoided terminal failure and resumed its electromechanical function. At 4.5 kV, a number of spikes can be observed; the intervals between two spikes varied from 1 second to several seconds. The occurrence of multiple localized dielectric breakdown events indicates that the electrical field introduced across the film at 4.5 kV has approached the dielectric strength of the silicone film.

For the actuator with carbon grease electrodes (Figure 3b), actuation was normal at 1 and 2 kV. At 3 kV, the current increased to a high value and remained there, which indicates the actuator underwent premature failure and permanently lost its electromechanical function.

Since the self-clearable SWNT electrodes can prevent premature failure during actuation testing, we can determine the maximum breakdown strength of the dielectric elastomer from the field at which we begin to see continuous breakdown-clearing events. As shown in Figure 4, the maximum breakdown strength is 125 V/μm at 25% prestrain, 210 V/μm at 50% prestrain, 250 V/μm at 75% prestrain, and 265 V/μm at 100% prestrain. The breakdown strength increased with increasing prestrain, a trend that matches the results for 3M VHB elastomers [27]. Due to the inability of carbon grease electrodes to undergo self-clearing, the calculated breakdown strengths are randomly distributed with prestrain.

The results indicate that the dielectric strength of silicone film increases with the prestrain; however, the stiffness of the film has also been shown to increase [19]. Using a simple linear approximation for the strain response of the material, which is valid at small strains and provides a rough estimate at larger strains, \( s = \varepsilon_0 \varepsilon \frac{E}{Y} \), where \( s \) is strain, \( E \) is electric field, \( Y \) is Young’s modulus, and \( \varepsilon \) and \( \varepsilon_0 \) are the dielectric constant of the elastomer and the permittivity of free space, respectively. We can see that the strain is affected by the dielectric strength and modulus; however, both are affected by the prestrain ratio. For 25% biaxial prestrain, even though the film is soft, the breakdown strength is low. Thus, the strain is lower, and a maximum area strain of about 50% is obtained. For 100% biaxial prestrain, the dielectric strength is highest, but it is stiffest due to the high degree of prestrain. Thus, the strain is also low, about 50% in area. For 50% and 75% biaxial prestrain, the dielectric strength of the films is around 210 and 250 V/μm, respectively, and the stiffness is moderate,

![Figure 4. Dielectric strength distribution for various prestrain ratios and electrode materials during step-increase actuation testing.](image)
thus, the maximum attainable strain is higher than for the other two prestrain levels. The highest actuation strain was obtained with a film prestrained biaxially by 75%.

3.2. Improvement in operational durability with carbon nanotube electrodes

The SWNT self-clearing process can prevent premature failure in dielectric elastomer actuators and thus prolong the operational lifetime. A circular actuator with an active area 20 mm in a diameter was actuated at a constant voltage of 4 kV to test the constant actuation lifetime, as shown in Figure 5. The actuator tested was fabricated using a silicone film with an initial thickness of 130 μm that was prestrained biaxially by 50%. At 4 kV, the actuated strain was about 50% in area. During the continuous test, which lasted for 260 minutes, the actuator survived six localized dielectric breakdown events that occurred at 35, 50, 120, 155, 200, and 210 minutes, as indicated by the spikes in current. The strain measured during the test remained in the range of 45% to 50%. The actuated strain remained steady at 50% until the fourth clearing event, after which the strain decreased to 45% due to the formation of inactive areas on the electrode surface after clearing.

3.3. Broad actuation temperature range of silicone

The glass transition temperature of silicone is around −120°C. Thus, actuators based on silicone films can be operated efficiently between −40 and 80°C, the typical range of operation temperatures required for consumer products. From Figure 6, we can see that at each temperature the strain increases with voltage as expected, and that at each voltage the actuation strain is higher at lower temperature; this trend is more pronounced at higher voltage.

This variation of actuation strain with temperature is likely related to the dependence of an elastomer’s modulus with temperature [28]. Since the elasticity of an elastomer is driven by entropy, the modulus will decrease with decreasing temperature. Therefore, the electrically induced strain will be higher at lower temperatures.

3.4. Linear strain with non-uniform prestraining

When the silicone sheet was subjected to non-uniform prestraining, namely, high prestrain along one direction and low prestrain along the orthogonal direction, it showed preferential

![Figure 5. Strain and chronoamperic curves for a lifetime test at a constant voltage of 4 kV.](image)
actuation strain along the low prestrained direction over the high prestrained direction. This phenomenon has also been observed with 3M VHB elastomers [8].

The silicone sheets were prestrained by 100% x 25%, 125% x 10%, and 125% x 50%. SWNT electrodes with a rectangular shape measuring 4.5 mm along the high prestrained direction and 1 mm along the low prestrain direction were applied using an air brush. The strain was measured along the direction with low prestrain.

The change in linear actuation strain along the low prestrain direction with electrical field is shown in Figure 7. It can be seen that under the same electrical field, the amount of electrically induced strain is higher for lower prestrain along that direction. For the three levels of prestrain tested, the maximum strain obtained was about 55% with the 100% x 25% sample. The higher strain observed in samples with this prestrain is attributed to the

Figure 6. Strain measurement at −40, 25 and 80°C with voltage increasing test.

Figure 7. Linear actuation strain along low prestrained direction under an applied electrical field for silicone actuators with non-uniform prestrain.
The more optimal combination of dielectric strength and stiffness after prestraining. As mentioned in Section 3.1, even though the dielectric strength is higher for the highly prestrained samples (125% × 50%), the higher stiffness will act to counter any increase in the actuation strain brought about by an increase in dielectric strength.

3.5. Strain frequency dependence

Figure 8 shows the strain decay of a silicone actuator with frequency. At 100 Hz, the strain is about 70% of the strain at 1 Hz. Thus, the 50% cut-off frequency for silicone is over 100 Hz.

For a 75% biaxially prestrained 100 μm thick LC-20 silicone actuator with SWNT electrodes, an area strain of approximately 80% can be achieved at 1 Hz. Thus, from the decay curve, we can estimate that over 50% area strain is obtained at 100 Hz.

4. Discussion

4.1. Thickness dependence of SWNT clearing on silicone

The clearing of SWNT electrodes on silicone is strongly dependent on the thickness of the silicone film. This was observed during self-clearing testing of SWNT electrodes on silicone films of different thickness. When tested on a 30 μm thick silicone film, the self-clearing process remained incomplete for 10 min, at which point the test was terminated. However, on a 120 μm thick silicone film, the clearing took a matter of seconds to complete. The effect of film thickness on the ability of silicone films to self-clear needs further investigation in order to find the optimum thickness for silicone dielectric elastomer actuators.

Three silicone sheets with thicknesses of 50, 110, and 160 μm were fabricated using a doctor blade coater. The resulting free standing films were biaxially prestrained by 25%, and circular actuators with an active region 20 mm in a diameter were prepared by spraying SWNT electrodes onto the films. The surface resistance of all SWNT electrodes was around 800 Ω/square. For the electrode self-clearing tests, defects were introduced into the active area of the actuators by puncturing through the silicone film with a pin; following this a voltage of 2 kV was applied to the actuator. A Keithley 2000 multimeter was used to record the current across the film during the clearing process and the results are shown in Figure 9.
Figure 9. Chronoamperic curves of SWNT clearing on (a) 50 μm, (b) 110 μm and (c) 160 μm thick silicone films, all biaxially prestrained by 25% × 25%. Each current spike corresponds to a localized dielectric breakdown caused by a pin puncture.
For SWNT electrodes on the 50 μm thick silicone films, the clearing period ranged from 2 to 6 min. When tested on the 110 μm silicone films, it decreased to 1 or 2 min. When tested on the 160 μm thick silicone films, it dropped further to tens of seconds. To confirm the repeatability of the clearing process, four or more defects were introduced into each sample and then cleared. From Figure 9, it can be seen that the self-clearing of the SWNT electrodes is very reproducible. The clearing duration varies with the thickness of the silicone films. For a specific thickness, the clearing durations are quite comparable from one puncture to the next.

The dependence of the self-clearing process on film thickness may be due to the possibility of the carbon nanotubes from the two opposing electrodes approaching each other during breakdown or puncturing. Typically, the length of SWNTs is between 5 and 15 μm; during breakdown or puncturing, the tubes around the hole will move in and form a conductive pathway between the two electrodes through the hole. This possibility increases with decreasing film thickness. The SWNT clearing duration relies on the high voltage discharge intensity between the electrodes. The higher is the intensity, the faster the clearing. However, the high voltage discharge intensity is determined by the conductance through the punctured hole [29]. If it is too conductive, as is the case with carbon grease electrode, an electrical short will be formed and maintained and the high voltage discharging will not take place. At sufficiently low conductance levels, high voltage discharging will become possible and the intensity of the discharging process will increase with decreasing conductance. Under high discharge intensity, the duration of SWNT clearing is short. On the other hand, the thinner the dielectric film, the higher the possibility of the opposing electrodes forming a conductive pathway which will decrease the discharge intensity and increase the clearing duration.

Why has this phenomenon not been observed during the SWNT clearing process on prestrained 3M VHB films [25]? The original thickness is 1 mm for 4910 and 0.5 mm for 4905 films. After the high prestrain, the VHB films tested were thinner than the thickest silicone film in Figure 9. When a small hole is punctured through the film, the VHB film around the hole relaxes because the high level of prestrain cannot be supported, and the film increases in thickness. Forming a conductive path through the edge of the hole is less likely. Thus, the intensity of high voltage discharging will be high and the SWNT clearing process on prestrained VHB films only lasts for about 3 to 10 seconds.

5. Conclusion

We have demonstrated the self-clearing properties of SWNT electrodes on silicone films. With SWNT electrodes, premature failure of silicone dielectric actuators can be avoided and the apparent breakdown strength of the material can be improved. For a 75% biaxially prestrained LC-20-2004 silicone film, an area strain of over 80% and a dielectric strength of 250 V/μm have been achieved. The duration of the SWNT clearing process depends on the thickness of the silicone film. The thicker the silicone film, the faster the clearing of SWNTs surrounding the breakdown site. A silicone film thickness of around 100 μm allows for an acceptable actuation voltage and clearing duration. Actuators based on silicone films can operate between −40 and 80°C. At the same actuation voltage, the strain is larger at lower temperature. This trend follows the expected behavior or an entropically elastic material. Linear actuation with a non-uniformly prestrained silicone sheet was observed and a linear strain of about 55% has been achieved with a sample prestrained to 100% × 25%. Using an optical apparatus, the 50% cutting-off frequency of silicone was found to be higher than 100 Hz. With fault tolerant silicone actuators, an area strain of around 50% can be obtained at 100 Hz.
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