Electronic Supplementary Material (ESI) for Materials Chemistry Frontiers.

Supporting Information for

The synergistic effect of nitrile and jeffamine structural elements towards stretchable and high-*k* neat polyimide materials

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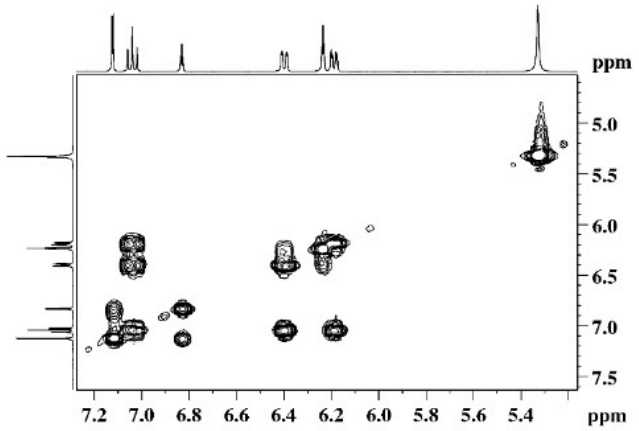
M2

(a)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6.0
6.5
7.0
7.5
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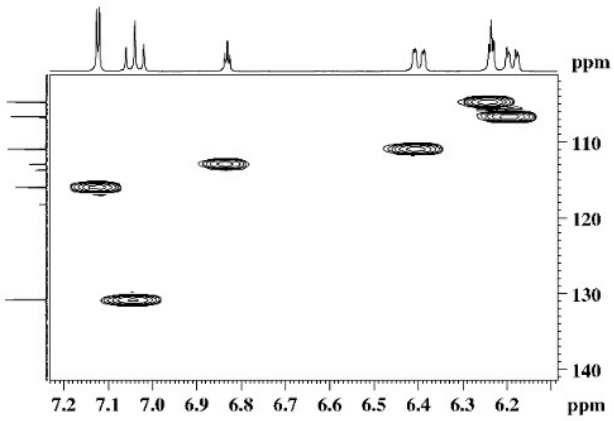
ppm

(b)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130
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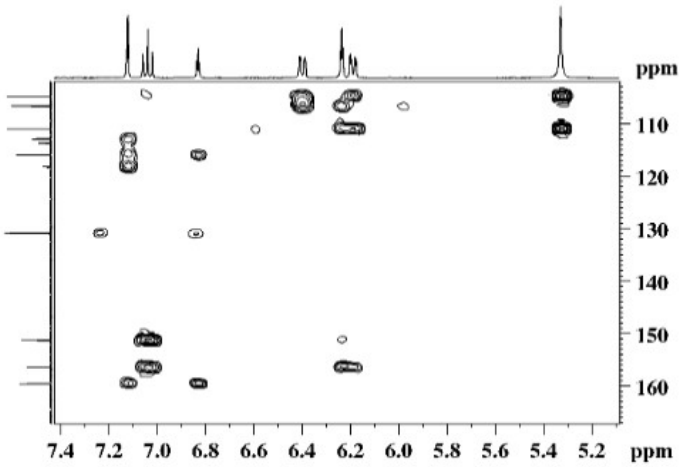
ppm

(c)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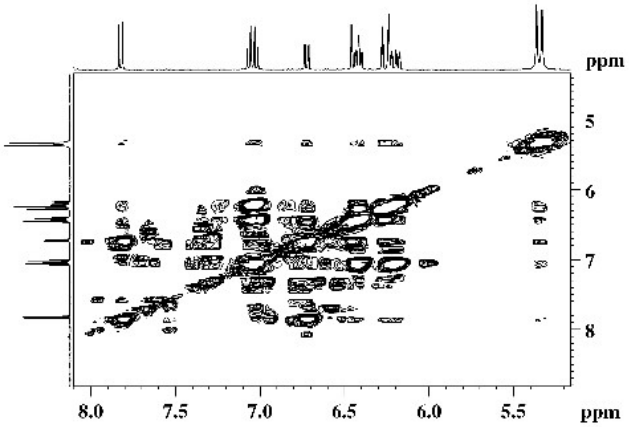
ppm

M3

(a)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7
8
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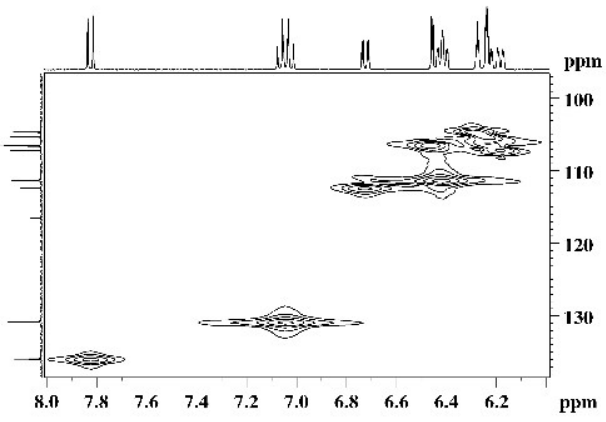
(b)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ppm

(c)

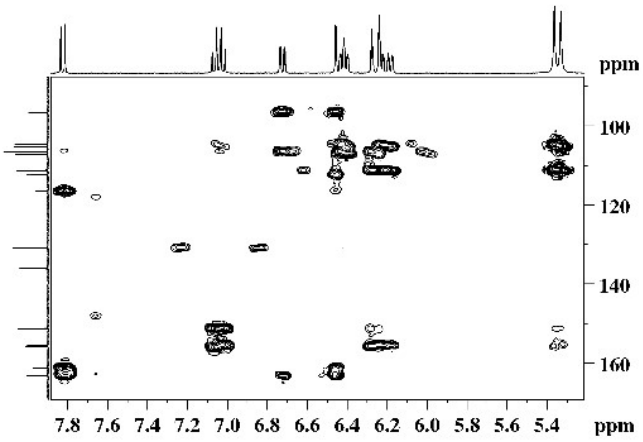
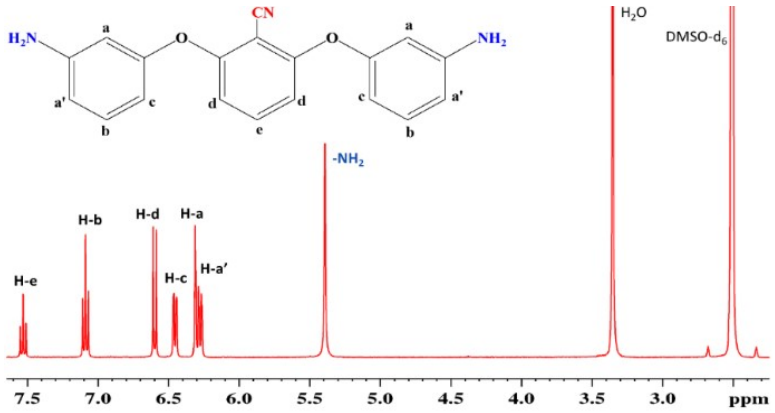


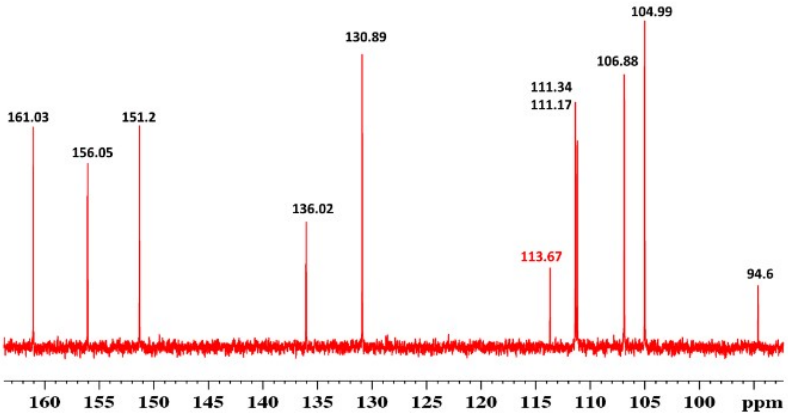
Figure S1. 2D NMR spectra of diamines **M2** and **M3**: (a) H,H-COSY; (b) H,C- HSQC; (c) H,C-HMBC.

M1

(a)

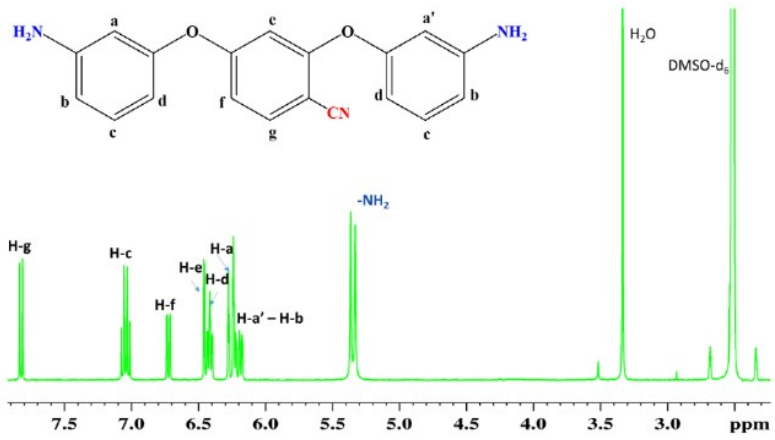


(b)



M3

(a)



(b)

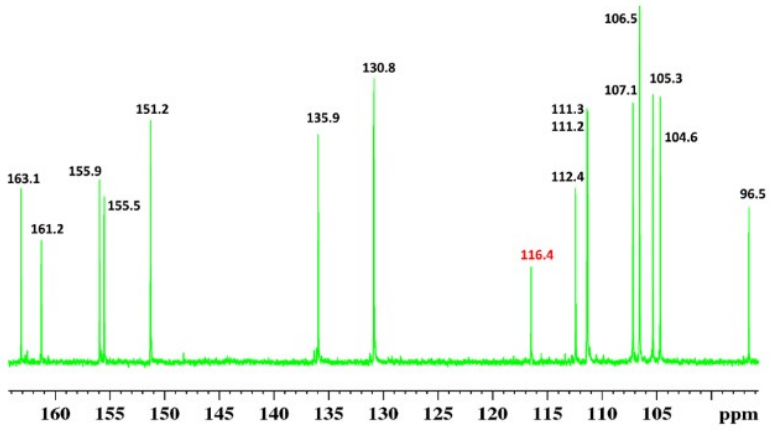


Figure S2. NMR spectra of diamines **M1** and **M3**: (a) ^{1}H-NMR and (b) ^{13}C-NMR.

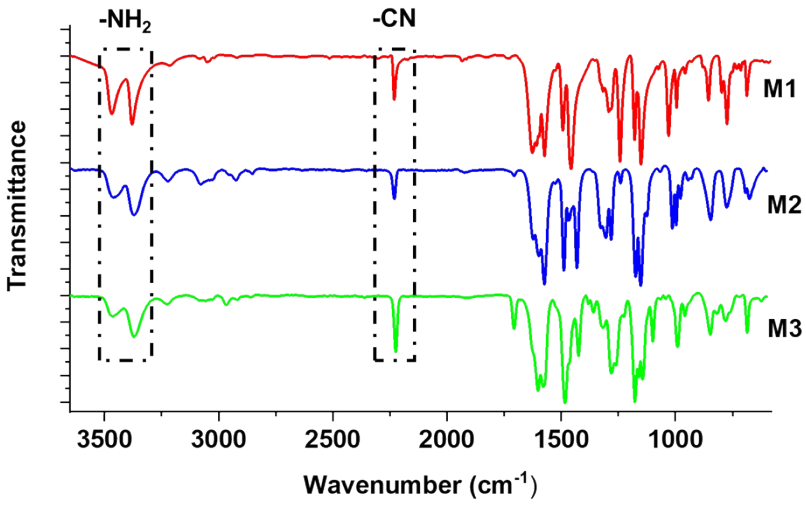


Figure S3. FTIR spectra of diamines **M1**, **M2** and **M3**.

Table S1. Targeted and experimental composition of the CN- and J-2000-based copolymers.

Copolymers	Targeted molar ratio of the structural units		Experimental molar ratio of the structural units from ^{1}H-NMR		Experimental wt % ratio of structural units from ^{1}H-NMR	
	M-BTDA	J-2000-BTDA	M-BTDA	J-2000-BTDA	M-BTDA	J-2000-BTDA
coPI-1	0.7	0.3	0.79	0.21	50.24	49.76
coPI-2	0.7	0.3	0.71	0.29	40.81	59.19
coPI-3	0.7	0.3	0.72	0.28	42.03	57.97

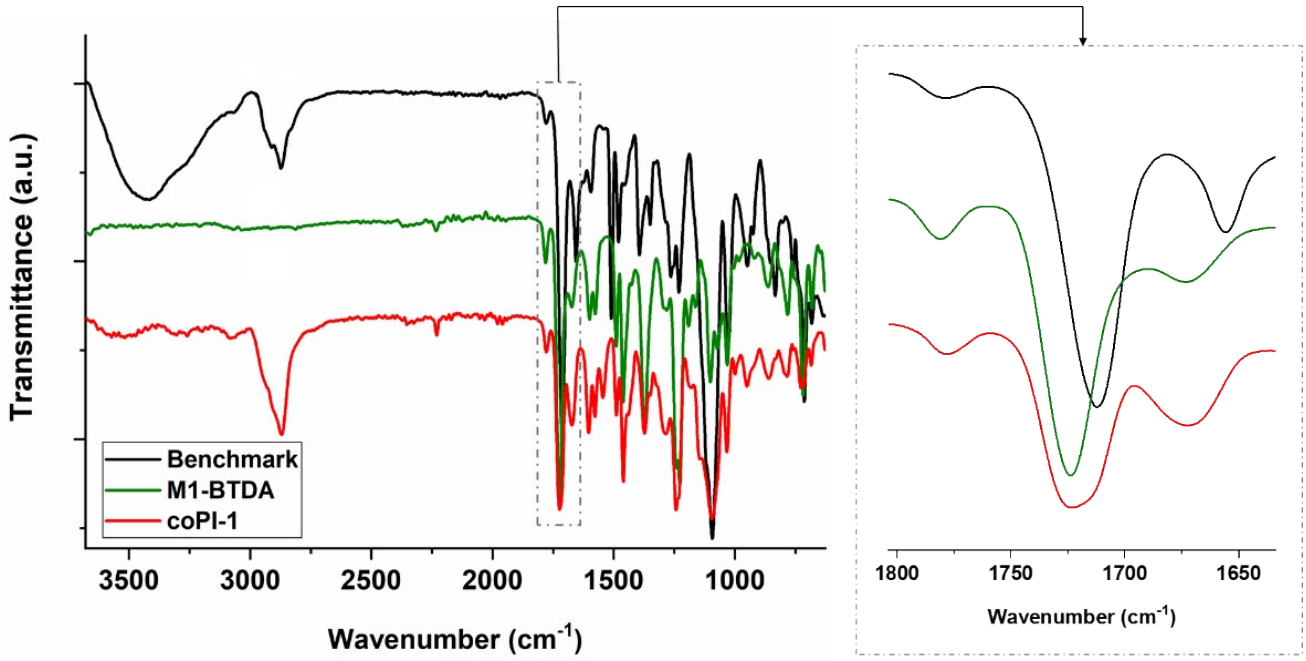


Figure S4. Compared FTIR spectra of **coPI-1**, benchmark and M1-BTDA.

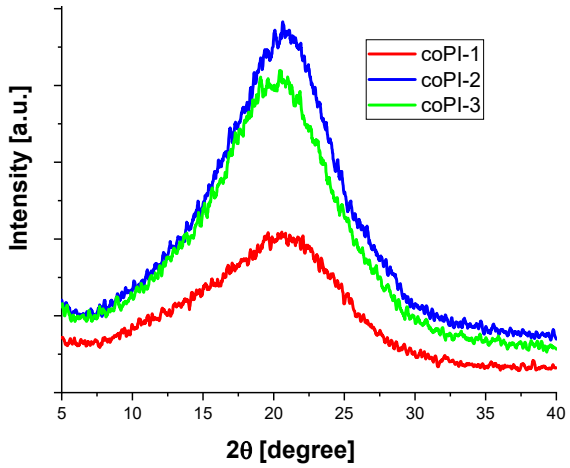


Figure S5. Wide-angle X-ray diffraction patterns of copolyimide films.

Table S2. The solubility tests and the molecular weights of copolymers.

Copolymer	Solvent								M_n (g/mol)	M_w (g/mol)	PDI (M_w/M_n)
	$CHCl_3$	THF	ACN	DCM	DMF	DMAc	DMSO	NMP			
coPI-1	±	++	-	±	±	++	++	++	404 000	1 008 600	2.49
coPI-2	±	++	-	±	±	++	++	++	197 900	311 300	1.57
coPI-3	++	++	-	++	++	++	++	++	155 800	218 300	1.40

++ soluble at room temperature; ± partially soluble/swelling; - insoluble/swelling even on heating

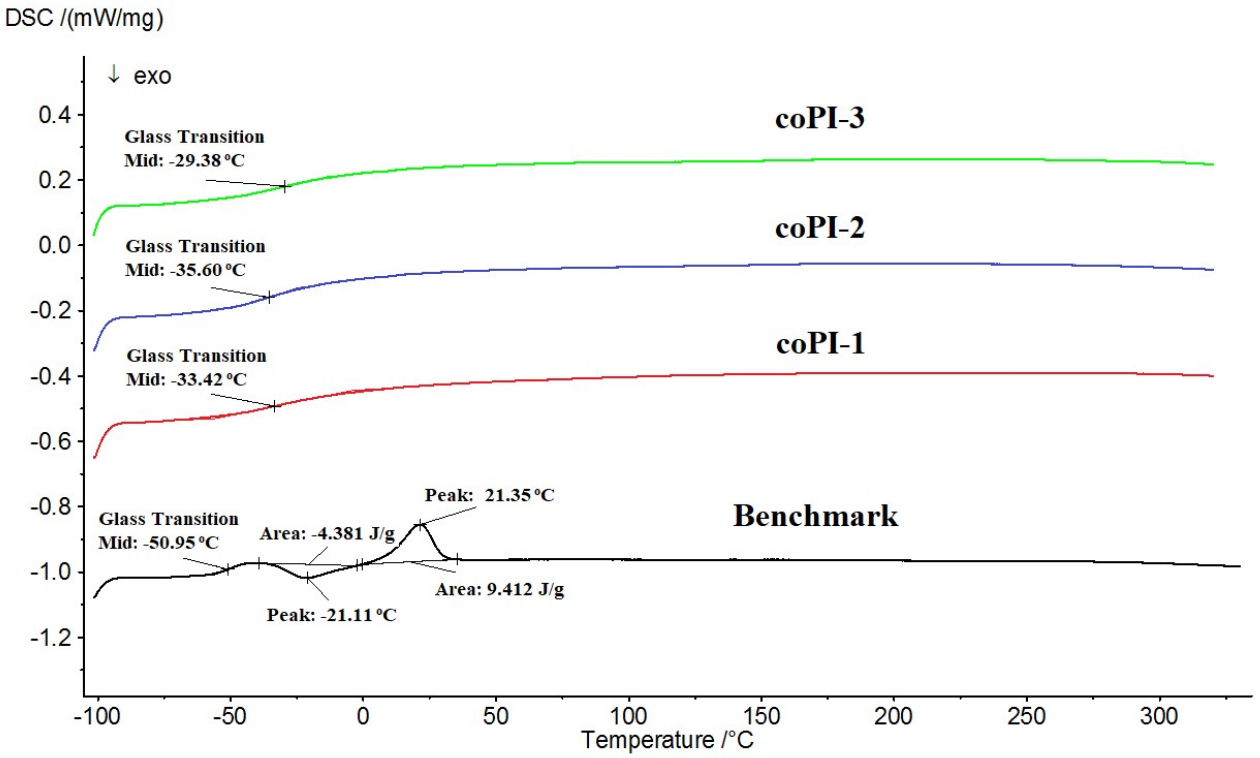


Figure S6. Comparative DSC curves of **coPI-1** – **coPI-3** and of the benchmark.

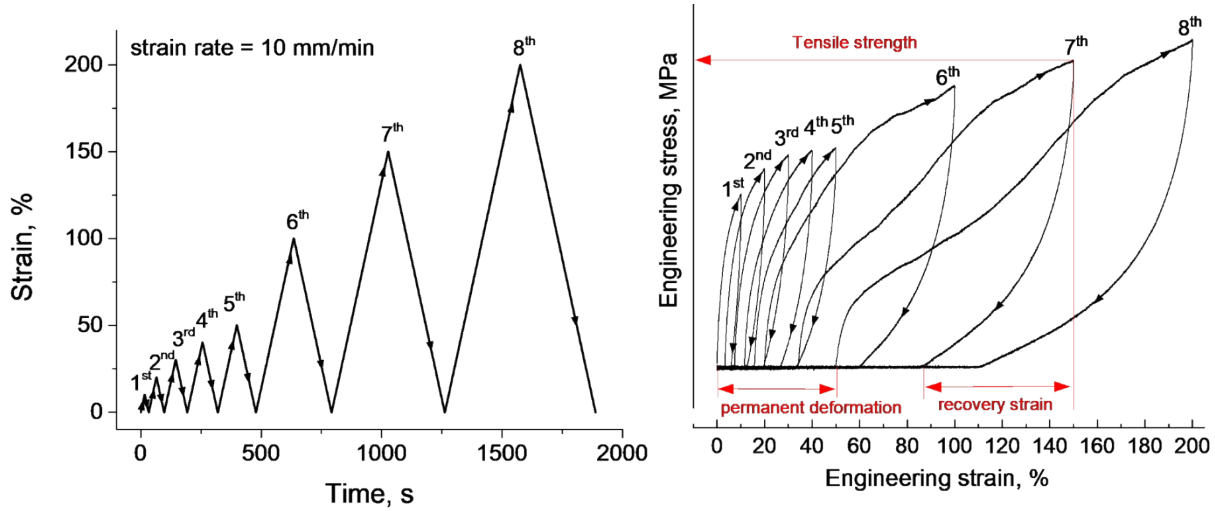


Figure S7. Loading protocol for the cyclic experiments and the obtained stress-strain curves. Definition of tensile strength, permanent deformation, recovery strain is illustrated.

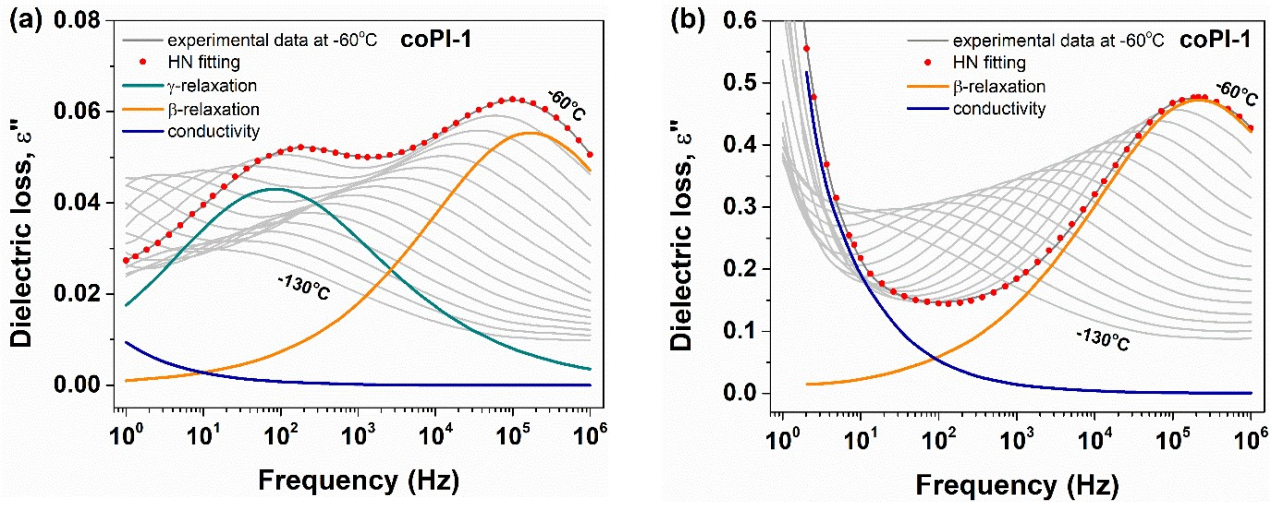


Figure S8. The evolution of dielectric loss with frequency at various temperatures, during the first (a) and second (b) heating cycles, for **coPI-1** film.

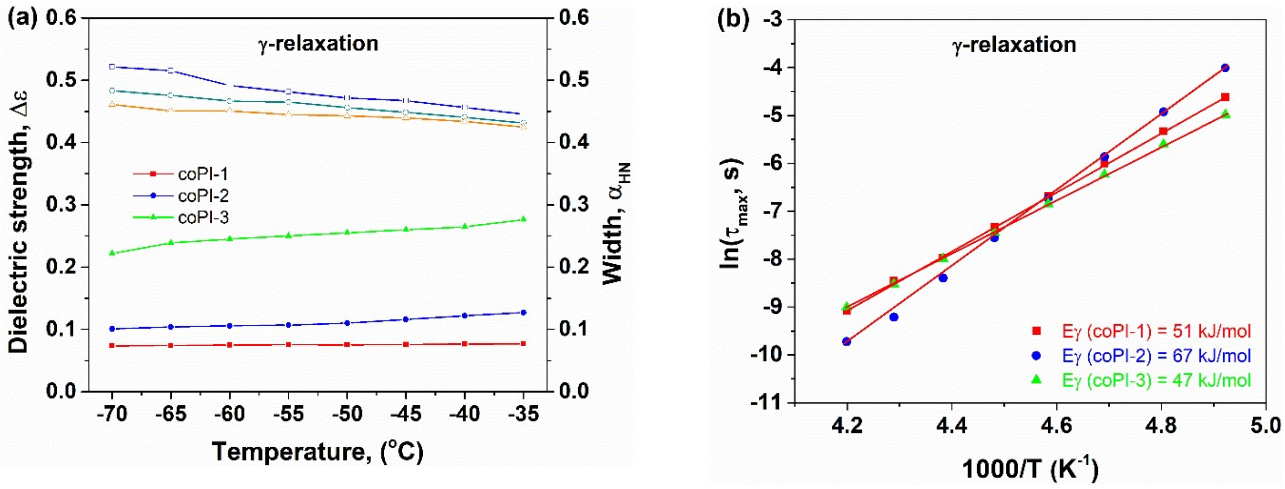


Figure S9. (a) The evolution of dielectric strength (solid symbols) and width (open symbols) with temperature and (b) Arrhenius plots and activation energy for γ-relaxation of the copolymers.

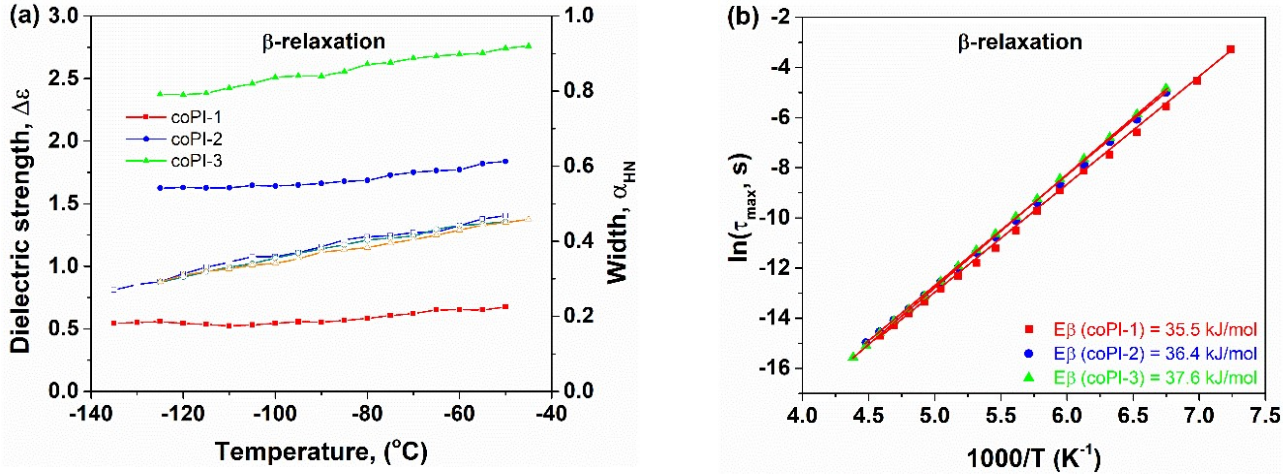


Figure S10. (a) The evolution of dielectric strength (solid symbols) and width (open symbols) with temperature and (b) Arrhenius plots and activation energy for β-relaxation of copolymers.