Electronic Supplementary Material (ESI) for Journal of Materials Chemistry C. This journal is © The Royal Society of Chemistry 2017^{adjust margins}



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PAA-1 - poly(amic acid)

Scheme S1. Obtaining route of poly(amic acid).



Scheme S2. Preparation scheme of polyimide-polydimethylsiloxane copolymer containing CN groups and 50 % PDMS in the backbone.



Scheme S3. Preparation scheme of polyimide-polydimethylsiloxane copolymer containing hexafluoropropilene groups and 20 % PDMS in the backbone.

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Figure S1. SEM images of crio-fractured cross-section of: a) Si-Blank, b) Elastosil, c) Si-1-2%, d) Si-1-5%, e) Si-2-2%, f) Si-2-5%, g) Si-2-10%, h) Si-3-2%, i) Si-3-5% and j) Si-3-10%.



Figure S2. EDAX mapping of Si-2-10% and Si-3-10% cryo-fracture section.



Figure S3. Topography (left) and phase (right) AFM images of Si-2-10% and Si-3-10%.



Figure S4. Dependence of loss factor vs. temperature for Si-Blank and Si-2-5%.





Figure S5. Temperature dependence of: a), b) loss factor and c) storage modulus for Si-Blank and SI-2-5%.



Figure S6. Dielectric losses as a function of temperature for Si-Blank and samples which contains 5% polyamic acid/polyimide.







Figure S8. Pellets of polyamic acid/polyimides before (top) and after (bottom) breakdown tests.



Figure S9. Dielectric loss in dependence on frequency for: a)Elastosil; b) PAA-1 series; c) PICOSI-1 series and d) PICOSI-2 series.



Figure S10. Voltage signal used for actuation measurements.

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Figure S11. Example of electrical breakdown during actuation tests.



Figure S12. FT-IR spectra of: a) polyamic acid, b) polyimide containing CN groups and c) polyimide containing hexafluoropropylidene groups.

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Figure **\$13.** ¹H-NMR spectrum of polyamic acid.

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Figure S14. ¹H-NMR spectrum of polyimide containing CN groups.





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Image S1. Electrical breakdown setup.



Image S2. Equiaxial stretching setup.

Table S1. The calculated maximum energy converted.

Sample	Energy gain ^{a)} , mJ·cm ⁻³	Energy gain ^{b)} , mJ·cm ⁻³
Elastosil	119.5	95.2
Si-Blank	18.4	13.9
Si-1-2%	27.9	21.1
Si-1-5%	51.2	39.2
Si-1-10%	-	-
Si-2-2%	39.4	30.0
Si-2-5%	73.2	55.2
Si-2-10%	129.8	100.3
Si-3-2%	46.2	35.2
Si-3-5%	76.1	60.6
Si-3-10%	164.5	132.7

 $^{a)}\text{eps'}$ @ 1Hz, $\text{E}_{\text{max}}\text{=}\text{Ebd}$ and $\text{s}_{\text{max}}\text{=}\text{maximum}$ strain;

 $^{\text{b})}\text{eps'}$ @ 1Hz, $\text{E}_{\text{max}}\text{=}\text{Ebd}$ and $\text{s}_{\text{max}}\text{=}100\%.$