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Silicone dielectric elastomers optimized by crosslinking pattern - a simple approach to high-performance actuators.

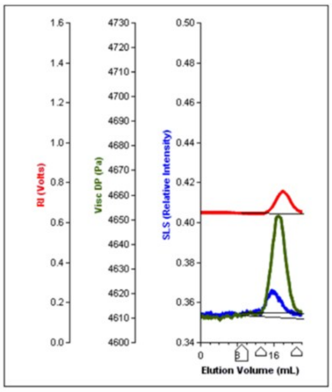
Codrin Tugui[a]*, George-Theodor Stiubianu[a], Manole-Stelian Serbulea[b,c], Maria Cazacu[a]

[a]Inorganic Polymers Department, Petru Poni Institute of Macromolecular Chemistry, Aleea Grigore Ghica Voda 41A, 700487, Iasi, Romania

[b]Technical University of Civil Engineering Bucharest, Soil Mechanics and Foundation Engineering Department, bd. Lacul Tei 122, sector 2, 022396, Bucharest, Romania

[c]Geotechnical Expert S.R.L., str. Fabrica de Gheață 16-18, ap. 55, sector 2, 022540, Bucharest, Romania

Email: tugui.codrin@icmpp.ro



Polymer A1

M_n	2.356e+04	g/mol
M_w	5.543e+04	g/mol
M_z	1.147e+05	g/mol
M_p	3.822e+04	g/mol
M_w/M_n	2.353	
M_z/M_w	2.069	
$<M_{10}$	2.734e+04	g/mol
$>M_{90}$	2.501e+05	g/mol
V_p	17.73	mL
$Area_{RI}$	2.450e+00	V*mL
Concentration	undetermined	mg/mL
dn/dc	0.8223	mL/g
Sample Recovery	undetermined	%
[η]	8.732e+02	mL/g

Polymer A2

M_n	1.816e+05	g/mol
M_w	2.972e+05	g/mol
M_z	5.102e+05	g/mol
M_p	2.147e+05	g/mol
M_w/M_n	1.636	
M_z/M_w	1.717	
$<M_{10}$	1.488e+05	g/mol
$>M_{90}$	1.032e+06	g/mol
V_p	15.67	mL
$Area_{RI}$	2.209e-01	V*mL
Concentration	undetermined	mg/mL
dn/dc	0.0741	mL/g
Sample Recovery	undetermined	%
[η]	1.717e+02	mL/g

Polymer A3

M_n	3.708e+05	g/mol
M_w	6.418e+05	g/mol
M_z	1.028e+06	g/mol
M_p	5.287e+05	g/mol
M_w/M_n	1.731	
M_z/M_w	1.602	
$<M_{10}$	3.420e+05	g/mol
$>M_{90}$	1.988e+06	g/mol
V_p	14.70	mL
$Area_{RI}$	2.853e-01	V*mL
Concentration	undetermined	mg/mL
dn/dc	0.0270	mL/g
Sample Recovery	undetermined	%
[η]	1.510e+02	mL/g

Figure S1. Molecular weight of A1, A2 and A3 determined by GPC.

Polymer B1

M_n	8.073e+04	g/mol
M_w	1.620e+05	g/mol
M_z	2.765e+05	g/mol
M_p	1.405e+05	g/mol
M_w/M_n	2.006	
M_z/M_w	1.707	
$<M_{10}$	8.625e+04	g/mol
$>M_{90}$	5.491e+05	g/mol
V_p	16.15	mL
$Area_{RI}$	1.660e-01	V*mL
Concentration	undetermined	mg/mL
dn/dc	undetermined	mL/g
Sample Recovery	undetermined	%
[η]	1.837e+02	mL/g

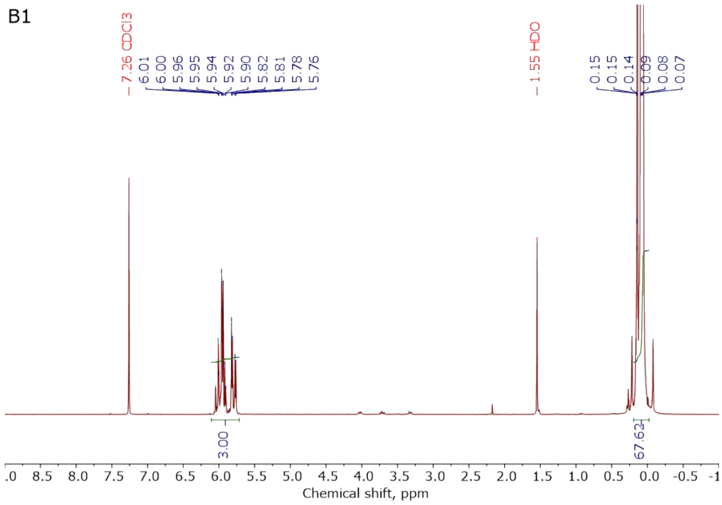


Figure S2. GPC data and NMR spectrum of polymer B1.

Polymer B2

M_n	1.557e+05	g/mol
M_w	3.456e+05	g/mol
M_z	5.889e+05	g/mol
M_p	3.186e+05	g/mol
M_w/M_n	2.220	
M_z/M_w	1.704	
$<M_{10}$	1.872e+05	g/mol
$>M_{90}$	1.150e+06	g/mol
V_p	15.23	mL
$Area_{RI}$	1.537e-01	V*mL
Concentration	undetermined	mg/mL
dn/dc	undetermined	mL/g
Sample Recovery	undetermined	%
[η]	2.617e+02	mL/g

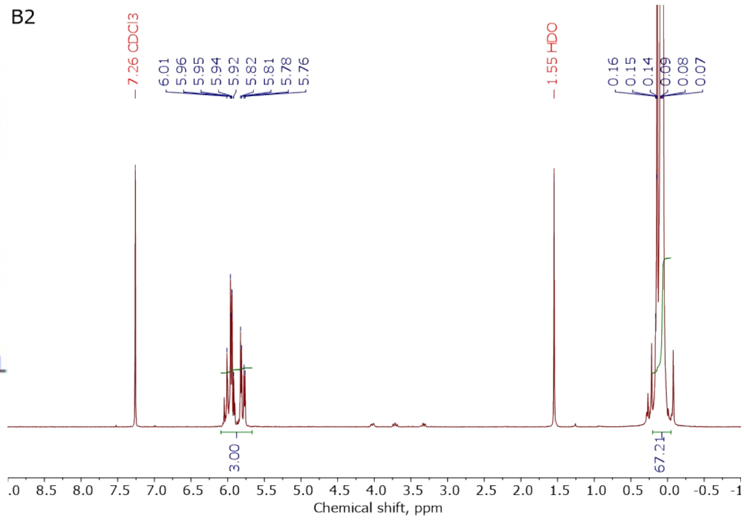


Figure S3. GPC data and NMR spectrum of polymer B2.

Polymer B3

M_n	2.647e+05	g/mol
M_w	5.422e+05	g/mol
M_z	8.904e+05	g/mol
M_p	5.120e+05	g/mol
M_w/M_n	2.048	
M_z/M_w	1.642	
$<M_{10}$	2.980e+05	g/mol
$>M_{90}$	1.703e+06	g/mol
V_p	14.73	mL
$Area_{RI}$	1.540e-01	V*mL
Concentration	undetermined	mg/mL
dn/dc	undetermined	mL/g
Sample Recovery	undetermined	%
[η]	4.103e+02	mL/g

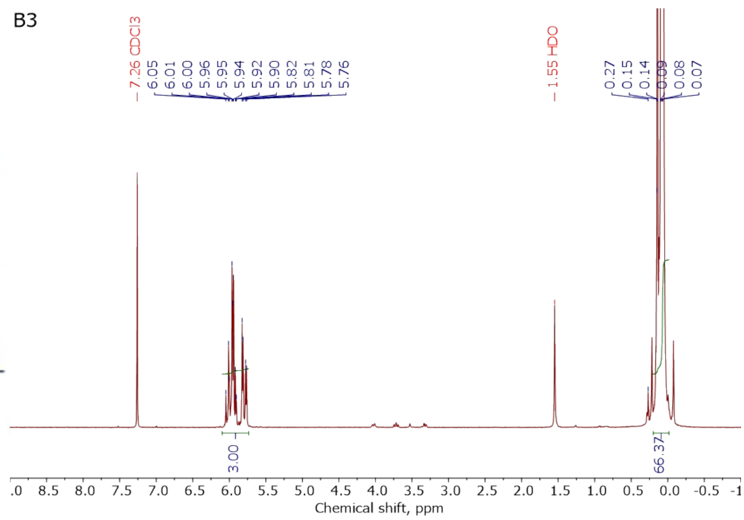


Figure S4. GPC data and NMR spectrum of polymer B3.

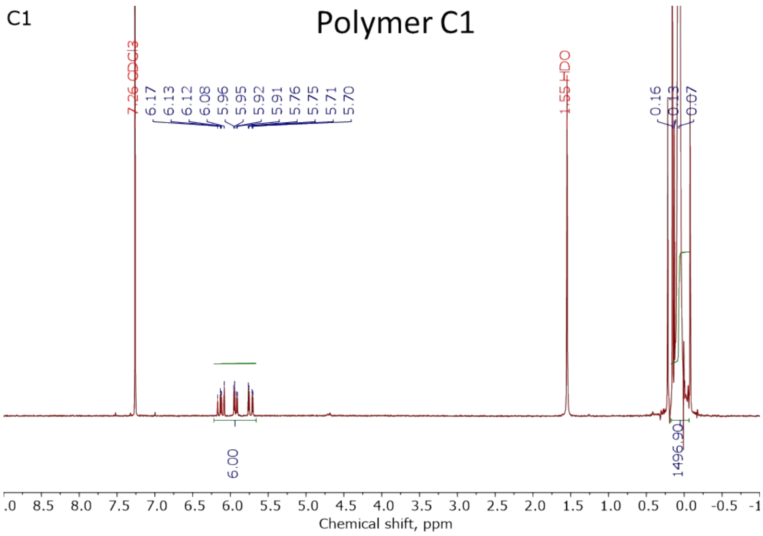


Figure S5. NMR spectrum of polymer C1.

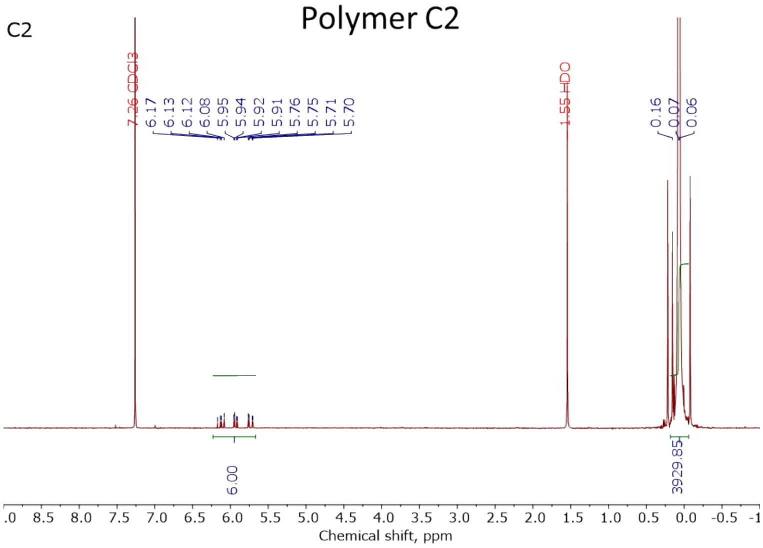


Figure S6. NMR spectrum of polymer C2.

Polymer X1

M_n	1.865e+04	g/mol
M_w	5.527e+04	g/mol
M_z	1.446e+05	g/mol
M_p	3.213e+04	g/mol
M_w/M_n	2.964	
M_z/M_w	2.617	
$<M_{10}$	2.458e+04	g/mol
$>M_{90}$	3.299e+05	g/mol
V_p	17.87	mL
$Area_{RI}$	4.174e-01	V*mL
Concentration	undetermined	mg/mL
dn/dc	0.0383	mL/g
Sample Recovery	undetermined	%
[η]	2.219e+01	mL/g

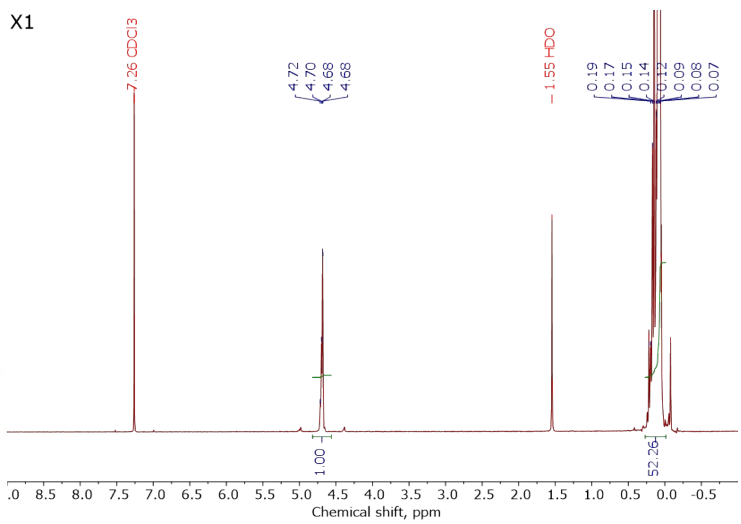


Figure S7. GPC data and NMR spectrum of polymer X1.

Polymer X3

M_n	5.733e+04	g/mol
M_w	1.593e+05	g/mol
M_z	4.390e+05	g/mol
M_p	8.580e+04	g/mol
M_w/M_n	2.778	
M_z/M_w	2.756	
$<M_{10}$	7.407e+04	g/mol
$>M_{90}$	1.094e+06	g/mol
V_p	16.73	mL
$Area_{RI}$	2.852e-01	V*mL
Concentration	undetermined	mg/mL
dn/dc	0.0154	mL/g
Sample Recovery	undetermined	%
[η]	3.063e+01	mL/g

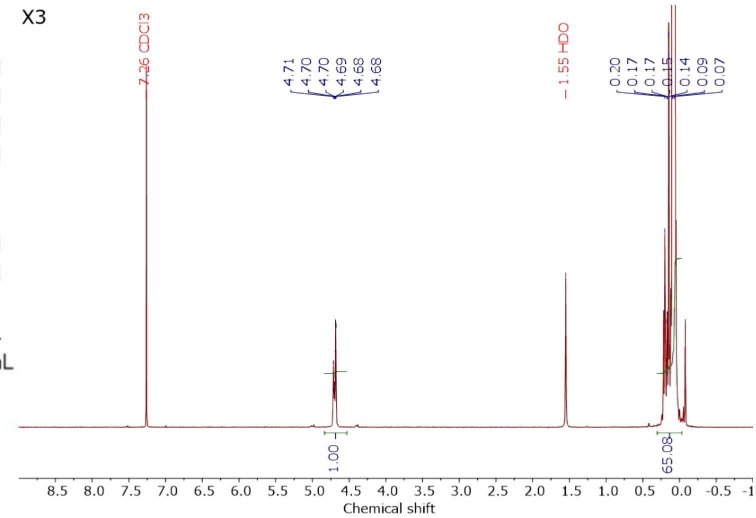


Figure S8. GPC data and NMR spectrum of polymer X3.

Polymer X2

M_n	3.584e+04	g/mol
M_w	8.435e+04	g/mol
M_z	2.104e+05	g/mol
M_p	4.725e+04	g/mol
M_w/M_n	2.354	
M_z/M_w	2.494	
$<M_{10}$	3.914e+04	g/mol
$>M_{90}$	5.165e+05	g/mol
V_p	17.47	mL
$Area_{RI}$	2.769e-01	V*mL
Concentration	undetermined	mg/mL
dn/dc	undetermined	mL/g
Sample Recovery	undetermined	%
[η]	5.194e+02	mL/g

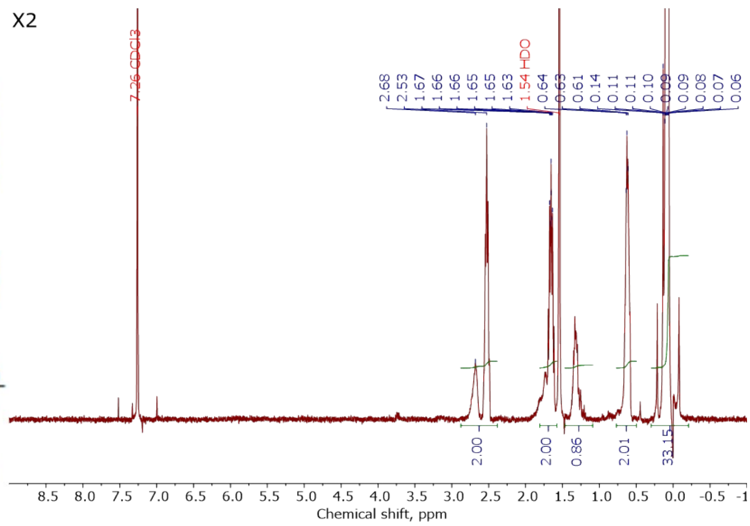


Figure S9. GPC data and NMR spectrum of polymer X2.

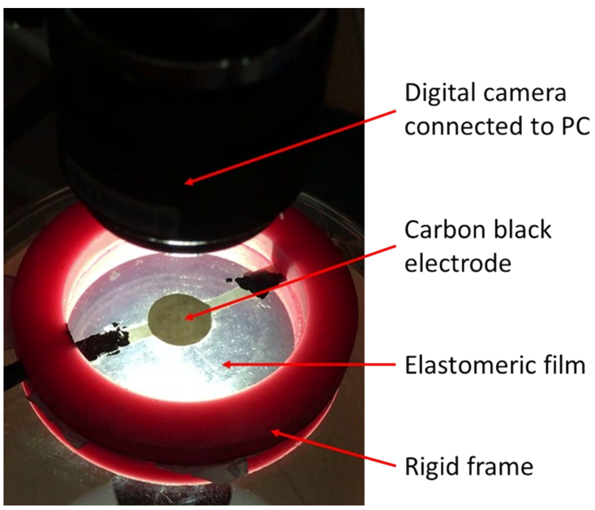


Figure S10. The actuator configuration for lateral strain measurements.

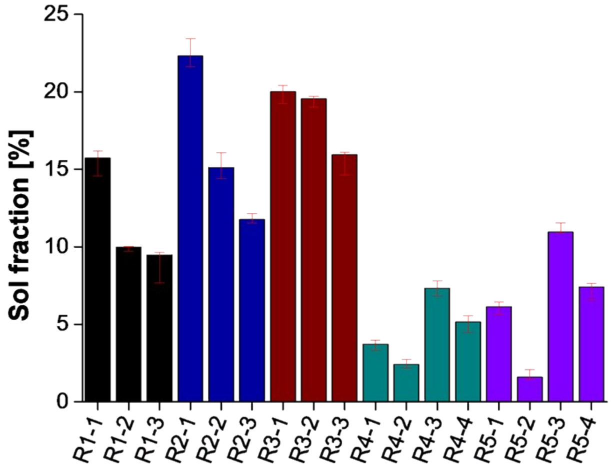


Figure S11. The amount of soluble fraction extracted from films.

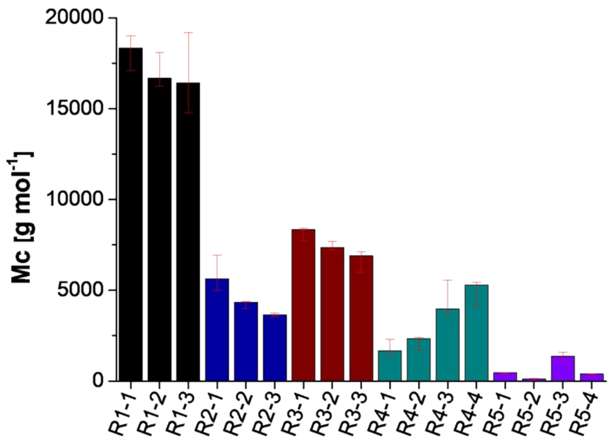


Figure S12. The average molecular weight of polymer chains between two consecutive junctions.

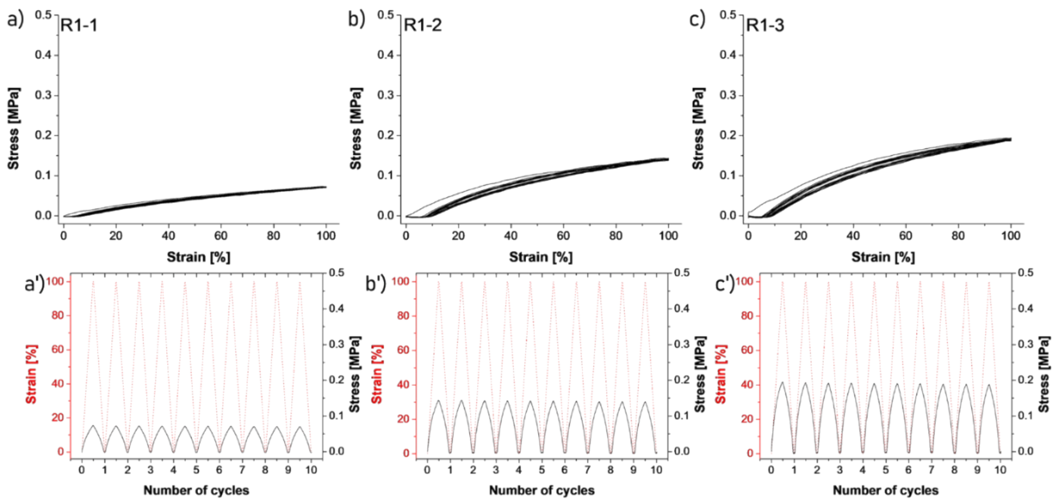


Figure S13. Cyclic stress-strain curves (top) and the corresponding stress relaxation (bottom) for Series R1.

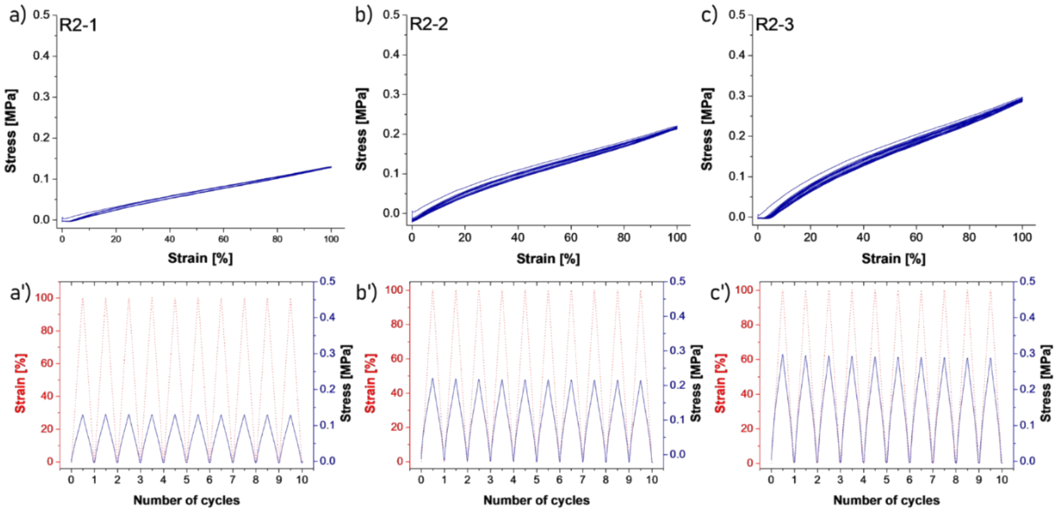


Figure S14. Cyclic stress-strain curves (top) and the corresponding stress relaxation (bottom) for Series R2.

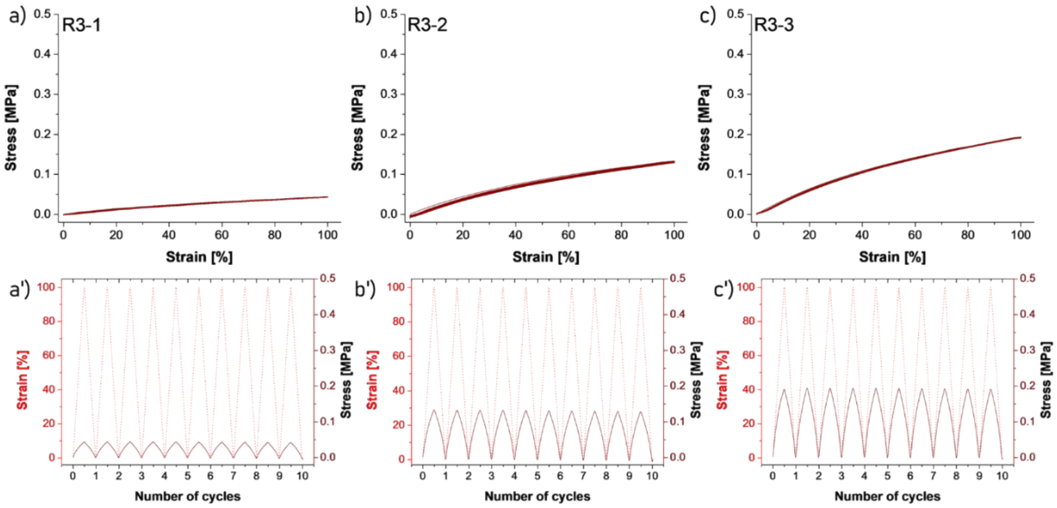


Figure S15. Cyclic stress-strain curves (top) and the corresponding stress relaxation (bottom) for Series R3.

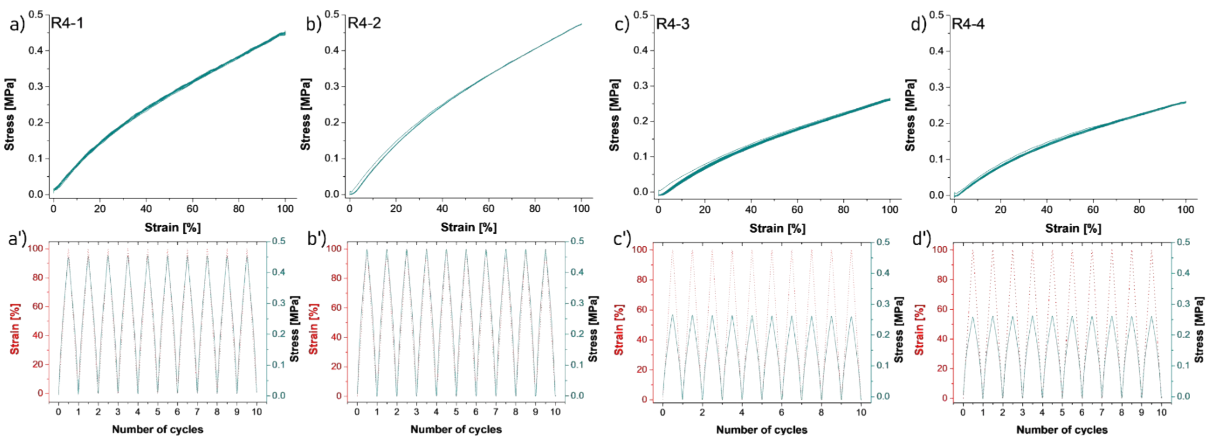


Figure S16. Cyclic stress-strain curves (top) and the corresponding stress relaxation (bottom) for Series R4.

In actuation mode, when an electric field is applied on film made of dielectric material, the Maxwell pressure generated between the two electrodes is given by:

$$\boldsymbol{p} = \boldsymbol{\varepsilon\varepsilon}_0 \boldsymbol{E}^2 \tag{1}$$

where p is the electrostatic pressure across the electrodes, ε is the relative permittivity, ε_0 is the permittivity of the free space and E^2 represents the electric fields applied on electrodes. If we use the linear-elasticity and boundary approximations, the actuation strain in z direction (thickness) is given by:

$$\boldsymbol{s}_z = -\frac{\boldsymbol{p}}{\boldsymbol{Y}} \tag{2}$$

where Y represents the Young's modulus.

Considering the elastomers are incompressible $At=const$, where A represents the area of the electrodes, t is the thickness of the dielectric film placed between electrodes, thereby it is possible to determine the actuated thickness of the elastomer t_1:

$$\boldsymbol{t}_1 = \boldsymbol{t}_0 - (\boldsymbol{s}_z \cdot \boldsymbol{t}_0) \tag{3}$$

where t_0 is the thickness of initial unactuated elastomer.

The actuated diameter of the electrodes will be:

$$\boldsymbol{d}_1 = \sqrt{\frac{\boldsymbol{d}_0^2 \cdot \boldsymbol{t}_0}{\boldsymbol{t}_1}} \tag{4}$$

where d_0 and d_1 are the initial and the final diameter of the circular electrode.

The theoretical in plane actuation (lateral actuation) s_x was calculated with the formula:

$$\boldsymbol{s}_x = \frac{\boldsymbol{d}_1 - \boldsymbol{d}_0}{\boldsymbol{d}_0} \cdot 100 \tag{5}$$

We substitute equation (1) in equation (2), then substitute equation (2) in (3), then we substitute (4) in equation (5) and we obtain the equation mentioned in the paper (numbered as equation 6):

$$\boldsymbol{s}_x = \frac{\sqrt{\dfrac{\boldsymbol{d}_0^2 \cdot \boldsymbol{t}_0}{\boldsymbol{t}_0 - \left(\dfrac{\boldsymbol{\varepsilon} \cdot \boldsymbol{\varepsilon}_0 \cdot \boldsymbol{E}^2}{\boldsymbol{Y}} \cdot \boldsymbol{t}_0\right)}} - \boldsymbol{d}_0}{\boldsymbol{d}_0} \cdot 100$$

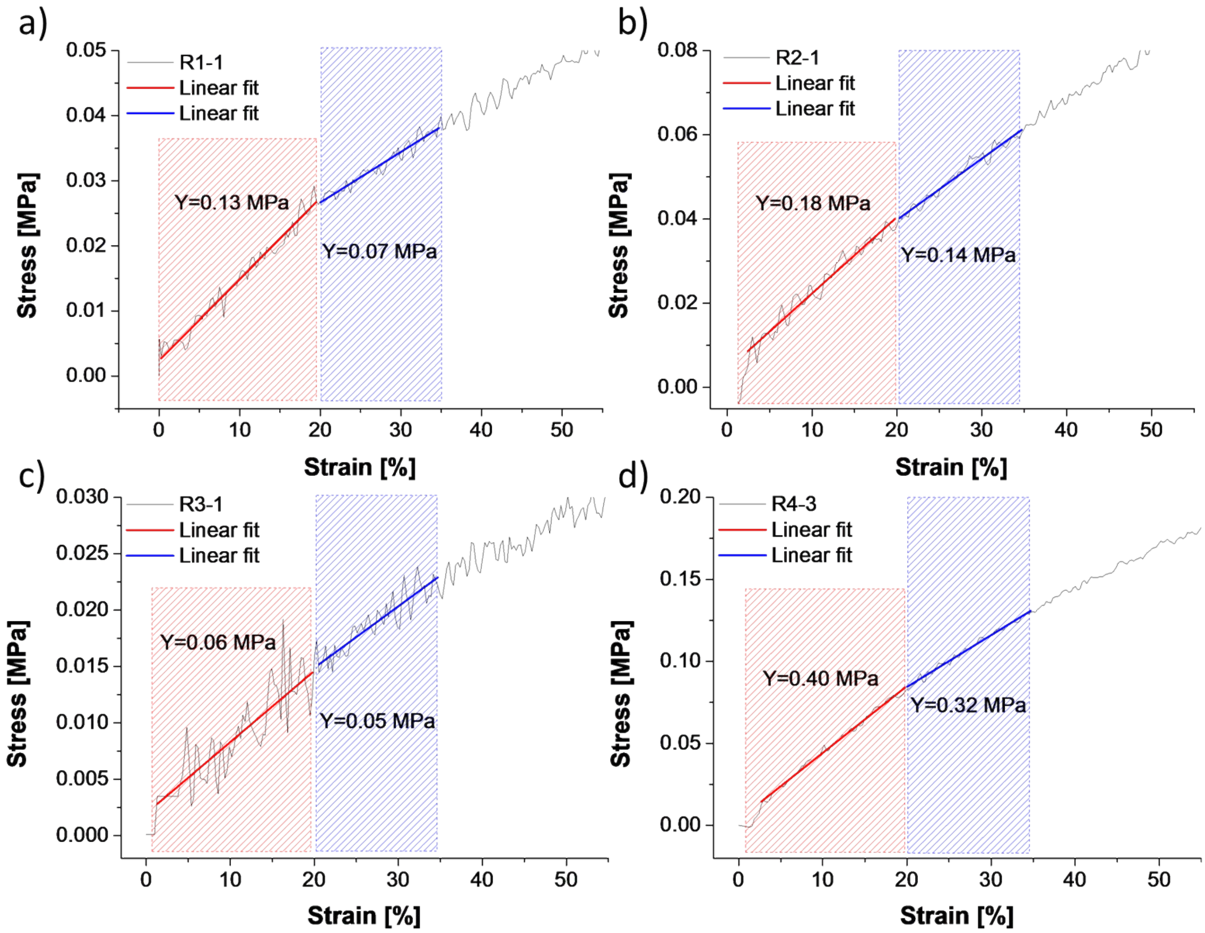


Figure S17. Young's modulus values of R1-1, R2-1, R3-1 and R4-3 determined from stress-strain curves within 0 - 20% and 20 - 35%, strain, respectively.