

Supporting Information

Synthesis of polysiloxane elastomers modified with sulfonyl side groups and their electromechanical response

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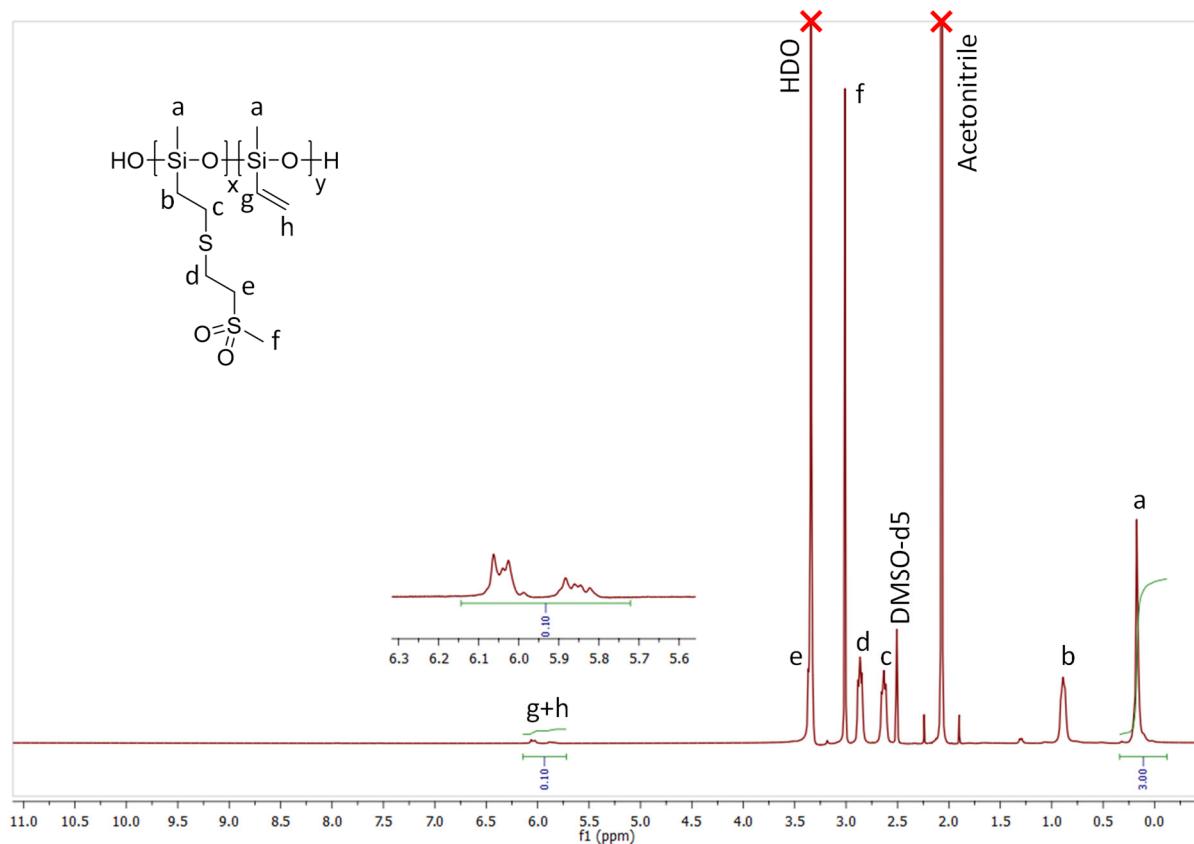


Figure S1. ¹H NMR spectrum of polysiloxane functionalized with 2-(methylsulfonyl)-ethanethiol side groups PSu (60 wt% in acetonitrile).

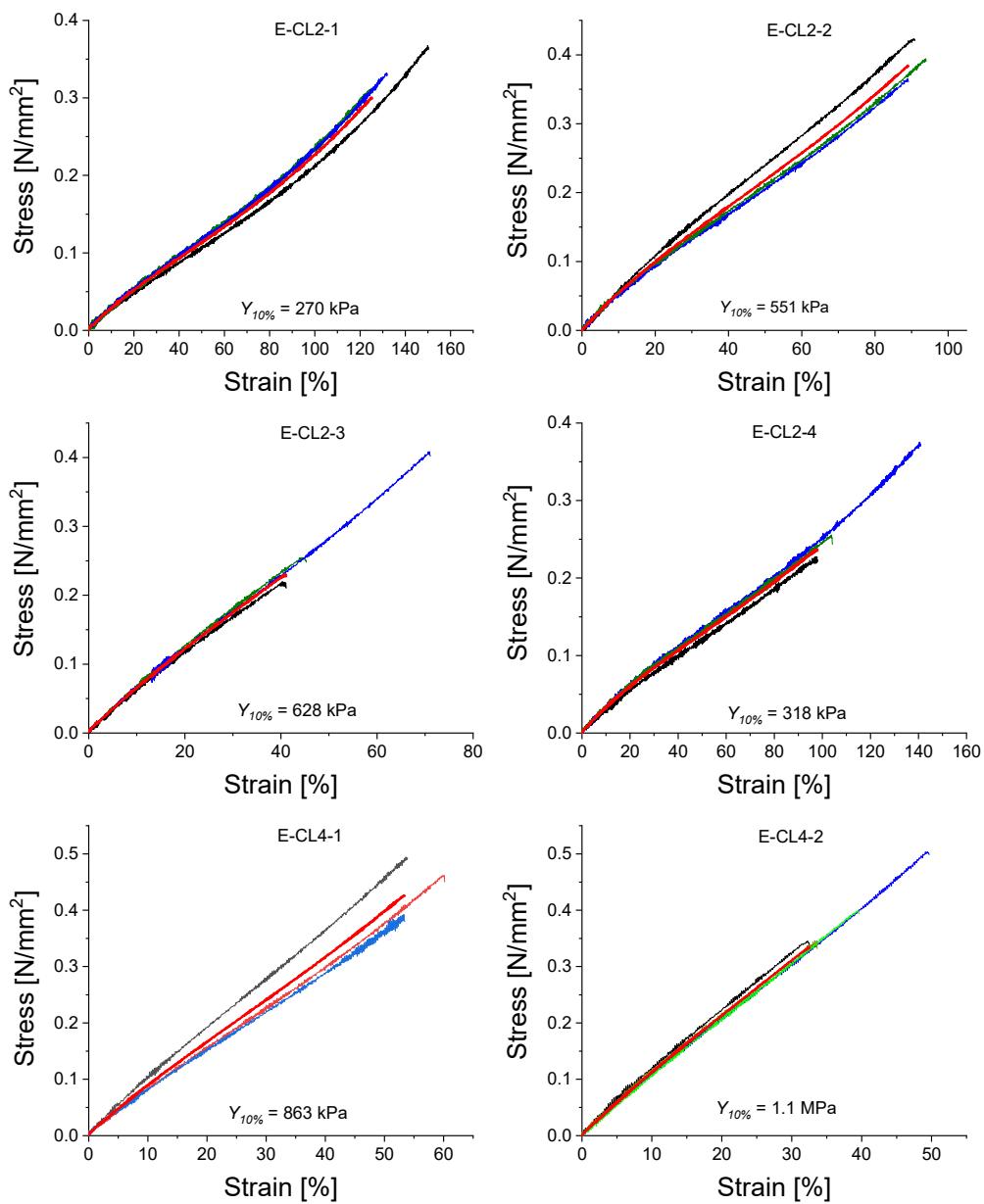


Figure S2. Stress-strain curves of elastomers **E-CLx-Y**. The red curve in each graph is the average of several measurements.

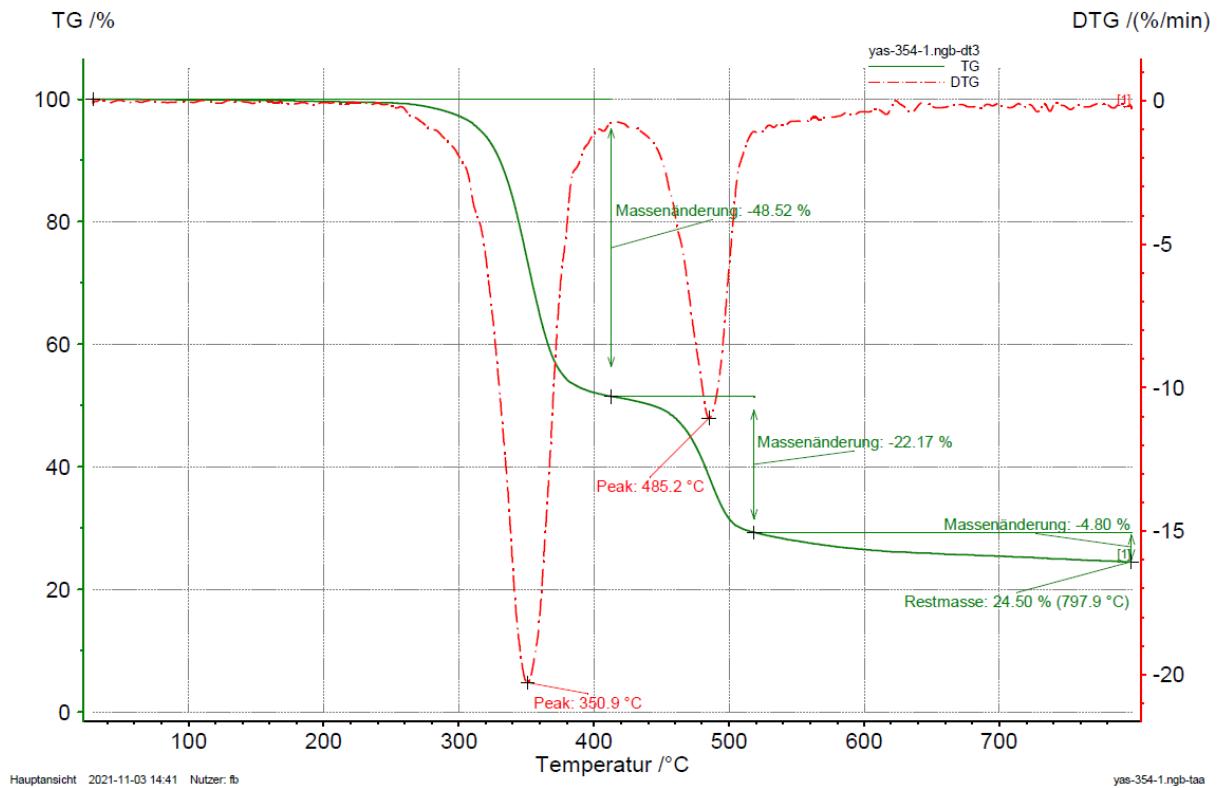


Figure S3. TGA curve of E-CL2-3

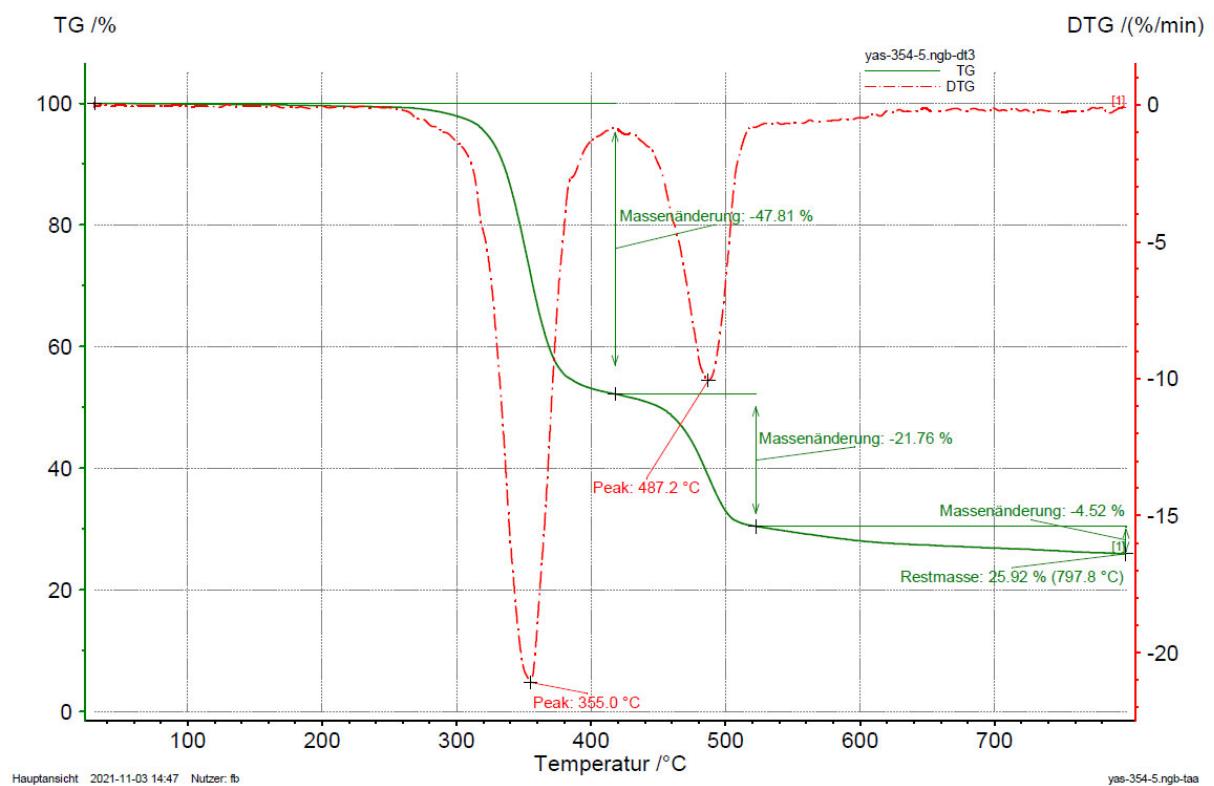


Figure S4. TGA curve of E-CL4-2.

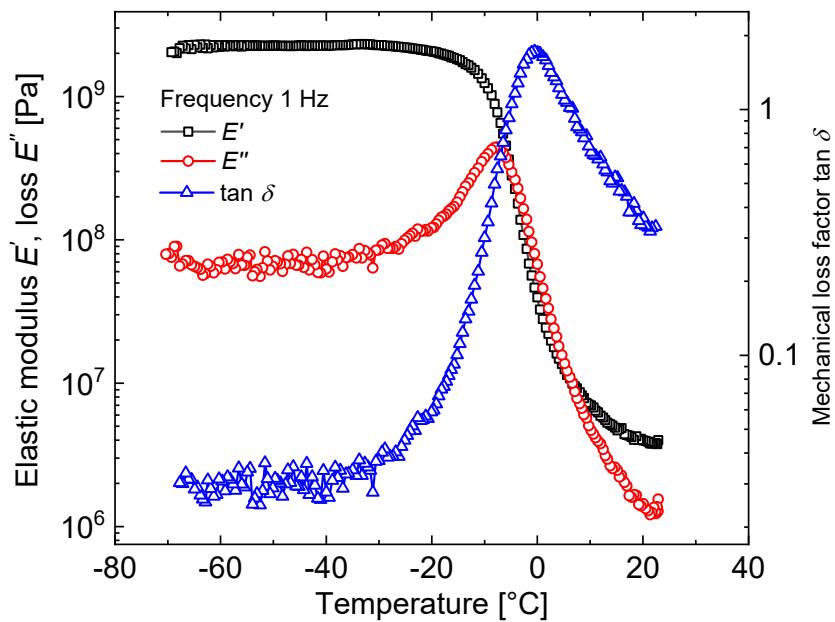


Figure S5. DMA measurement of **E-CL4-2** from -70 to 20 °C at 1 Hz. The peak in $\tan \delta$ at 0 °C represents the glass transition temperature.

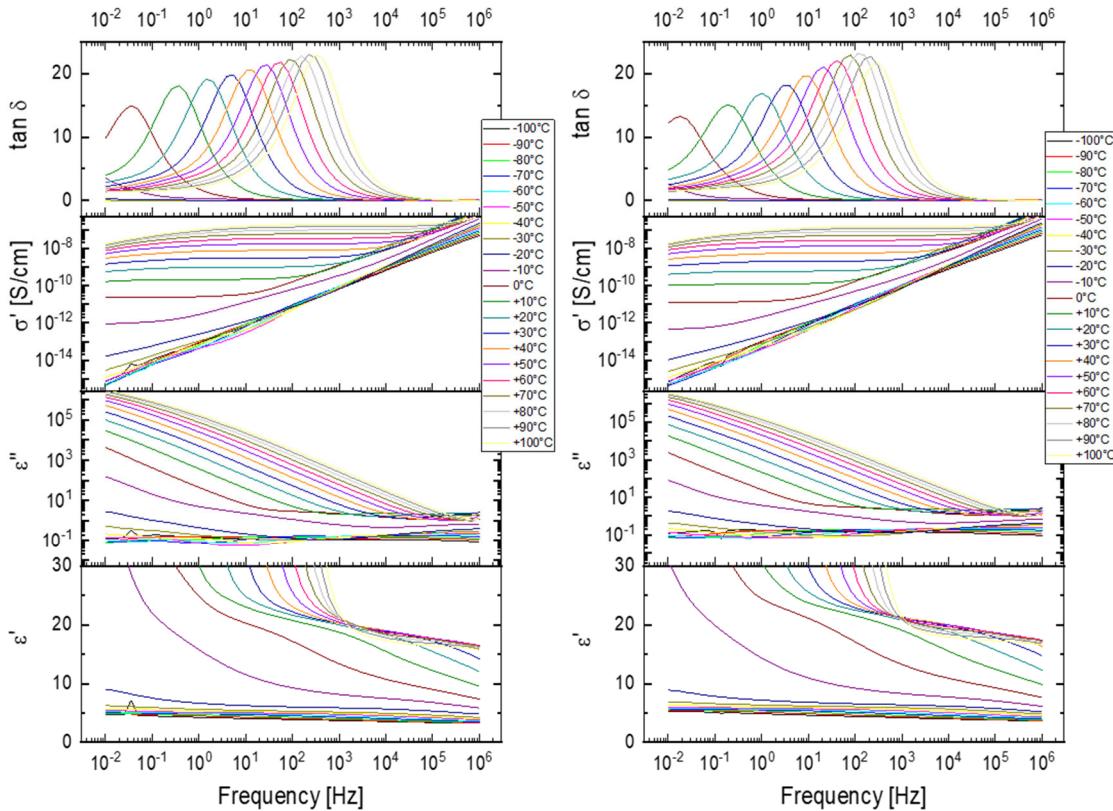


Figure S6. Dielectric permittivity (ϵ'), dielectric loss (ϵ''), conductivity (σ'), and $\tan \delta$ of **E-CL2-3** (left) and **E-CL4-2** (right) at different temperatures and frequencies.

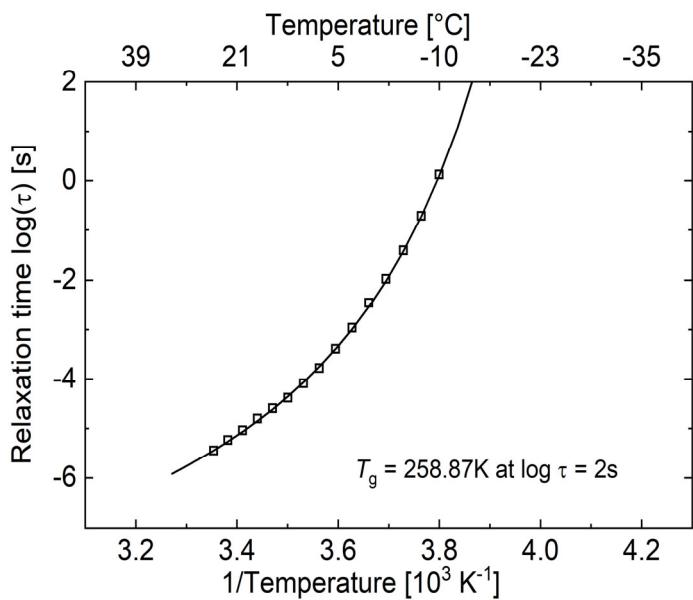


Figure S7. Arrhenius plot of the relaxation time versus temperature exhibiting Vogel-Fulcher-Tamman (VFT) behavior. A T_g of $-14.3\text{ }^\circ\text{C}$, close to the DSC value, was calculated at a relaxation time of 100 s ($\log \tau = 2\text{ s}$).

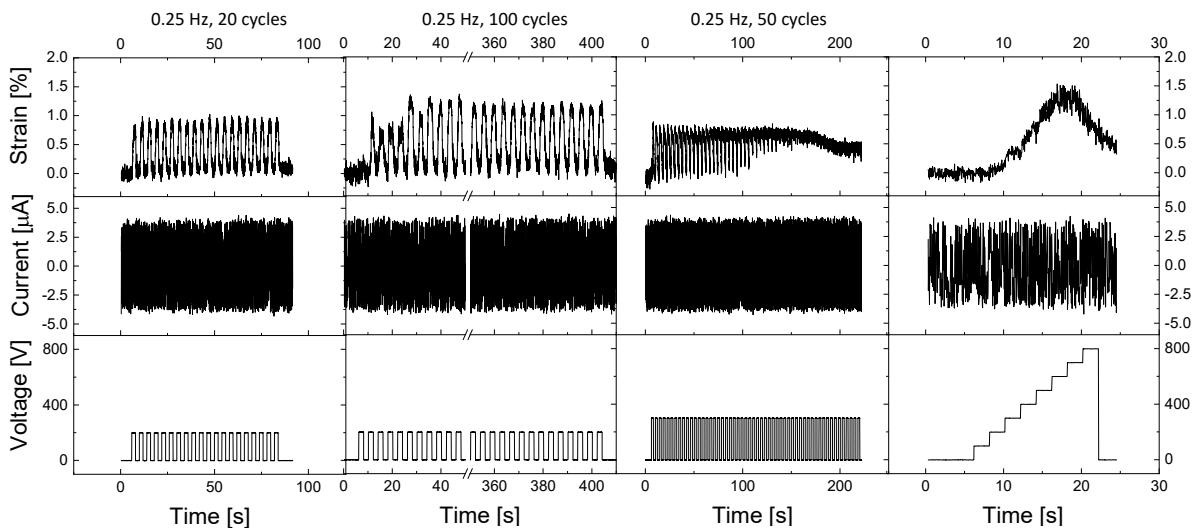


Figure S8. Actuator is made of material **E-CL2-3** with a thickness of $101\text{ }\mu\text{m}$. Four of more than ten measurements are presented, which describe the typical behavior of this actuator. First, the measurements were done at 200 V . When voltage was increased to 300 V , deterioration of actuation happened. Subsequent application of 800 V didn't result in a higher actuation strain.

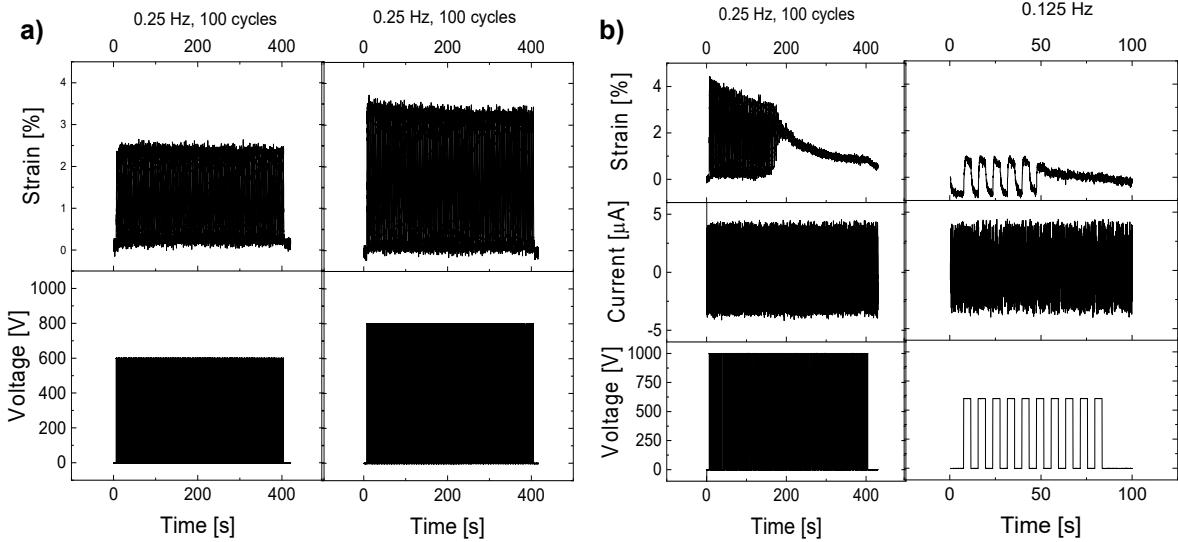


Figure S9. Actuation of a 99 μm thick actuator made of material **E-CL4-2**: a) stable actuation over 100 cycles at 600 V and 800 V and 0.25 Hz; b) degradation of actuation during the 100 cycles test at 1000 V and 0.25 Hz, and subsequent bad actuation at 600 V and 0.125 Hz.

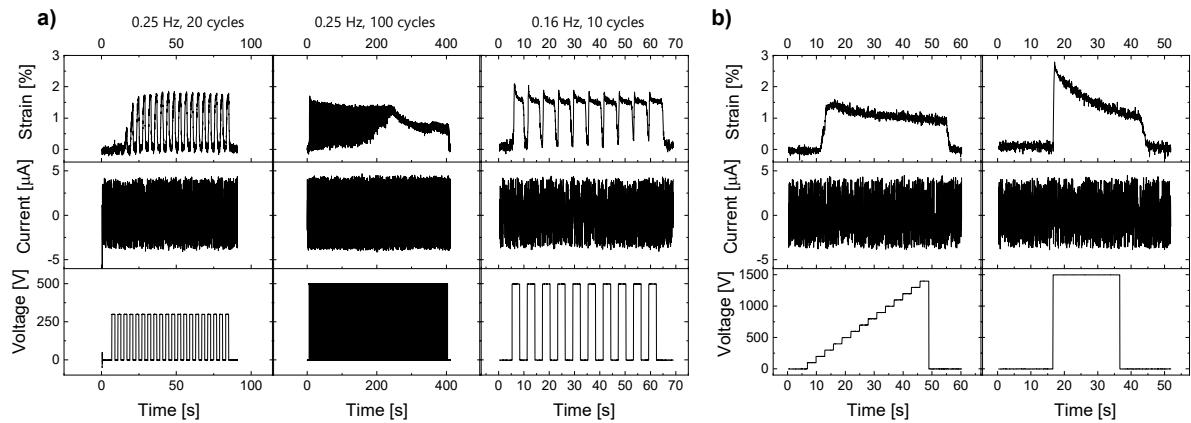


Figure S10. Actuation of a 101 μm thick actuator made of material **E-CL4-2**: a) a 1.8% strain is detected at 300 V and 0.25 Hz. Increasing the voltage to 500 V resulted in a deterioration of actuation. Subsequent application of 500 V at lower frequency (0.16 Hz) resulted in 1.8% actuation, but it can be seen that the actuator doesn't have enough time to relax back to the initial state; b) step voltage increase measurement up to 1500 V and a constant application of 1500 V withing 20 s.

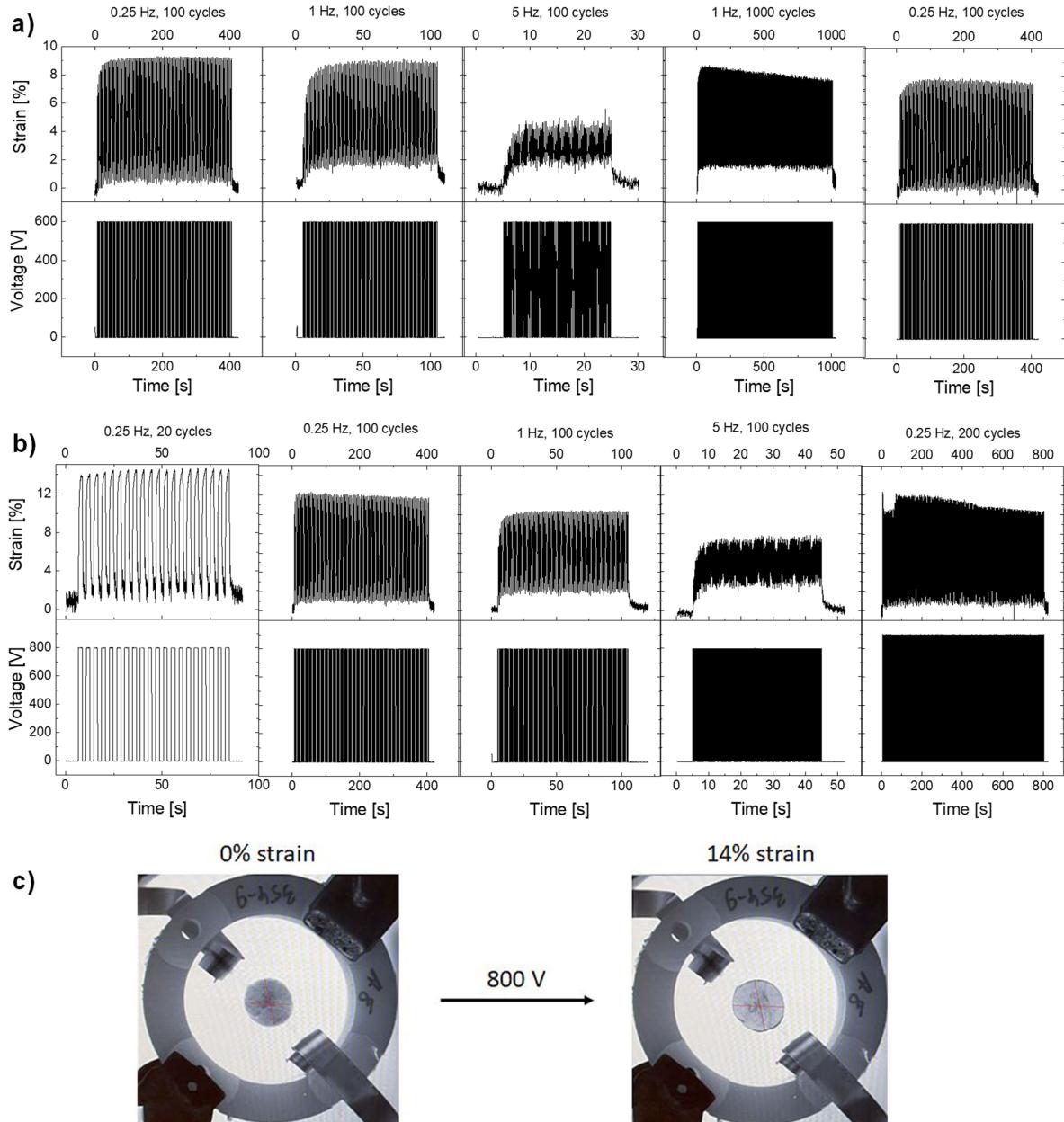


Figure S11. Actuation of a 33 μm thick actuator made of material **E-CL2-3**: a) actuation at 600 V and different frequencies; b) actuation at 800 V and different frequencies; c) photo of the actuator in relaxed and actuated (800 V, 0.25 Hz, 20c) states.

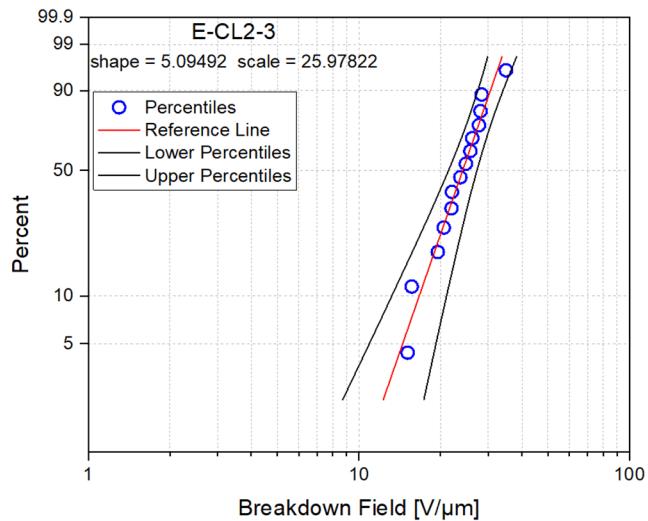


Figure S12. Weibull probability plot of breakdown field for materials **E-CL2-3**. The breakdown strength was tested by placing the material between two metallic electrodes of 1 mm^2 and gradually increasing the voltage until the breakdown was reached. At least ten samples with thicknesses in the range of 100-130 μm were tested. The average breakdown strength was $24 \text{ V } \mu\text{m}^{-1}$.

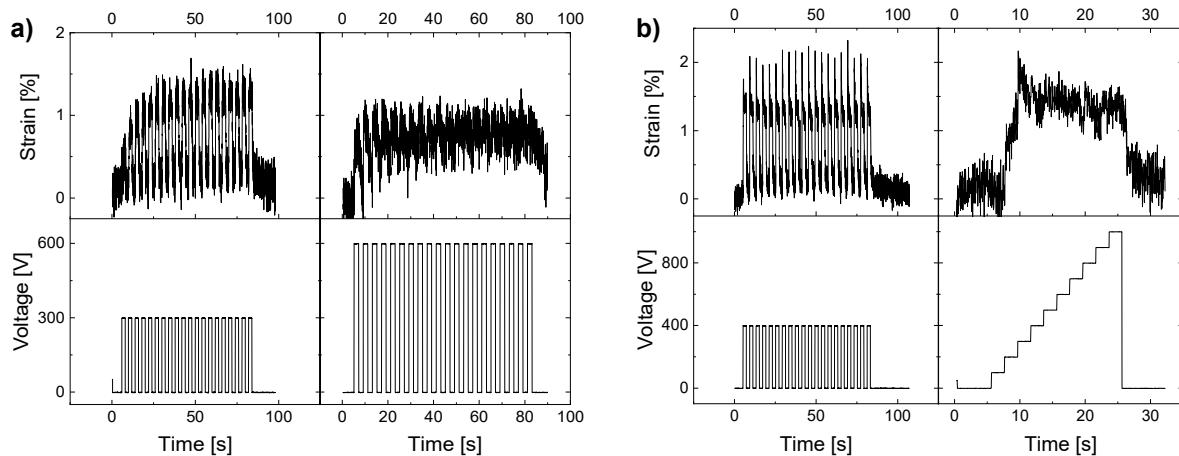


Figure S13. Actuation response of **E-CL4-2**: a) 34 μm film; b) 33 μm film.