Supporting Information

A polyphosphazene elastomer containing 2,2,2-trifluoroethoxy groups as a dielectric in electrically responsive soft actuators

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NMR Spectra

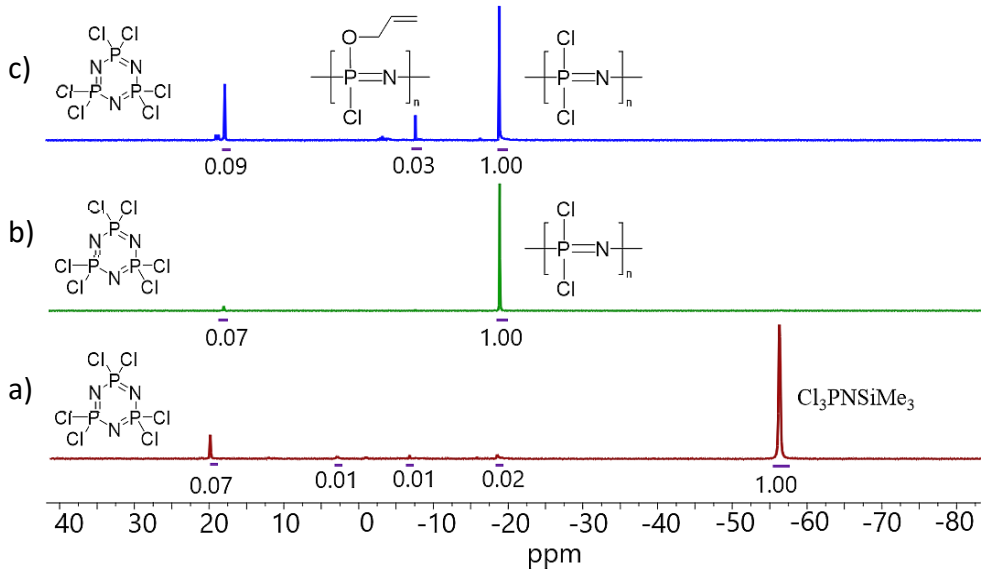

Figure S1. $^{31}P\{^{1}H\}$ NMR spectra ($CDCl_3$) of monomer $Cl_3PNSiMe_3$ with cyclohexachlorotriphosphazene (0.7 %<) and some oligomer impurities (0.2 %<) (a); polymer with cyclohexachlorotriphosphazene (0.7 %<) impurity (b); polymer after allyl alkoxide substitution (c).

GPC Spectrum

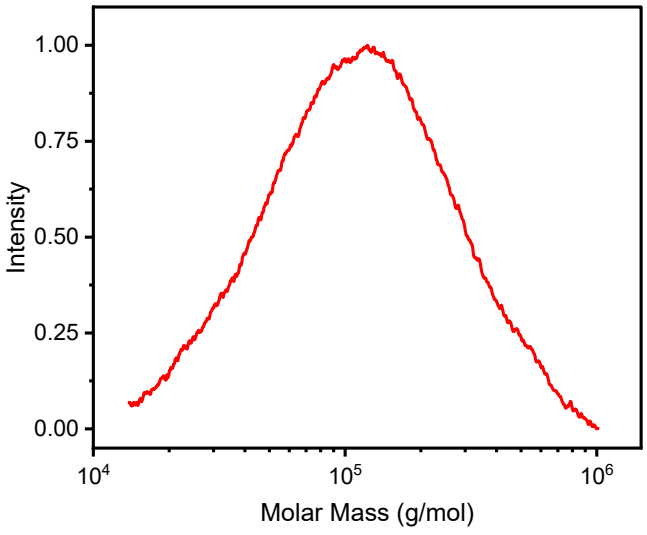

Figure S2. GPC elugram of poly[bis(2,2,2-trifluoroethoxy)]phosphazene.

Table S1. The molar mass and molar mass distributions result from GPC characterization.

Sample	M_n (g/mol)	M_w (g/mol)	PDI
poly[bis(2,2,2-trifluoroethoxy)]phosphazene	71926	149189	2.074

The polymer was synthesized by cationic polymerization of monomer $Cl_3PNSiMe_3$ in the presence of PCl_5 initiator. In general, the molecular weight is in the range of 10^5 g/mol with a polydispersity index of 1.2 to 2.5.[1]

FTIR Spectra

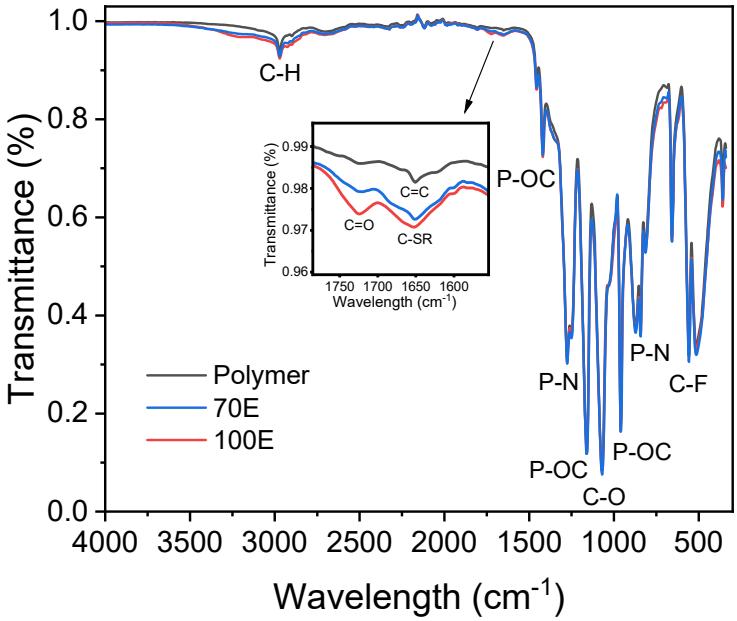

Figure S3. FTIR spectra of the poly[bis(2,2,2-trifluoroethoxy)]phosphazene, **E70** and **E100**.

FTIR spectra were used to determine the functional groups of the polymer and elastomers. Polyphosphazene backbone P-N-P shows an asymmetric stretching peak between 1200 and 1400 cm^{-1}. In general, electro-negative side groups raise the strength of the P-N bond and increase the vibrational frequency. The OCH_2CF_3 substituted P-N asymmetric stretching band was observed at 1270 cm^{-1}.[2] The symmetric P-N-P stretching appeared at 833 cm^{-1}. P-O-C band was observed at 1416 cm^{-1}, 1163 cm^{-1}, and 955 cm^{-1}.[3] The peaks at 2972 cm^{-1} belong to the aliphatic C-H bond, at 573 cm^{-1} belong to the C-F bond, and at 1066 cm^{-1} is associated to the C-O bond stretching of the side group.[4] Additionally, the allyl peak of the polymer appears at 1650 cm^{-1}. Peaks corresponding to C-SR at 1653 cm^{-1} and C=O at 1724 cm^{-1} were observed after the thiol-ene addition reaction with the tetrakis(3-mercaptopropionate) crosslinker. No -SH peaks were detected in the region of 2600 – 2550 cm^{-1}, indicating that all thiol groups were consumed in the thiol-ene addition reaction.

DSC Spectra

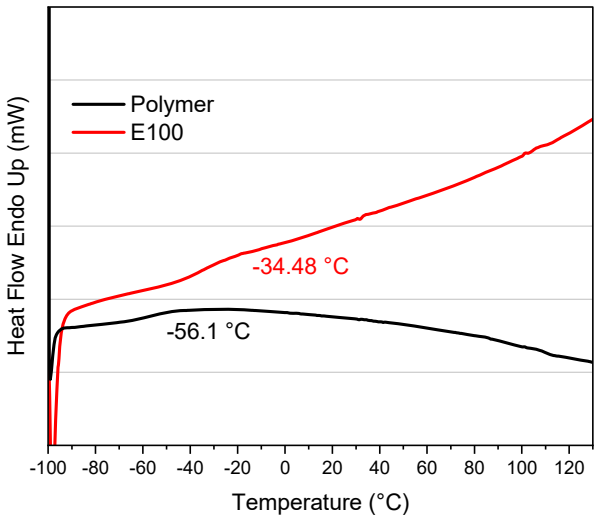

Figure S4. DSC graph of the poly[bis(2,2,2-trifluoroethoxy)]phosphazene, and **E100**.

Differential Scanning Calorimetry was employed to determine the glass transition temperature of both the polymer and the cross-linked material. Upon comparison, it was observed that the polymer exhibits a lower Tg than the cross-linked material, indicating reduced flexibility in the latter due to the presence of a network structure..

TGA Spectra

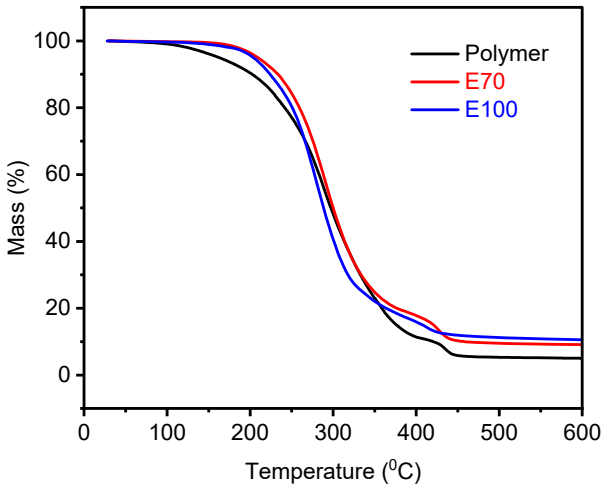

Figure S5. TGA graph of the poly[bis(2,2,2-trifluoroethoxy)]phosphazene, **E70** and **E100**.

Thermogravimetric analysis (TGA) was conducted using a Perkin Elmer TGA7 with a heating rate of 10 °C min^{-1} under a nitrogen gas flow. TGA was performed to analyze the thermal stability and decomposition behavior of the polymer and the films. The analysis revealed that polymer decomposition was initiated below 160 °C. In contrast, this occurred at elevated temperatures for the elastomers, which can be attributed to the necessity of higher temperatures to disrupt the cross-linked network. The principal weight loss was observed below 350 °C due to the materials' decomposing into cyclic species.[5,6]

Dielectric Spectra

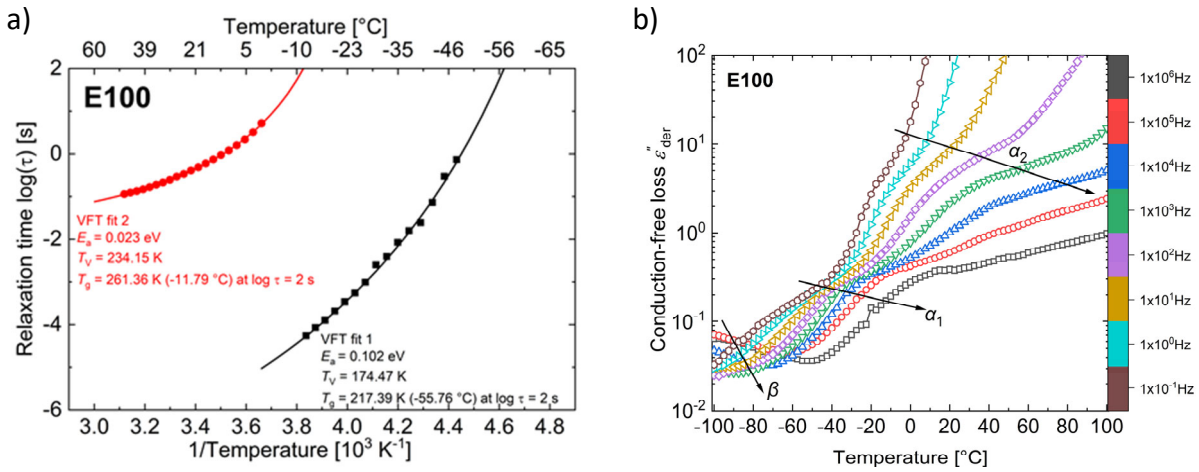

Figure S6. a) Arrhenius relaxation plot with VFT fits for **E100**, b) isochronal representation of the conduction-free dielectric loss (ε''_{der}) calculated for **E100** sample between 10^{-1} and 10^{6} Hz.

Actuation Measurements

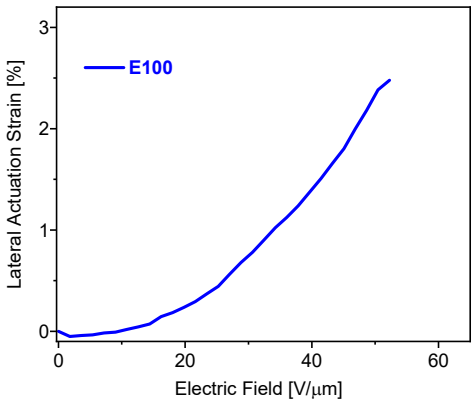

Figure S7. Lateral actuation strain of **E100** at different electric fields.

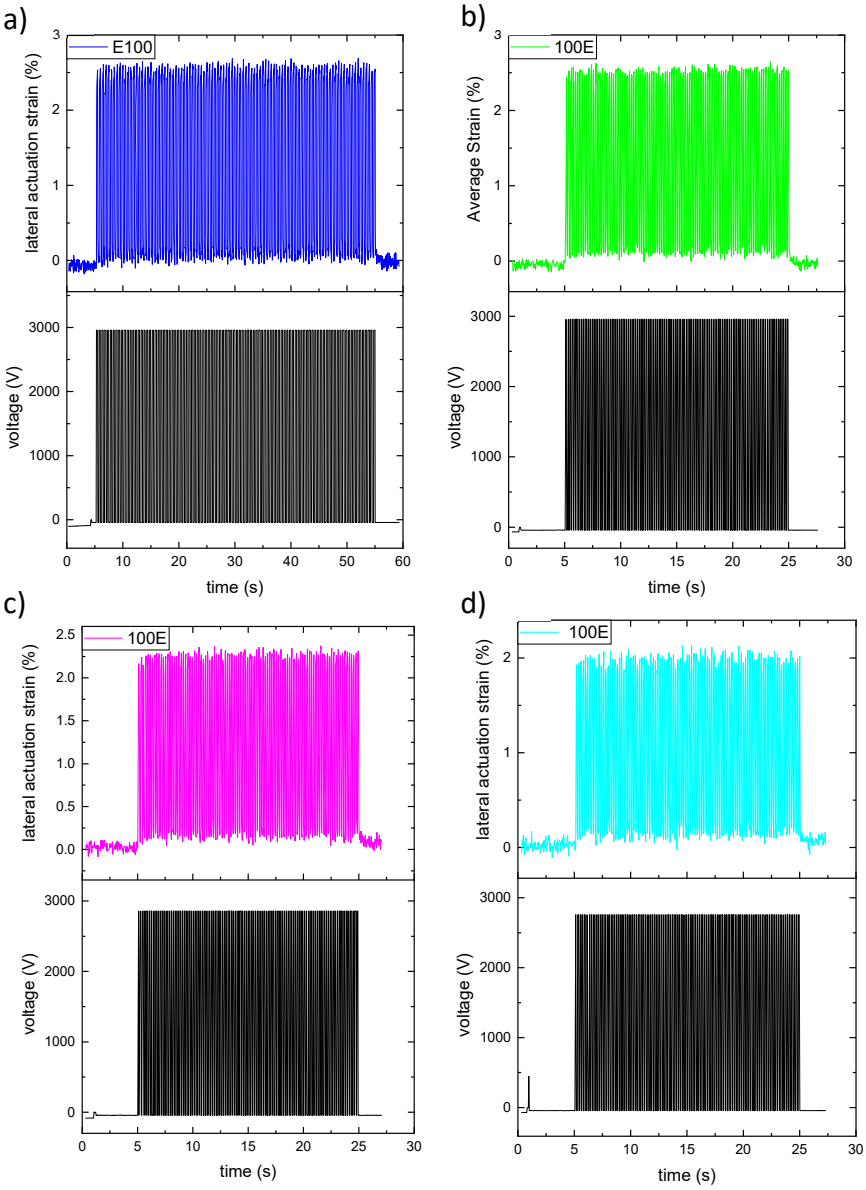

Figure S8. An actuator constructed from a 55 μm thick **E100** film exposed to an electric field of 52 V/μm and varying frequencies of 1, 2, 4, and 10 Hz for 100 cycles, respectively, exhibited stable and reversible actuation.

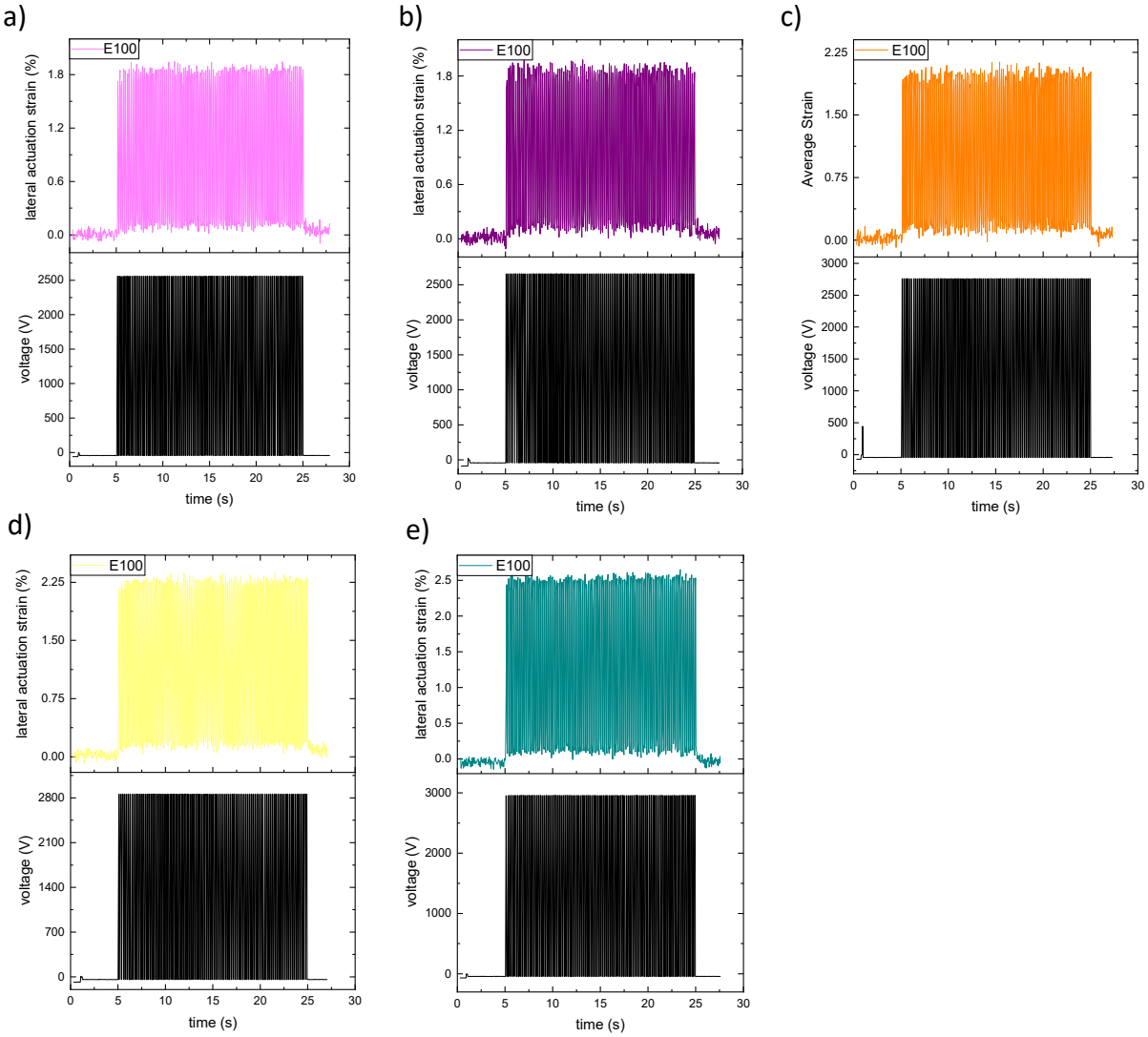

Figure S9. Film **100E**, 55 μm thick, subjected to different voltages of: a) 2600 V; b) 2700 V; c) 2800 V; d) 2900 V and e) 3000 V at 0.1 s for 100 cycle.

Weibull probability plot

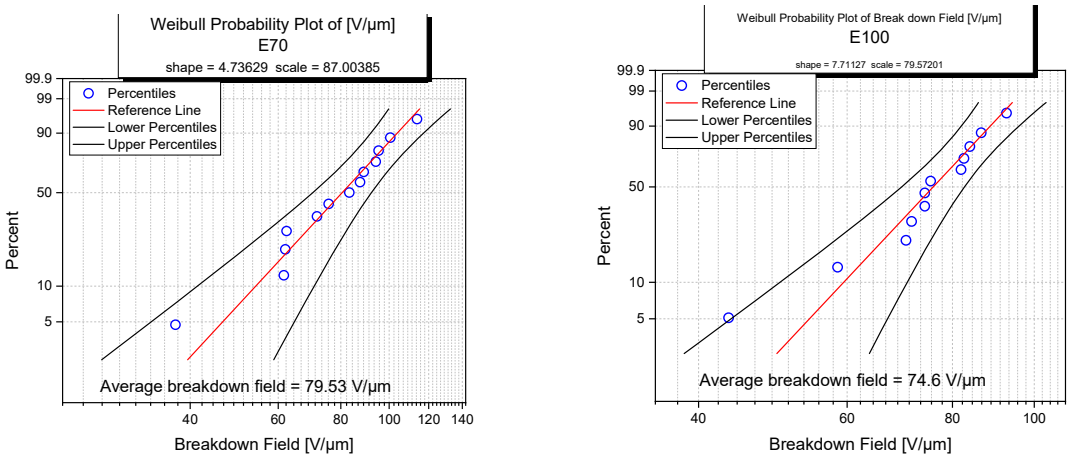

Figure S10. Weibull probability plot of a) **E70** and b) **E100**

Calculation of electrostatic pressure

The electrostatic pressure acting on the films is $p = \varepsilon'\varepsilon_0 E^2$ where p is pressure, E is electric field and, ε' and ε_0 represents relative, and free-space ($\varepsilon_0 = 8.854187817\times10^{-12}$ F/m) permittivity, respectively The p is calculated as:

0.32 MPa for **E70** (80 V μm^{-1}, $\varepsilon' = 5.65$) and

0.17 MPa for Elastosil (85 V μm^{-1}, $\varepsilon' = 3.00$).[7]

References

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