

Highly Stretchable Silicone Elastomer Applied in Soft Actuators

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In this work, a highly stretchable silicone elastomer is incorporated into dielectric elastomer actuators (DEAs) in order to decrease operation voltages by applying high prestretches. Results show that the fabricated DEAs (5 mm diameter circle active region) can be actuated to a lateral strain of 30% at 4.3 kV for a 122 µm thick prestretched film, and to a lateral strain of 2.5% at only 250 V for a 6.9 µm thick prestretched film. Due to the significant viscous component of the silicone elastomer, the DEAs respond more slowly (2-14 s to reach 90% of full strain) and show greater strain changes over time compared to conventional silicone-based DEAs. While this inherent viscosity is not universally favorable, it can be advantageous in applications where actuator damping is desirable. The studied DEAs' mean lifetimes under DC actuation range significantly-from 0.9 h to more than 123.0 h-depending mainly on initial electrical fields (17.8–36.3 V μ m⁻¹). For instance, DEAs with a 150 µm initial thickness and a prestretch ratio of 3 show 1.4-2.6% lateral strains for the mean lifetime (123.0 h) at only 300 V. Given the strains achieved at low voltage, such DEAs show promise for applications that do not require fast response speeds.

1. Introduction

A dielectric elastomer actuator (DEA) is an electromechanical transducer that consists of a dielectric elastomer (DE) sand-wiched between two compliant electrodes.^[1] When a voltage is applied, electrostatic pressure causes the DE to decrease in thickness and expand in area.^[2] DEAs enable noiseless, reversible deformations via the cyclic charging and discharging of electrodes. DEA development has progressed rapidly due to

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promising applications in numerous fields such as soft robotics,^[3,4] tunable optics,^[5,6] and compliant grippers.^[7,8] However, one of the main obstacles to more widespread DEA use is that high voltages (>1 kV) are usually required to achieve desirable strains.^[2] Such high operation voltages require stringent safety regulations, restricting DEAs' use in certain practical applications such as wearables. Additionally, high operation voltages tend to induce lower reliability of the DEAs as dielectric breakdown becomes more likely to occur.

Silicone elastomers are the preferred material for DEAs due to their high efficiency, reliability, and broad operation temperature range.^[9,10] Silicone elastomers with a wide range of properties are commercially available, and extensive research has been conducted to map their performance.^[11,12] Within the vast range of available formulations, however, elastic moduli values remain in the relatively narrow range of

0.05–2.4 MPa. Further elastomer functionalization enables improved DEA performance, and numerous studies have therefore attempted to decrease operation voltages by decreasing either the thickness or Young's modulus and/or increasing the dielectric permittivity of the silicone elastomers used in their formulation.^[2]

Silicone elastomers' dielectric permittivity has been increased by physically or chemically incorporating high permittivity components such as particles, oils, and functional moieties.^[2,13] While the physical incorporation of high permittivity components is easy to achieve, it often leads to increased stiffness, phase-separation, and/or long-term instability of the resulting elastomers.^[14,15] In contrast, silicone elastomers covalently grafted with high permittivity components can overcome such disadvantages.^[16,17] However, these materials often suffer from increased dielectric loss, which in turn commonly leads to reduced dielectric breakdown strength.^[18]

Multiple approaches for developing softer silicone elastomers have been proposed as alternative paths to improving actuation strains: soft silicone elastomers were prepared by adding solvents,^[19–22] through crosslinking with offstoichiometric ratios,^[23–25] or by crosslinking with bottle-brush polydimethylsiloxane (PDMS).^[26,27] While the first two strategies are relatively easy to implement, the resulting elastomers are prone to electromechanical instability due to potential phase separations.^[19] Crosslinking with bottle-brush PDMS, on the





Figure 1. Illustration of two silicone elastomers resulting from curing reactions: a conventional silicone elastomer resulting from a hydrosilylation reaction; a highly stretchable silicone elastomer resulting from chain extension and crosslinking in a one-pot reaction where the chain extension reaction proceeds considerably before any significant crosslinking occurs.

other hand, has been shown to result in ultrasoft yet stable silicone elastomers; however, this strategy often requires relatively complex synthesis.^[28]

The fabrication of thinner DE films is desirable because actuation strains are inversely proportional to the squared film thickness. Most reported film thicknesses are in the range of 20-100 µm, as reliable fabrication becomes challenging below this range. Nevertheless, a DEA with a 3 µm thick pad-printed film has been reported, which exhibited a 7.5% lateral strain at only 245 V.^[29] A DEA with a thinner film of 1.4 µm was subsequently fabricated by solvent-casting a DE film and using Langmuir-Schaefer transferred electrodes, enabling a 4.0% lateral strain at the impressively low voltage of 100 V.^[30] Disadvantageously, fabrication of thin DE films may decrease dielectric breakdown strength due to possible nonuniformities in the films, such as air bubbles, foreign particles and thickness variations.^[30] As characteristic dimensions of the nonuniformities do not scale down with the film thickness, their relative impact increases for thin DEAs, leading to premature local dielectric breakdown. Another approach for reducing DE film thickness is through prestretching. This process circumvents the need to prepare reliable ultrathin films (<20 µm), and it is a widely known technique to improve dielectric breakdown strength^[31,32] however, some challenges remain. First, the reduction in thickness is limited by the stretchability of the elastomers used, as conventional silicone elastomers usually have maximum strains below 900% in uniaxial deformations.^[12] Second, prestretching requires that actuators possess rigid frames, which may limit their practical applications; in addition, a high degree of prestretching can often lead to significantly increased stiffness in the resulting DE films due to strain hardening effects and loss of tension over time for viscous DE films. In order to overcome these limitations for devices in which rigid prestretch frames are acceptable, a silicone elastomer with high stretchability and a low modulus under high degrees of prestretch is therefore needed.

In our previous work,^[33] highly stretchable silicone elastomers were prepared using an air-mediated reaction of telechelic hydride functional PDMS, telechelic vinyl functional PDMS, and a platinum catalyst in excess of hydride groups (**Figure 1**). Two types of reactions take place simultaneously in this one-pot reaction, in which the hydrosilylation reaction between vinyl groups and hydride groups results in chain extensions, and the reaction of hydride groups with methyl groups, oxygen, and water leads to crosslinking. Due to the significant kinetic advantage of the former reaction over the latter reaction, the length of the PDMS chains is significantly increased before any meaningful SCIENCE NEWS _____ www.advancedsciencenews.com



Figure 2. a) Uniaxial true stress-strain curves. b) Frequency dependence of storage and loss moduli. c) Dielectric permittivity (ϵ ') as a function of frequency.

crosslinking occurs, enabling significantly increased stretchability of the elastomers. In this work, a highly stretchable silicone elastomer is prepared with the same reaction strategy. The distance between chemical crosslinks is much longer (around 5 times) in the resulting elastomer compared to the conventional silicone elastomer resulting from a hydrosilylation reaction (Figure 1). In addition, the modulus of the highly stretchable silicone elastomer produced here remains low even being uniaxially stretched to 1500% strain. This material is then used to fabricate thin DEAs with equibiaxial prestretch ratios of 3 and 6, with the aim of decreasing operation voltages. Strain–voltage curves, response speeds, and long-term stability of the fabricated DEAs are studied.

2. Results and Discussion

2.1. Characterizations of Silicone Elastomers

An elastomer's ultimate strain determines its highest achievable extent of prestretch. **Figure 2**a presents uniaxial true stress versus strain curves for the highly stretchable silicone elastomer produced here compared to a conventional silicone elastomer. The former shows a maximum strain of 2300%, which is around five times as large as that of the conventional silicone elastomer (480%). Such high stretchability permits a significant reduction in thickness from large prestretches. The elastic modulus only increases from 0.19 to 0.38 MPa under 1500% strain. The combination of high stretchability and low modulus under large strain

allows implementing high prestretches on DEAs to significantly reduce thickness, with low strain hardening effects.

Elastomers' linear viscoelastic properties are shown in Figure 2b. An obvious relaxation behavior is observed for the highly stretchable silicone elastomer, as evidenced by the reduced storage modulus with decreasing frequency. This relaxation behavior suggests that the entangled chains are unraveled due to their sliding ability under the measured shearing frequencies between 0.01–100 Hz. Moreover, the highly stretchable elastomer shows a much higher viscous loss, as tan (δ), a ratio of loss modulus to storage modulus, at 0.01 Hz is 15-fold larger than that of the conventional elastomer. Both the observed relaxation behavior and high viscous loss of the highly stretchable silicone elastomer can be attributed to its highly entangled and sparsely crosslinked structures.

Figure 2c shows that the dielectric permittivity of the highly stretchable silicone elastomers remains almost constant in the frequency range of 10^{-2} – 10^{5} Hz. The dielectric permittivity of the highly stretchable silicone elastomer without prestretch is around 3.3 at the measured frequencies, which is close to that of the conventional silicone elastomer. When the elastomer is stretched to ratios of 3 and 6, the dielectric permittivity is reduced to 2.4 and 2.5, respectively. Because the highly stretchable silicone elastomer consists of the same repeating (SiO(CH₃)₂)) units as the conventional silicone elastomer, their dielectric permittivities should be similar. As the elastomer is stretched equibiaxially, the PDMS chains become oriented in the stretching directions. When applying an electrical field perpendicular to the stretching plane, the movements of dipoles on the chains are constrained

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Figure 3. a) DEA strain (s_x , lateral strain)-voltage plots. b) DEA strain-electrical field plots. Electrical field is calculated by $E = \frac{U}{d_0} (1 + s_x)^2$, where U is the applied voltage, d_0 is the prestretched thickness of a film before a voltage is applied, and E_{BD} is the dielectric breakdown strength.

by the mechanical force. It is therefore expected that the dielectric permittivity perpendicular to the stretching direction will be lower than in the stretching direction,^[34–36] and that a decrease in dielectric permittivity will be observed.

2.2. Actuation Performances of DEAs

DEAs' initial thickness and prestretch ratio affect their response to applied voltage, as they govern the final thickness of prestretched films and the prestretch ratio can also affect Young's modulus. DEAs with different initial thicknesses (1100, 280, and 150 µm) and equibiaxial prestretch ratios (3 and 6) were fabricated. Actuation strains mentioned in this work represent lateral strains (s_x) , which were determined by Equation (1). As shown in Figure 3a, DEA_1100 µm_3 can be maximally actuated to a strain of 30% at 4300 V-a much larger actuation strain than the reported maximum strains of 10-15% for conventional siliconebased DEAs.^[31,37-39] This increased strain threshold can be attributed to the low modulus even being highly stretched. By decreasing the initial thickness or increasing the prestretch, DEAs can be actuated to the same strains at much lower voltages. Specifically, DEA_280 µm_3 shows a maximum strain of 11.8% at 1200 V, DEA_280 µm_6 shows a maximum strain of 2.5% at only 250 V, while DEA_1100 µm_3 requires 3 and 9 times the voltage, respectively, to enable the same strains. In terms of their achieved strains, the studied DEAs show promise for use at low voltage, circumventing the challenge posed by the typically high voltages (>1 kV) needed to actuate silicone-based DEAs.^[2]

Lateral strain-electrical field plots are shown in Figure 3b. For DEAs with initial thicknesses of 1100 and 280 μ m and the same prestretch ratios, these plots are mostly overlaid; this is consistent with the theoretical relationship between the lateral strain and the electrical field,^[40] since DEAs with the same prestretch ratio should possess identical mechanical behaviors and dielectric permittivities. The curve of the thinnest prestretched DEAs (DEA_150 μ m_3) is far below the curves of the DEAs with the prestretch ratio of 3. This discrepancy may be due to amplificatory effects of defects (such as air bubbles and foreign particles) in the thin films, which could originate from film and electrode fabrication processes. Additionally, larger standard error on the



Figure 4. Strain response as a function of actuation time for different prestretched actuators at relevant voltages.

measured thickness of the thin films also might be a factor for the discrepancy. The DEAs undergo electrical breakdown when the electrical field is further increased from the maximum strengths tested in Figure 3b. The dielectric breakdown strength was reduced from 60.7 to 39.3 V μ m⁻¹ by decreasing the initial thickness and increasing the prestretch ratio from 3 to 6. The prestretch has been reported to improve dielectric breakdown strength.^[31,32] In our case, the contrary effect of the prestretch on the dielectric breakdown strength could be because the increase of defect propagations under such an unprecedentedly high prestretch ratio.

DEA response speed is an important factor for practical applications, and response times ranging from tens of milliseconds to a few seconds have been reported for silicone-based DEAs.^[41,42] As shown in **Figure 4**, the strain keeps increasing over time and gradually approaches a plateau. Overall, it takes 0.1–0.5 s to achieve 50% of full strain and 2–14 s to reach 90% (see **Table 1**). Such slow responses result from the highly entangled and sparsely crosslinked silicone network: the entangled structures slow down the response due to their sliding ability upon

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Table 1. Required times to reach 50% and 90% of achieved strain under DC conditions.

		Time for achiev	Time for achieved strain [s]		
Dielectric elastomer actuator	<i>U</i> [V]	50%	90%		
DEA_1100 μm_3	4000	0.40	14.3		
DEA_1100 μm_6	1200	0.19	5.1		
DEA_280 µm_3	1000	0.21	9.7		
DEA_280 μm_6	250	0.14	2.1		
DEA_150 μm_3	400	0.46	7.2		

external stress, while the covalently crosslinked chains respond faster because they are fixed until break.

DEAs with a prestretch ratio of 6 respond faster to voltage compared to DEAs with a prestretch ratio of 3. In particular, DEA_280 μm_3 requires 0.21 s and 9.7 s to reach 50% and 90% of the full strain, respectively, while DEA_280 μm_6 exhibits lower respective response times of 0.14 and 2.1 s. Faster response times can be explained by the shorter slippage distance of the entangled structures under increased constraint.

Actuator lifetime, defined as the time required to reach 100% failure of devices, is another important feature when evaluating DEAs for practical applications. The failures of the DEAs in this study were caused by electrical breakdown, which was confirmed by the presence of visible pinhole defects after testing. As Table 2 shows, mean lifetime for each batch DEA vary considerably under DC conditions, from 0.9 h to more than 123.0 h, depending primarily on the electrical field applied. For example, DEA_280 µm_3 samples fail after applying an initial electrical field of 36.3 V µm⁻¹ for only 0.9 h. DEA_150 µm_3, on the other hand, survives for 123.0 h before failure under a much lower initial electrical field of 19.6 V µm⁻¹. However, lower applied electrical fields often lead to lower strains, as confirmed by the strain ranges of 4.9-9.3% versus 1.4-2.6% for DEA_280 µm_3 and DEA_150 µm_3, respectively. Actuator lifetime and achieved strain are thus the most important considerations when selecting which electrical field to apply. A commercial silicone (Wacker Elastosil 2030/20) based DEA is compared with the DEAs in this study. Table 2 shows that reported DEA can be actuated to lateral strains of 2.5-3.0% for more than 100 h under 600 V. In comparison, DEA_150 µm_3 can be actuated to lateral strains of 1.4-2.6% under only 300 V for 123.0 h. In addition, the much thicker initial thickness of DEA_150 μm_3 usually means easier fabrication than that of the 20 μm -thick silicone film.

The prestretch has been reported to have a detrimental effect on DEAs' lifetime.^[32] In our case, it can be noted that DEAs with an equibiaxial prestretch ratio of 6 (**Figure 5b**,d) displayed a high incidence of early failure (4/5), while the failure of DEAs with an equibiaxial prestretch ratio of 3 is more evenly dispersed over time. Early failures of DEA samples may come from defects and thickness variations during the DEA fabrications. This more frequent early failure may be caused by relatively larger sizes of the defects with respect to the prestretched thickness under the higher equibiaxial prestretch ratio.^[43] Considering the limited number (5 or 6) of DEAs in parallel per type of DEA, more DEA testing data are required to more reliably confirm the trend.

Enabling steady strains over time under fixed voltages is important to ensure DEA function for long-term applications. Representative strain-time plots are shown in Figure 6. Overall, strain in the presence and absence of an electrical potential increases with time until electrical breakdown. Strain increase over time with an applied potential results from the fact that constant actuation exercises a softening effect on the silicone elastomer. The strain does not go back to zero when the voltage is turned off, indicating a response hysteresis, i.e., permanent set. What is more, increasing the strain leads to reduced film thickness and increased electrical field, likely reducing the lifetime. However, there is no clear indication that the electrical breakdown originates from pull-in instability due to the limited strain. Strain ranges for the various DEAs studied here are summarized in Table 2. DEA_1100 µm_3 shows the widest strain change: from 3.5% to 15.2% over its full actuation lifetime. Although a high strain (over 10%) is achieved, such a high degree of strain instability over time is an obstacle to practical DEA applications. With a larger prestretch, DEA_1000 µm_6 exhibits a much narrower strain range of 2.8-4.0% over its actuation lifetime (Table 2). As the observed results for DEAs with an initial thickness of 280 µm show, higher prestretch significantly reduces strain change over time (Table 2). Additionally, variations of strain ranges are observed between different DEAs of the same category (Figure 5). They may be caused by the variations of initial thickness and prestretch ratio, which eventually induce the variations of prestretched thickness and Young's modulus.

As with their faster observed responses, the narrower strain ranges of DEAs under larger prestretch can be explained by the

Table 2. Summary of lifetime tests of the DEAs in this study, and a reported DEA which is fabricated from a 20 μ m thick silicone film (Elastosil 2030/20, Wacker) and carbon black powder electrodes.^[48] All the DEAs in this study were tested under 25 °C and 50% relative humidity. The reported DEA was tested under 20 °C and 30%/90% relative humidity.

	Average prestretched thickness	U	Range of s _x	Range of electrical field	Mean lifetime
DEA	[μm]	[kV]	[%]	[V µm ⁻¹]	[h]
DEA_1100 μm_3	120	2.0	3.5-15.2	17.8–22.1	>42.0
DEA_1100 μm_6	27	0.9	2.8-4.0	34.9-35.7	6.7
DEA_280 μm_3	30	1.0	4.9-9.3	36.3–39.4	0.9
DEA_280 μm_6	8	0.3	1.1–1.7	37.0–37.3	47.9
DEA_150 μm_3	16	0.3	1.4–2.6	19.6–20.1	123.0
DEA based on Elastosil 2030/20	12	0.6	2.5-3.0	52.1–52.6	>100 h



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Figure 5. Failure incidence over time for various DEAs. Five or six of each type of DEA were tested in parallel under the same DC voltage source. The applied voltage and strain range for each DEA is shown above, where the minimum strain refers to the strain once the voltage is first applied and the maximum strain refers to the strain at failure.



Figure 6. Example plots of strain over time for each type of DEA. DEAs are cyclically actuated with DC voltages. For each cycle, the target voltage is constantly applied for 1 h and then decreased to zero for 30 s.





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Figure 7. DEA configuration and actuation setup. a) Prestretched DEAs were fabricated with carbon black powder electrodes and an active 5 mm diameter circular region in the center; b) a previously reported setup was used to test the lifetime the fabricated DEAs,^[43] in which the DEAs were actuated by a controlled DC voltage source and the actuations were monitored by camera from the top of each DEA. c) Control procedure of applied voltages for lifetime tests ($U_{applied}$ for each DEA is shown in Table 2); d) control procedure of applied voltages for response speed tests.

Table 3. Sample specifications of fabricated DEAs. DEA_1100 µm_3 indicates an initial film thickness of 1100 µm and an equibiaxial prestretch ratio of 3
Prestretched DEA thicknesses were measured by transmission interferometer, except that of DEA_1100 μm_3, which had to be estimated due to nois
signals.

Dielectric elastomer actuator	Initial thickness [µm]	Equibiaxial prestretch ratio	Prestretched thickness [µm]		
			Strain-voltage curve tests	Response speed tests	Lifetime tests
DEA_1100 μm_3	1096± 16	3	≈ 122	≈ 122	≈ 122
DEA_1100 μm_6		6	30.0 ± 0.0	28.3 ± 0.5	27.3± 1.3
DEA_280 μm_3	277± 2	3	37.5± 1.7	29.0± 2.2	30.3± 2.6
DEA_280 μm_6		6	6.9± 0.0	9.3± 0.1	8.3± 1.2
DEA_150 μm_3	148± 6	3	12.0± 0.4	13.0± 0.0	15.7± 0.9

more constrained network shape and shorter slippage distance of entangled chains.

While the relatively slow response time and obvious strain changes over time of the DEAs studied here are not universally favorable for all potential practical applications, they are advantageous in applications which favor actuator damping. Various strategies have also been developed to overcome the viscous effects of acrylic-based DEAs, including regulating applied electric fields,^[44,45] modeling viscoelastic behavior,^[46] and changing states of robotic or mechatronic systems in discrete steps.^[47] By applying these strategies in the DEAs described above, it should be possible to control strain more accurately in the future.

3. Conclusion

In this work, we fabricated and tested a highly stretchable silicone elastomer. The maximum uniaxial strain of this elastomer is 2300%. The elastomer exhibits an obvious stress relaxation behavior and a relatively high viscous loss compared to conventional silicone elastomers. The relative dielectric permittivity of the elastomer in a free standing state is 3.3, which is reduced to 2.5 and 2.4 for equibiaxial stretch ratios of 3 and 6, respectively.

DEAs with initial thicknesses of 1100, 280, and 150 μ m, and equibiaxial prestretch ratios of 3 and 6 were fabricated from the highly stretchable silicone elastomer. Strain–voltage plots show that DEA_1100 μ m_3 fabricated with a 122 μ m thick prestretched film can be actuated to a 30% strain at 4.3 kV—a much larger degree of strain than is achievable with common silicone-based DEAs. Applying a higher prestretch ratio or a lower initial thickness can greatly reduce the applied voltage. For example, DEA_280 μ m_6 fabricated with a 6.9 μ m thick prestretched film can be actuated to a strain of 2.5% at only 250 V.

Due to the viscous nature of the highly stretchable elastomer used here, the fabricated DEAs respond slowly and show strain increases over time compared to conventional silicone-based DEAs—taking 0.1–0.5 and 2–14 s to reach 50% and 90% of fully strain, respectively. DEA_1100 μ m_3 shows the widest strain range of 3.5–15.2%, while applying a higher prestretch ratio of 6 narrows this to 2.8–4.0%. This inherent viscosity is not favorable for all potential actuator applications, but is advantageous for those in which actuator damping is desirable, such as in loudspeakers. Importantly, prestretch ratio can be used to tune DEAs' response behavior.

DEAs ' mean lifetimes vary remarkably—from 0.9 h to more than 123.0 h—depending primarily on the applied electrical field. For example, DEA_150 µm_3 can be actuated to a lateral strain of 1.4–2.6% for a mean lifetime of 123.0 h under an applied voltage of 300 V. Given the strains it is possible to achieve, DEAs show promise for use in applications where a high degree of prestretch is permissible and fast response times are not required.

4. Experimental Section

Materials: Vinyl-terminated PDMS (DMS-V41, $M_n = 63$ kDa; DMS-V25, $M_n = 14$ kDa), hydride-terminated PDMS (DMS-H25, $M_n = 14$ kDa), methyl hydrosiloxane-dimethylsiloxane copolymer (HMS-301, 1.9 kDa), and platinum-divinyl tetramethyldisiloxane complex (SIP6830.3, 3.0 wt% Pt) were purchased from Gelest, Inc. Platinum cyclo-vinylmethyl siloxane complex (catalyst 511, 1.0 wt% Pt) was purchased from Hanse Chemie. Dry carbon black powder (Ketjenblack EC-600JD) was purchased from Nouryon.

Silicone Elastomer Preparation—Preparation of Highly Stretchable Silicone Elastomer: A vinyl-terminated PDMS (DMS-V25, 7.08 g, 5.06×10^{-4} mol) and a platinum catalyst (SIP 6830.3, 1.5 mg, 2.31×10^{-7} mol) were mixed for 4 min at 2500 rpm using a speed mixer. Subsequently, a hydride-functional PDMS (DMS-H25, 7.92 g, 5.66×10^{-4} mol) was added into the mixture and mixed for 2 min at 2500 rpm. In order to prepare a 1100 μ m thick film, the mixture was poured onto a Teflon mold. For thinner films (280 μ m thick films and 150 μ m thick films), the mixture was cast onto PET film using an applicator. The cast mixture was then placed in an oven at 100 °C for 24 h.

Preparation of Conventional Silicone Elastomer: Before a crosslinking reaction, parts A and B were prepared. Part A was prepared by mixing a vinyl-terminated PDMS (DMS-V41, 10.00 g, 1.59×10^{-4} mol) with a methyl hydrosiloxane-dimethylsiloxane copolymer (HMS-301, 0.14 g, 7.33×10^{-5} mol) using a speed mixer at 3500 rpm for 2 min. Part B was prepared by mixing a vinyl-terminated PDMS (DMS-V41, 4.86 g, 7.75×10^{-5} mol) with a platinum complex catalyst (catalyst 511, 6 mg, 3.08×10^{-6} mol) using a speed mixer. Parts A and B were then mixed using a speed mixer at 3500 rpm for 30 s. The final mixture was poured into a mold and placed in an oven at 100 °C for 10 h.

Silicone Elastomer Characterization: Tensile Test: A universal testing system (Instron 3340 series, US) was used for uniaxial tensile tests. Specimens were cut into a dumbbell shape using ASTM D-638 Type V (width × length × thickness = 3.18 mm × 9.53 mm × 1 mm) as a standard. Specimens were then stretched with a crosshead speed of 500 mm min⁻¹, and true stress was determined by multiplying the measured engineering stress by the stretch ratios.

Linear Viscoelasticity Analysis: Elastomers' linear viscoelastic properties were analyzed using a strain-controlled rheometer (ARES G2, TA Instruments). Cylinder-shaped specimens (\approx 1 mm thick and 8 mm in diameter) were analyzed at room temperature with a fixed shear strain of 1% and a frequency range of 0.01–100 Hz.

Dielectric Permittivity Analysis: Dielectric relaxation spectra were obtained using a Novocontrol Alpha-A high-performance frequency analyzer (Novocontrol Technologies GmbH & Co, Germany). Specimens, including free-standing films and prestretched films, were coated with round silver layers (each 20 mm in diameter and 5 nm thick) on two sides using a rotary pumped coater (Q150R Plus, Quorum). Tests were performed at a frequency range of 10^{-2} – 10^5 Hz at room temperature and an electrical field of 1 V mm⁻¹.

DEA Fabrication: Silicone elastomer films were equibiaxially prestretched using a radial prestretcher with 8 metallic fingers. The prestretched films were then fixed by rigid frames. Details of both the prestretching process and the frames have been previously published.^[49] Except for the thickest prestretched films, which had an initial thickness of 1096 \pm 16 μ m and a prestretch ratio of 3, the thickness of all prestretched films was measured using a transmission interferometer. The thickest prestretched film could not be measured this way due to noisy signals, but was estimated to be 1096 μ m/(3 \times 3) = 122 μ m. Dry carbon black powder was applied with a paintbrush to the two sides of the prestretched films through a mask. The overlaid area of the two-sided electrodes was a 5 mm diameter circle in the center. The fabricated DEAs are shown in Figure 7a, and their specifications are given in Table 3. The names of the DEAs contain information regarding initial thickness and prestretch ratio: for example, DEA_1100 µm_3 indicates an initial thickness of 1100 µm and an equibiaxial prestretch ratio of 3.

Actuation Test: Strain–voltage curve and lifetime tests were conducted using the MAPLE setup.^[43] For strain–voltage curve tests, step-increased voltages were applied on the DEAs until electrical breakdown occurred: step duration was set at 20 s, and the lateral strain was measured every 5 s. For lifetime tests, 6 DEAs in parallel were cyclically actuated with the same voltage source. For each cycle, voltages were increased stepwise until the target voltage was reached ($U_{applied}$ values are shown in Table 2), with a step duration of 20 s. Once reached, target voltages were maintained for 1 h and subsequently dropped to zero for 30 s (Figure 7c). By doing this, permanent sets and strain–voltage curves of the DEAs at each hour can be monitored. The lateral strain was measured every 5 s during the high voltage stage. The MAPLE setup was operated inside an environmental chamber in which the temperature and relative humidity were set to 25 °C and 50%, respectively.

Response speed tests were conducted by recording DEA actuations with a camera. Voltages were supplied by a single channel high voltage power supply, and were manually increased stepwise as shown in Figure 7d. For each step, the voltage was maintained for 30 s and then dropped to zero for another 30 s. The 30 s-actuation duration was set due to the highly viscous property of the films. The recorded videos (pixel size: 640×480 , 15.02 frames s⁻¹) were analyzed using Tracker software.

Lateral strains (s_x) are determined as

$$s_{\rm x} = \frac{D_{\rm act} - D_{\rm ref}}{D_{\rm ref}}$$

where D_{act} is the diameter of the actuated circular electrode. For both strain–voltage curve and lifetime tests, D_{ref} is the diameter of the circular electrode before actuation; for response speed tests, D_{ref} is the diameter of the circular electrode when voltage is turned off for each step. Assuming incompressibility of the silicone material, the thickness strain (s_z) is determined as

$$s_z = \frac{1}{(s_x + 1)^2} - 1$$

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Conflict of Interest

The authors declare no conflict of interest.

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Data Availability Statement

The data that support the findings of this study are available from the corresponding author upon reasonable request.

Keywords

dielectric elastomer actuators, high stretchability, long-term stability, lowvoltage actuation, silicone elastomers

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