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Supplementary Materials

Binary Silicone Elastomeric Systems with Stepwise Crosslinking as a Tool for Tuning Electromechanical Behavior

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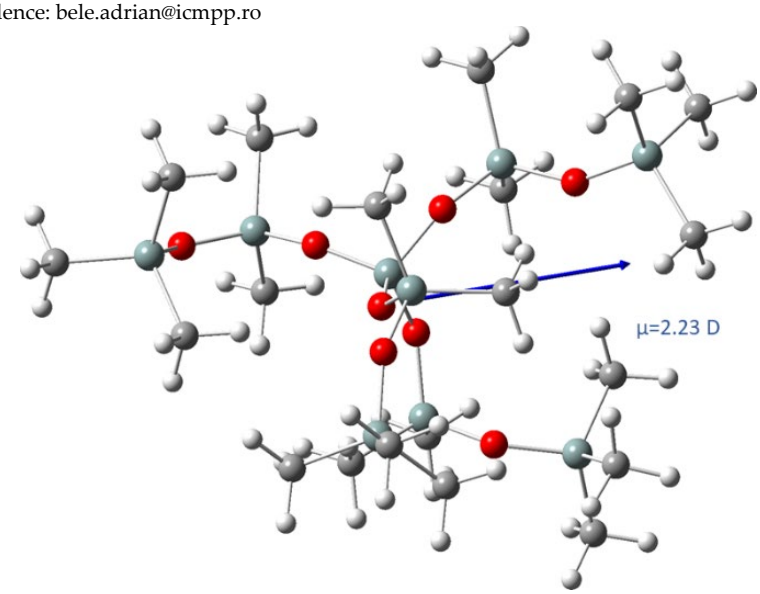

Figure S1. Representation of the equilibrium geometry and dipole moment orientation for structure A into the ground state. Theoretical calculations performed with the LC-wPBE/6-31G(d,p) method.

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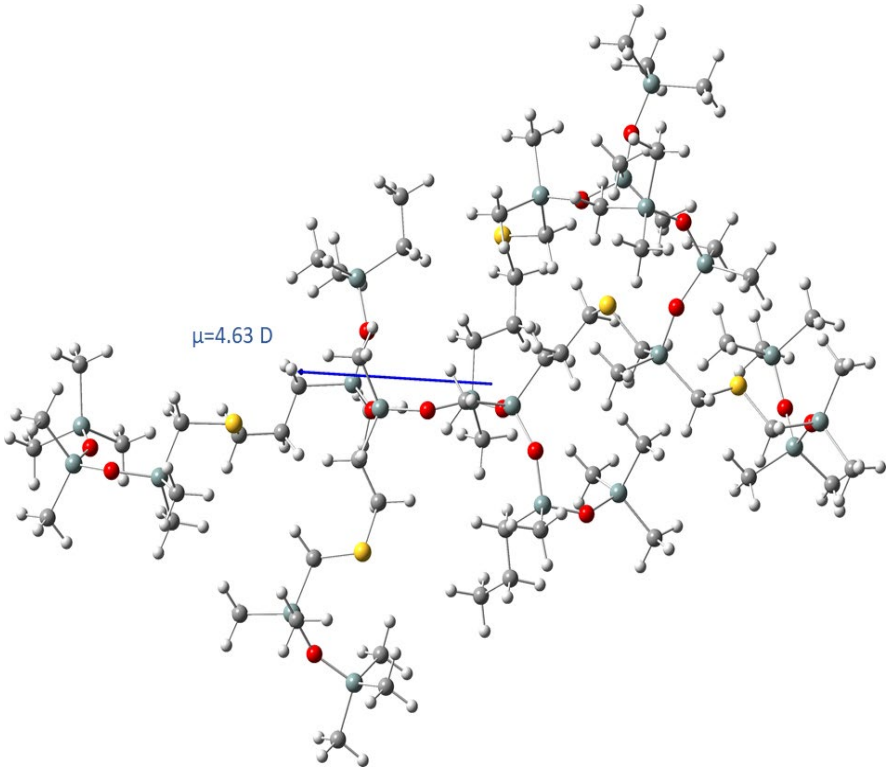

Figure S2. Representation of the equilibrium geometry and dipole moment orientation in the case of the structure B (or C) into the ground state. Theoretical calculations performed with the LC-WPBE/6-31G(d,p) method.

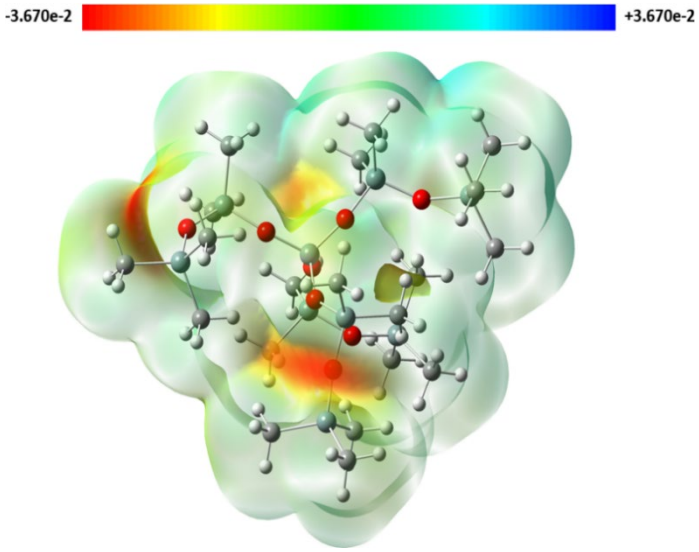

Figure S3. Electrostatic potential rendered as a mapped surface in the vicinity of molecule A (computation done at LC-wPBE/6-31G(d,p) level of theory).

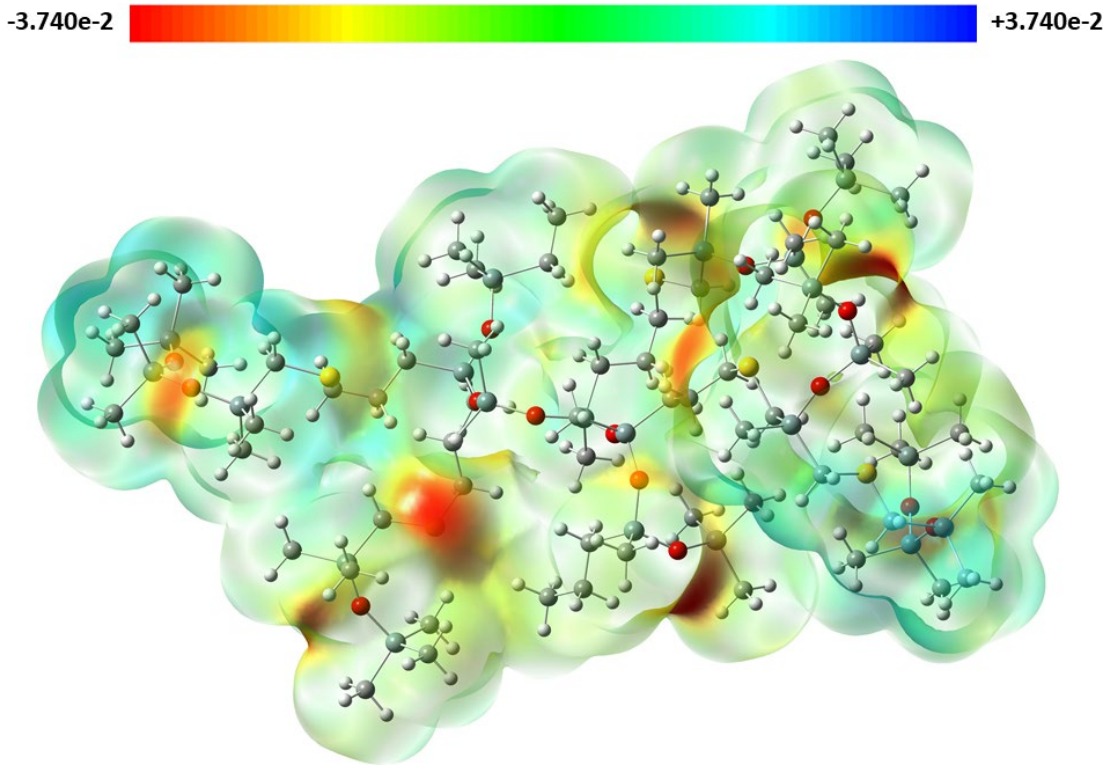

Figure S4. Electrostatic potential rendered as a mapped surface in the vicinity of molecule A (computation done at LC-wPBE/6-31G(d,p) level of theory).

Table S1. The amounts used for obtaining the IPNs and the reference samples

Reagent/Sample	A		B		C		A1B1		A2B1		A1B2		A1C1		A2C1		A1C2	
	g	mmol	g	mmol	g	mmol	g	mmol	g	mmol	g	mmol	g	mmol	g	mmol	g	mmol
FD 80	2	0.028					2	0.028	2	0.028	1	0.014	2	0.028	2	0.028	1	0.014
DMSV31			2	0.071			2	0.071	1	0.014	2	0.071						
DMSV22					2	0.21							2	0.21	1	0.106	2	0.21
TEOS	0.93	4.48					0.93	4.48	0.93	4.48	0.466	2.236	0.93	4.48	0.93	4.48	0.466	2.236
DBTDL	0.01	0.016					0.01	0.016	0.01	0.016	0.005	0.008	0.01	0.016	0.01	0.016	0.005	0.008
GP-367			0.176	0.048	0.258	0.071	0.176	0.048	0.088	0.024	0.176	0.048	0.258	0.071	0.129	0.035	0.258	0.071
DMPA			0.108	0.42	0.113	0.44	0.108	0.42	0.054	0.21	0.108	0.42	0.113	0.44	0.056	0.218	0.113	0.44

Table S2. Thermal characteristics extracted from DSC data

Sample	T_{g1} (°C)	T_{g2} (°C)	T_{m1} (°C)	ΔH_{m1} (J/g)	T_{m2} (°C)	ΔH_{m2} (J/g)	T_{cr} (°C)	ΔH_{cr} (J/g)	C_p (J/g °C)	$\chi_{PDMS/IPN}$ (%)
PDMS	– 122	– 120	– 42	23.46	– 42	24.55	– 68	– 24.34	0.067	0.40
A	– 123	– 123	– 42	24.73	– 42	24.67	– 70	– 24.24	0.074	0.40
B	– 119	– 119	– 45	21.52	– 45	21.15	– 81	– 22.27	0.111	0.34
C	– 121	– 121	– 52	16.25	– 52	16.81	– 94	– 10.42	0.115	0.27
A1B1	– 121	– 121	– 43	19.36	– 42	20.34	– 70	– 21.17	0.151	0.33
A2B1	– 122	– 122	– 43	24.55	– 42	26.86	– 70	– 24.86	0.108	0.40
A1B2	– 120	– 121	– 43	17.35	– 43	17.98	– 73	– 17.7	0.079	0.29
A1C1	– 119	– 119	– 43	12.53	– 43	12.55	– 70	– 14.12	0.083	0.20
A2C1	– 120	– 120	– 43	24.29	– 43	24.9	– 70	– 22.7	0.086	0.40
A1C2	– 117	– 117	– 43	17.84	– 43	17.78	– 71	– 17.5	0.096	0.29

T_{g1} – glass transition temperature corresponding to the first heating run; T_{g2} – glass transition temperature corresponding to the second heating run; T_{m1} – melting temperature corresponding to the first heating run; T_{m2} – melting temperature corresponding to the second heating run; ΔH_{m1} – enthalpy of the melting profile corresponding to the first heating run; ΔH_{m2} – enthalpy of the melting profile corresponding to the second heating run; T_{cc1} – cold crystallization temperature corresponding to the first heating run; T_{cc2} – cold crystallization temperature corresponding to the second heating run; ΔH_{cc1} – enthalpy of the cold crystallization profile corresponding to the first heating run; ΔH_{cc2} – enthalpy of the cold crystallization profile corresponding to the second heating run; ΔH_{cr} – enthalpy of the crystallization profile; C_p – heat capacity; $\chi_{PDMS/IPN}$ – degree of PDMS crystallinity in the networks.

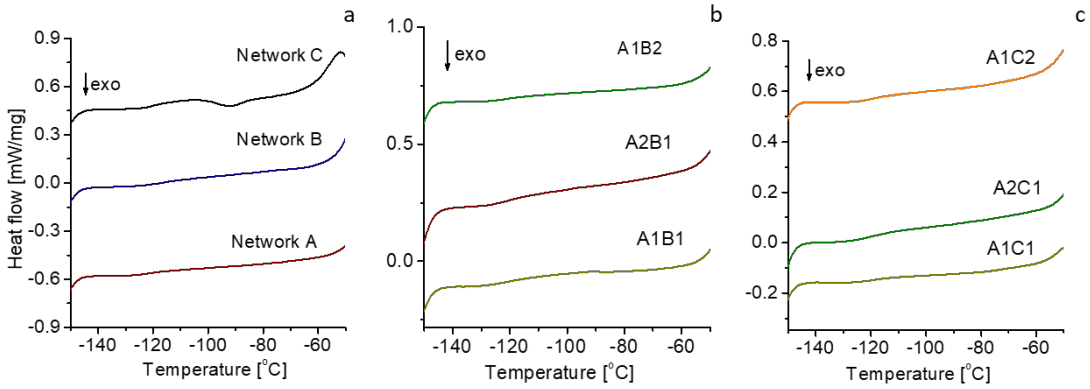

Figure S5. Inset of DSC data emphasizing the glass transition temperature.

Table S3. DMA data of reference sample A and AxCy series

Sample	E′ (Pa)	T_g		T_m (°C)
		tan δ	T(°C)	
A	$2.94x10^8$	0.101	-118.08	-36.13
A2C1	$5.75x10^8$	0.112	-112.9	-35.5
A1C1	$1.44\ x10^9$	0.105	-115.1	-36.2
A1C2	$2.06x10^9$	0.085	-113.93	-38.4

Table S4. Mechanical and dielectric data of prepared samples in comparison with representative IPNs from literature.

Sample	Sm (%)	Tnm (MPa)	Y (MPa)	ε'	Ebd (V/μm)	UTT, (kJ/m³)	ΔW/V (mJ/cm³)
A	256	0.31	0.42	3.25	41	4.50	8.23
B	325	0.07	0.22	3.00	39	1.30	7.79
C	100	0.15	0.41	3.17	39	0.90	6.54
A1B1	267	0.70	1.27	4.08	40	11.00	10.39
A2B1	270	0.41	0.42	3.72	63	5.70	9.48
A1B2	247	0.81	0.61	3.71	25	11.20	9.36
A1C1	520	1.00	0.60	3.66	35	29.10	9.80
A2C1	720	1.50	1.00	3.86	47	63.00	10.46
A1C2	320	0.60	0.45	3.97	46	11.40	10.30
Elastosil 3060	310	3.15	1.24	2.58	90	42.70	6.67
ref 14 (sample 15wt%)	600	3.10	-*	-*	45	-**	-*
ref 15 (sample IPN-P1)	484	0.30	0.27	2.92	60	-**	9.00
ref 15 (sample IPN-F2)	512	0.35	0.11	3.01	12	-**	1.90***
ref 15 (sample IPN-CN1)	486	0.27	0.03	3.62	15	-**	3.60***
ref 15 (sample IPN-F2)	692	0.32	0.13	2.93	29	-**	8.10
ref 16 (sample Si_A_IN10)	560	3.30	0.26	3.30	55	-**	8.90
ref 16 (sample Si_B_IN10)	670	2.70	0.22	3.80	53	-**	10.1

Sm – Tensile strain (values given at break); Tnm – Tensile stress (values given at break); Y – Young's modulus (calculated at 5 % Sm); ε' – Dielectric permittivity (values given at 10^3 Hz); UTT – Ultimate tensile toughness; ΔW/V - Energy output at 25 V/μm; * - values not available; ** - no values calculated due to the fact that raw mechanical data is needed in order to make calculations; *** - calculated at maximum Ebd provided by the authors.