Supporting Information

Dielectric elastomer actuators with increased dielectric permittivity and low leakage current capable of overcoming electromechanical instability

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Figure S1. GPC elution curve of P0.
Figure S2. GPC analysis of P0 ($M_n = 90,000 \text{ g mol}^{-1}$, $M_w = 175,000 \text{ g mol}^{-1}$, $PDI = 1.9$).

Figure S3. $^1$H NMR spectrum of P2.
Figure S4. $^{13}$C NMR spectrum of P2.

Figure S5. $^1$H NMR spectrum of P3.

Figure S6. $^{13}$C NMR spectrum of P3.
Figure S7. $^1$H NMR spectrum of P4.

Figure S8. $^{13}$C NMR spectrum of P4.

Figure S9. $^1$H NMR spectrum of P6.
Figure S10. $^{13}$C NMR spectrum of P6.

Figure S11. $^1$H NMR spectrum of P8.

Figure S12. $^{13}$C NMR spectrum of P8.
Figure S13. $^1$H NMR spectrum of P10.

Figure S14. $^{13}$C NMR spectrum of P10.

Figure S15. $^1$H NMR spectrum of P12.
**Figure S16.** $^{13}$C NMR spectrum of P12.

**Figure S17.** DSC curves of P2.
Figure S18. DSC curves of P2.

Figure S19. DSC curves of P6.
Figure S20. DSC curves of P8.

Figure S21. DSC curves of C10.
Figure S22. DSC curves of P12.

Figure S23. Dielectric properties of Cl-CL and CN-CL. Dielectric permittivity $\varepsilon'$ was taken at $10^6$ Hz where the contribution of ions can be neglected.
Figure S24. TGA curve of P2.

Figure S25. TGA curve of P3.
Figure S26. TGA curve of E2-Cl-33.

Figure S27. TGA curve of E2-Cl-20.
Figure S28. TGA curve of E2-CN-33.

Figure S29. TGA curve of E2-CN-20.
Figure S30. TGA curve of E3-Cl-33.

Figure S31. TGA curve of E3-Cl-20.
Figure S32. TGA curve of E3-CN-33.

Figure S33. TGA curve of E3-CN-20.
Figure S34. TGA curve of Er-Cl-33.

Figure S35. TGA curve of Er-Cl-20.
Figure S36. DSC curves of E2-Cl-20.

Figure S37. DSC curves of E2-Cl-33.
Figure S38. DSC curves of E2-CN-20.

Figure S39. DSC curves of E2-CN-33.
Figure S40. DSC curves of E3-Cl-20.

Figure S41. DSC curves of E3-Cl-33.
**Figure S42.** DSC curves of E3-CN-20.

**Figure S43.** DSC curves of E3-CN-33.
Figure S44. Tensile tests of Er. Three independent tests were performed.

Figure S45. Tensile tests of E2. Three independent tests were performed.

Figure S46. Tensile tests of E3. Three independent tests were performed.
Figure S47. Tensile tests of E4. Three independent tests were performed.

Figure S48. Tensile tests of E6. Three independent tests were performed.

Figure S49. Tensile tests of E8. Three independent tests were performed.
Figure S50. Tensile tests of Er-Cl-20. Three independent tests were performed.

Figure S51. Tensile tests of Er-Cl-33. Three independent tests were performed.

Figure S52. Tensile tests of E2-Cl-33. Two independent tests were performed.
Figure S53. Tensile tests of E2-Cl-20. Three independent tests were performed.

Figure S54. Tensile tests of E3-Cl-33. Three independent tests were performed.

Figure S55. Tensile tests of E3-Cl-20. Three independent tests were performed.
Figure S56. Tensile tests of E2-CN-33. Three independent tests were performed.

Figure S57. Tensile tests of E2-CN-20. Three independent tests were performed.

Figure S58. Tensile tests of E3-CN-33. Three independent tests were performed.
Figure S59. Tensile tests of E3-CN-20. Three independent tests were performed.

Figure S60. Tensile tests of Elastosil®Film. Three independent tests were performed.

Figure S61. Lateral actuation strain of Elastosil®Film 30% prestrained measured at various frequencies (0.5-8 Hz) and an electric field of 90 V μm⁻¹.
Video 1. Circular membrane actuator constructed from **E3-Cl-20** operated for 100 cycles with 53 V μm⁻¹ at 0.5 Hz. The diameter of the film is 25 mm, the electrode diameter is 8 mm. Carbon black powder was used as electrode.

Video 2. Circular membrane actuator constructed from **E3-Cl-20** operated for 1000 cycles at 42 V μm⁻¹ at 10 Hz.

Video 3. Circular membrane actuator constructed from **E3-Cl-20** tested at different voltages. The movie was constructed by combining photos which were taken at certain time intervals.