Supporting Information

Uniform Conductivity in Stretchable Silicones via Multiphase Inclusions

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A note on the **Supplemental Video 3**.
Data at timestep t was normalized as $\overline{S_t} = \frac{1/S_t}{1/S_0} = \frac{S_0}{S_j}$, such that the initial reading for each sensor is one. This corresponds to normalized capacitance C/C_0, since capacitance is linearly related to the voltage reported by the MPR 121.

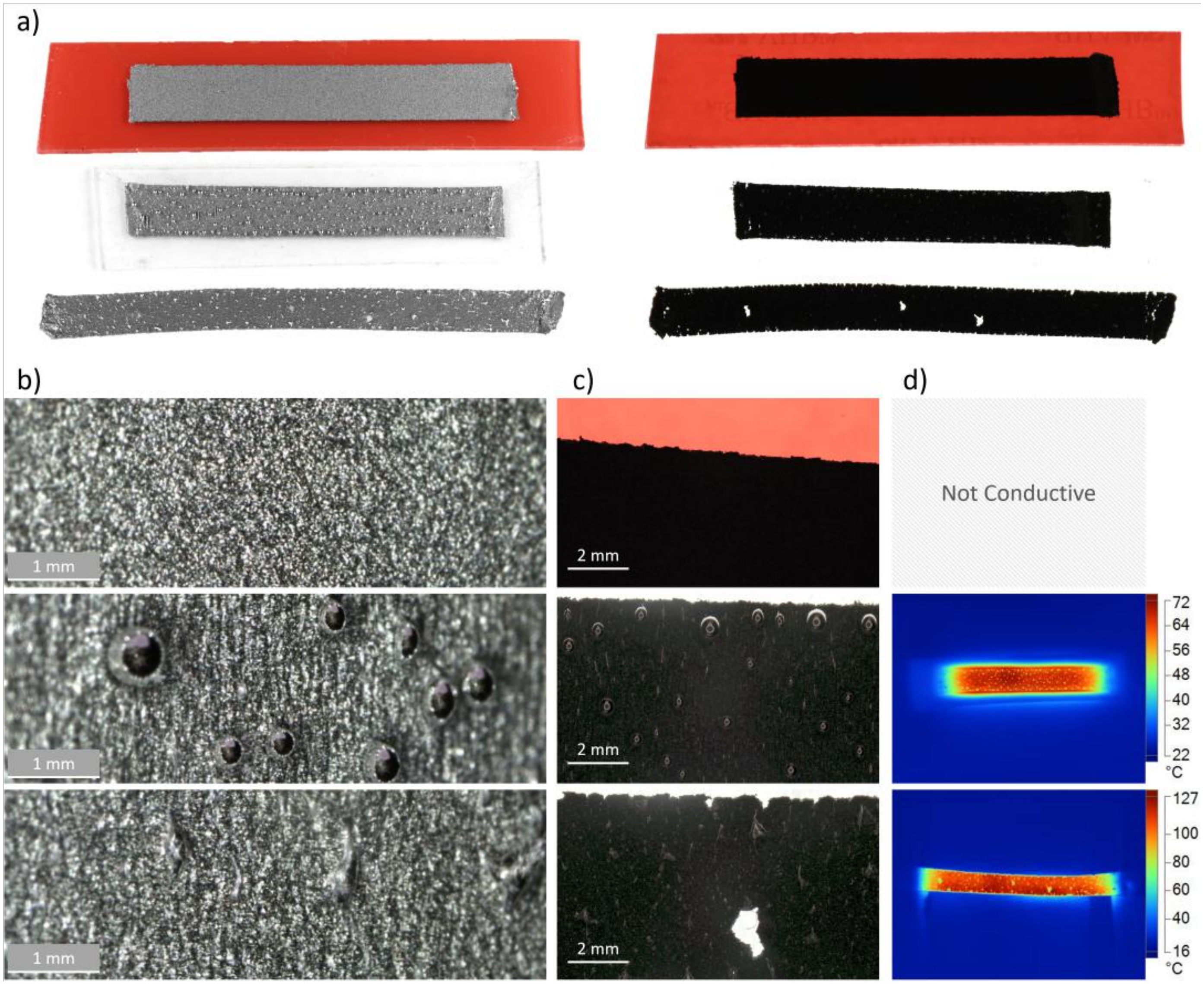

Fig. S1 The MPC silicone is robust enough to function without a silicone backing layer, if it is supported by a removable backing during the activation strain event. a) Front and back lighting of three 5 cm x 1 cm samples (as cast dimensions) of MPC silicone that are (top) unstrained, (mid) activated via 300% strain activation, and (bottom) activated then removed from the backing. (Top) The MPC silicone is laid on top of a stretchable adhesive tape, which serves as a support backing layer (VHB 4910, 3M). The tape is dispensed with a red, inextensible release liner which helps ensure that the sample does not strain prematurely. (Middle) The MPC silicone is stretched with the tape backing, which has been removed from the red release liner. (Bottom) The MPC has been removed from the tape backing. Note the sample extension from it's as-cast length — the internal microcracking that resulted in material activation result in a new, longer, rest state. b) Front-lit microscope images of the same three samples. Note the wrinkling in the activated sample (middle) which diminishes after the sample is allowed to relax when removed from the temporary backing layer. c) Back-lit microscope images showing the damage that is added to the material and is an integral part of the activation process. d) IR images demonstrate that even without a permanent backing layer, the material still activates resulting in a uniform electrical resistivity. Note that once completely removed from the adhesive backing (bottom IR image), there is very little mass in the system and so it heats up very quickly (< 3 s at 2 A).

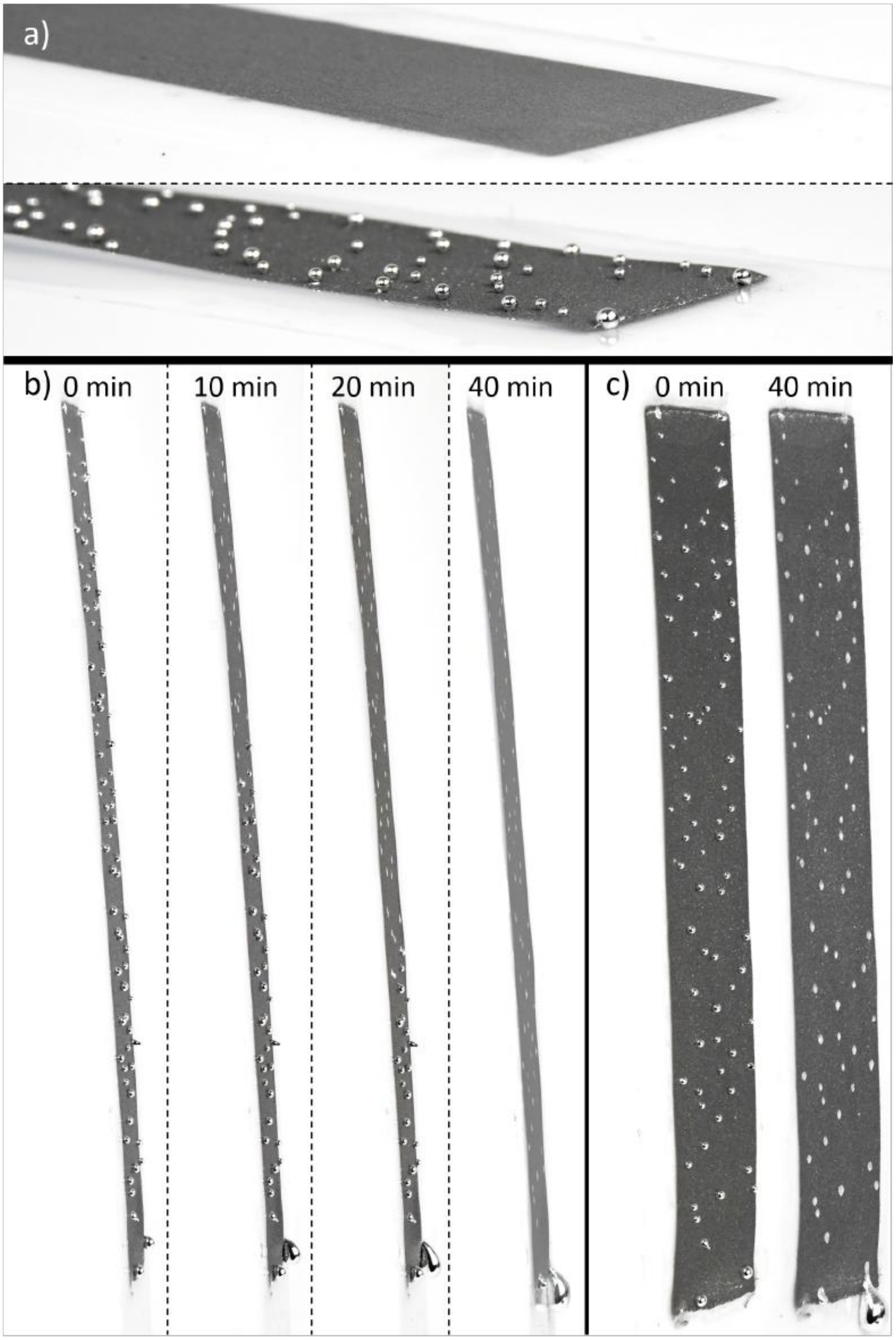

Fig. S2 Evidence of the internal liquid metal network fully percolating the sample end-to-end. a) A sample lying flat before and after a pre-mature activation strain of only 200% (mature, complete activation requiring at least 250%). Although this is insufficient strain for uniform heating, it does allow for larger beads of liquid metal to form on the surface, for easier observation. b) Sample held vertically for 40 minutes results in a withdrawal of the liquid metal from most of the surface beads, pooling in the bottom-most bead due to gravity. c) A front view of the sample before and after the 40 minutes shows that the liquid metal did not track down the surface, but rather it flowed through an internal interconnected network. Sample is 20 cm x 4 cm.

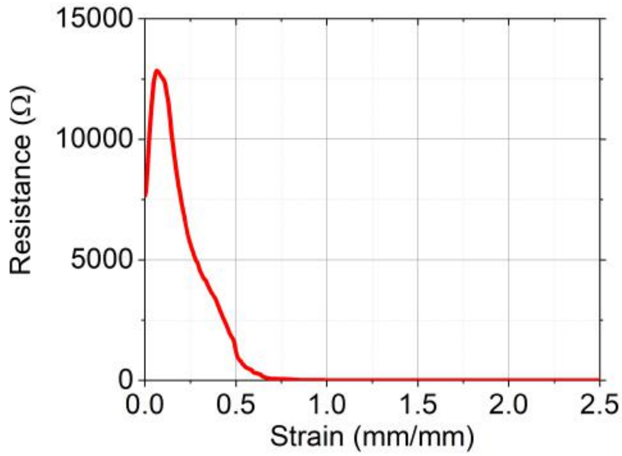

Fig. S3 Resistance drop of a representative 60-40-1 sample with the dimensions of 20 × 10 × 0.2 mm strained up to 250% with a strain rate of 5 mm/s.

Table S1 Standard deviations (σ) of the four test conditions described in Materials and Methods. Each reported value was calculated using 60 seconds of data recorded at 100 Hz for each sensor, shifted to have zero mean, and then aggregated prior to calculating standard deviation. Three graphite sensors were tested, while eight MPC sensors were tested. MPR is a 10-bit (valid range from 0 to 1023) system with typical full-scale range of ≈ 400 over 200% strain, while our 16-bit (0 to 65,535) charge integration circuit (described in [56]) changed ≈ 13000 over 200% strain. It is unclear why standard deviations decreased when a motor was placed near the sensors while using the MPR circuit.

Sensor Type	**MPR 121**		**Custom Circuit** [56]	
	No Motor	Motor	No Motor	Motor
Graphite	0.61	0.46	25.02	188.02
MPC	0.25	0.20	16.07	146.36

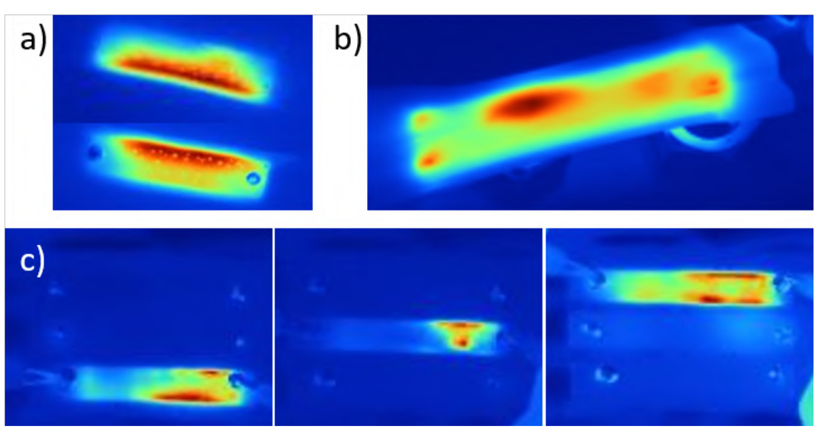

Fig. S4 IR images of samples of MPC silicone (5 cm x 1 cm) that did not activate uniformly, even though their resistance dropped below 10 Ω. Non-uniformity is caused by a number of reasons: a) uneven cross-sectional thickness (variance of over 100%), b) bad material composition (not enough liquid metal), and c) bad activation (non-uniform stress fields during activation).

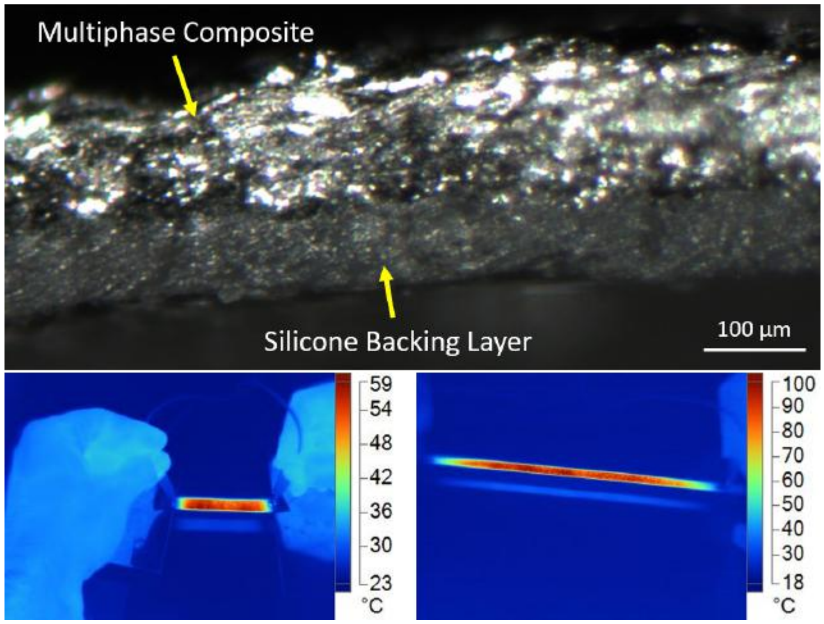

Fig. S5 Microscope images of the cross section of MPC silicone showing the active portion (approximately 150 μm thick), and the supporting silicone backing layer (approximately 75 μm thick). IR image demonstrating the uniform conductivity across the sample even when stretched to 200% strain. We note that the thickness can often vary across a sample as much as 30 μm (20% variance), an insufficient amount to effect the resultant electrical uniformity.

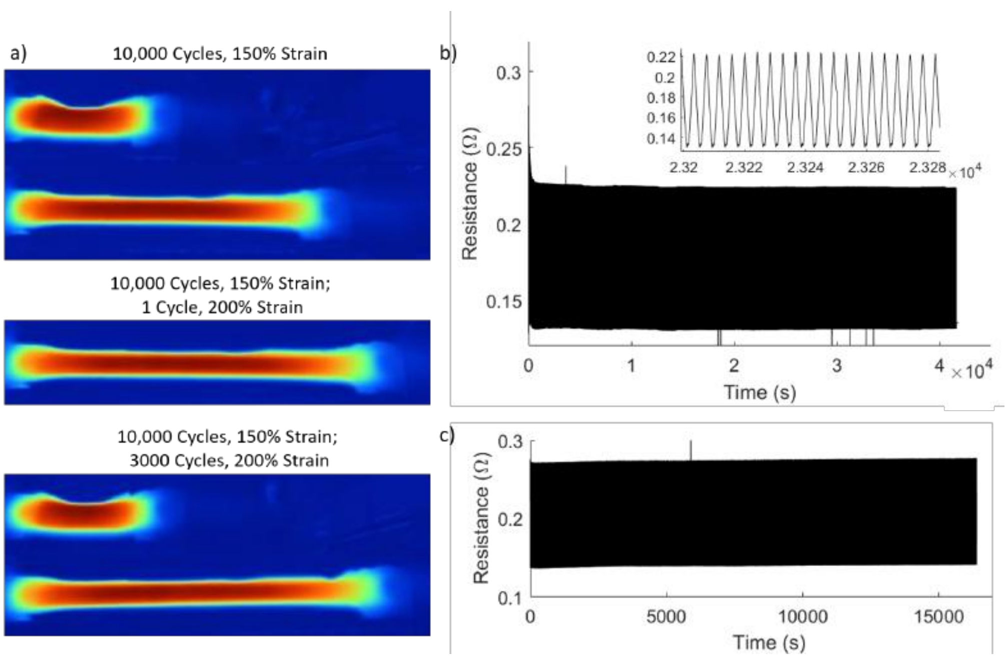

Fig. S6 Cyclic testing results of a 3 cm x 1 cm MPC silicone coupon, with only a single reinforcing backing layer of silicone (the other side is open to air). a) The sample continues to show consistently uniform conductivity (via Joule heating) after 10,000 cycles at 150% strain, and then after an additional 3,000 cycles at 200% strain. b) The resistance of the sample for all 10,000 cycles at 150% strain, with an inset showing 1 minute of cycling. c) The sample's resistance for its first 3,000 cycles at 200% strain. The sample subsequently failed (tore in half) after an additional 900 cycles at 200% strain.

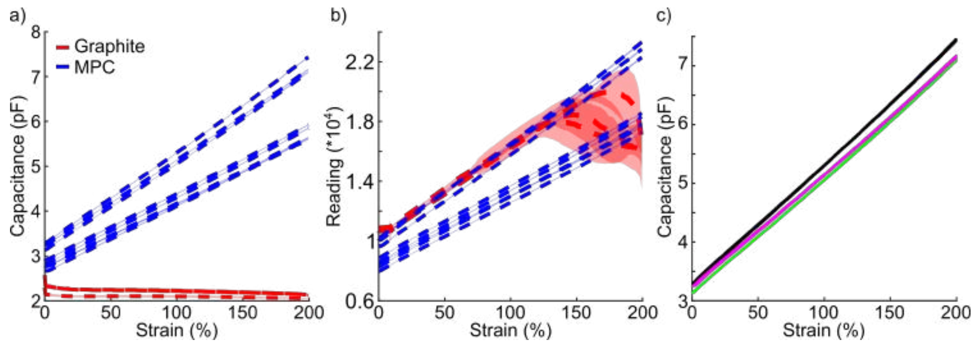

Fig. S7 Raw data for cyclic testing of strain sensors. a) The commercial circuit, MPR121, used to generate this plot, has excellent noise rejection but is ineffective at measuring capacitance of systems with high electrode resistance. b) The low, stable resistance over large strains results in stable capacitance readings when the MPC is used as an electrode material in a 3-layer capacitive strain sensor. Manufacturing variability leads to each sensor having slightly different intercept (corresponding to a rest capacitance) and slope, but are linear over small strains (both sensor types) and over large strains up to 200% (MPC silicone only). a,b) Dashed lines represent the mean for a single sensor over 10 strain cycles. Shaded regions represent 99% confidence intervals of a sample. c) All raw data, plotted independently without any filtering or transformations, for three MPC sensors cycled 1000 times to 200% strain. Each sensor is plotted in a different color (black, magenta, green).

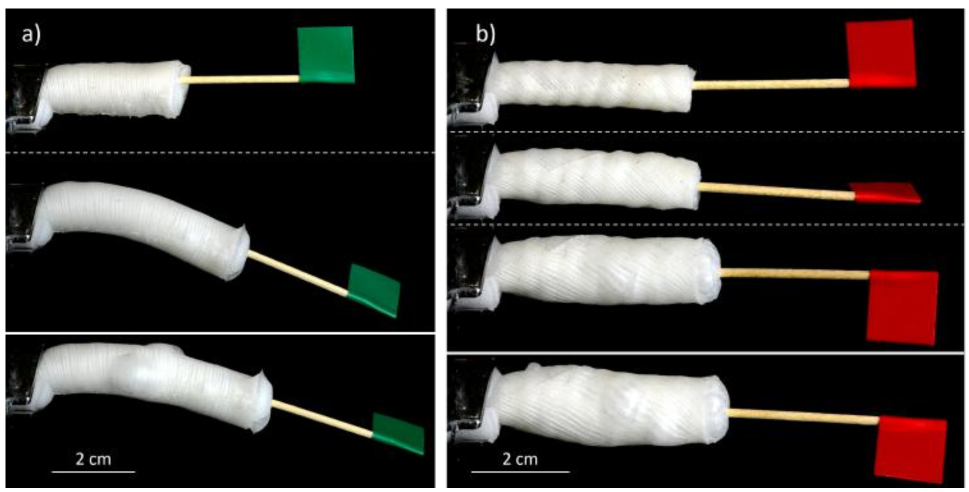

Fig. S8 Different actuation modes generated by changing the angle of the fiber wrap around the cylindrical tube of ethanol silicone. a) Fibers perpendicular to the length of the tube result in extension. b) A fiber angle of 45° results in $> 200^{\circ}$ rotation. Note: these samples were heated externally with a heat gun to allow fiber wrap to be visible.

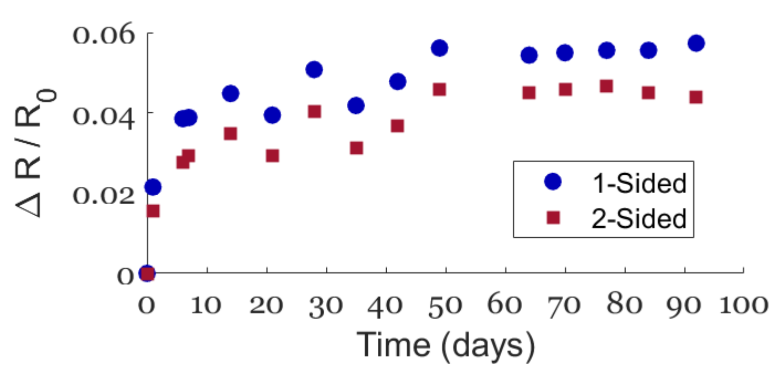

Fig. S9 The resistance across a 3 cm × 1 cm sample of MPC silicone over 92 days. After an initially sharp rise in resistance over the first 7 days, both samples settled into a very gradual increase for the remaining time.