Supplementary data

Shape-Memory Capability of Binary Multiblock Copolymer Blends with Hard and Switching Domains Provided by Different Components

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Table S1a. Mechanical properties of polymer blends obtained by co-precipitation method determined in tensile tests. E: E-Modulus, σ_y: stress at yield, ε_y: elongation at yield, σ_b: stress at break, ε_b: elongation at break.

Polymer blend ID [a)]	E [b)] (MPa)	σ_y [b)] (MPa)	ε_y [b)] (%)	σ_b [b)] (MPa)	ε_b [b)] (%)	E [c)] (MPa)	σ_b [c)] (MPa)	ε_b [c)] (%)
PDA(42)/PCA(47)[21/24/42]	15.5 ± 1.3	-	-	12.5 ± 3.5	1,295 ± 335	2.3 ± 0.4	1.1 ± 0.2	240 ± 40
PDA(42)/PCA(47)[28/16/42]	23.3 ± 3.6	-	-	9.5 ± 2.1	1,070 ± 30	4.5 ± 0.4	1.9 ± 0.2	270 ± 20
PDA(42)/PCA(47)[34/10/44]	16.8 ± 2.2	-	-	5.5 ± 0.7	445 ± 105	7.2 ± 1.5	2.5 ± 0.1	210 ± 70
PDA(42)/PCA(68)[14/45/28]	32.7 ± 1.3	4.3 ± 0.8	35 ± 7	12.5 ± 0.2	860 ± 95	0.7 ± 0.5	0.8 ± 0.2	1,055 ± 385
PDA(42)/PCA(68)[21/34/32]	25.6 ± 0.8	5.1 ± 0.2	58 ± 11	9.6 ± 3.0	795 ± 270	2.6 ± 0.1	1.0 ± 0.1	325 ± 60
PDA(42)/PCA(68)[28/22/36]	24.5 ± 2.1	-	-	8.9 ± 1.6	925 ± 180	4.1 ± 0.6	2.0 ± 0.2	320 ± 105
PDA(42)/PCA(68)[34/14/40]	16.9 ± 0.8	-	-	10.5 ± 0.7	1,280 ± 1	5.9 ± 0.1	1.9 ± 0.1	75 ± 1
PDA(50)/PCA(47)[25/24/37]	15.9 ± 2.4	4.9 ± 0.0	51 ± 3	9.8 ± 0.8	750 ± 110	6.5 ± 0.8	2.3 ± 0.1	410 ± 15
PDA(50)/PCA(47)[33/16/37]	20.5 ± 4.6	5.6 ± 0.4	51 ± 10	10.6 ± 1.1	705 ± 160	10.0 ± 1.7	2.7 ± 0.0	340 ± 10
PDA(50)/PCA(47)[40/9/37]	32.0 ± 2.0	6.3 ± 0.2	58 ± 6	11.8 ± 0.6	845 ± 90	14.9 ± 2.8	5.0 ± 0.2	575 ± 90
PDA(50)/PCA(68)[17/45/25]	33.9 ± 1.8	-	-	9.1 ± 0.2	970 ± 100	1.7 ± 0.1	1.2 ± 0.0	550 ± 95
PDA(50)/PCA(68)[25/34/28]	36.0 ± 2.0	-	-	13.6 ± 1.2	1,130 ± 150	4.9 ± 0.5	2.2 ± 0.1	460 ± 70
PDA(50)/PCA(68)[34/22/31]	37.0 ± 2.1	5.1 ± 0.3	78 ± 9	13.8 ± 1.8	1,055 ± 25	7.1 ± 1.0	3.0 ± 0.1	465 ± 65
PDA(50)/PCA(68)[40/14/34]	41.7 ± 3.1	5.8 ± 0.3	72 ± 12	15.5 ± 1.6	1,510 ± 20	17.6 ± 3.1	5.0 ± 0.2	480 ± 40

a) The three numbers in squared brackets of the sample ID give the content in wt% of PPDO, PCL and PADOH in the binary polymer blends.
b) determined at 20 °C
c) determined at 50 °C

Table S1b. Mechanical properties of polymer blends obtained by co-extrusion method determined in tensile tests. E: E-Modulus, σ_y: stress at yield, ε_y: elongation at yield, σ_b: stress at break, ε_b: elongation at break.

Polymer blend ID [a]	E [b] (MPa)	σ_y [b] (MPa)	ε_y [b] (%)	σ_b [b] (MPa)	ε_b [b] (%)	E [c] (MPa)	σ_b [c] (MPa)	ε_b [c] (%)
PDA(42)/PCA(47)[21/24/42]	23.8 ± 2.4	-	-	8.8 ± 0.8	735 ± 60	2.4 ± 0.6	0.7 ± 0.1	300 ± 70
PDA(42)/PCA(47)[28/16/42]	24.2 ± 1.1	-	-	6.6 ± 0.7	460 ± 35	7.0 ± 0.8	1.9 ± 0.2	240 ± 75
PDA(42)/PCA(47)[34/10/44]	27.3 ± 2.4	-	-	7.6 ± 0.7	530 ± 65	4.9 ± 0.6	1.9 ± 0.5	315 ± 15
PDA(42)/PCA(68)[14/45/28]	36.8 ± 3.7	4.5 ± 0.2	33 ± 2	16.4 ± 1.6	820 ± 40	0.3 ± 0.3	0.1 ± 0.05	360 ± 60
PDA(42)/PCA(68)[21/34/32]	39.4 ± 1.2	4.7 ± 0.1	39 ± 2	11.9 ± 0.4	705 ± 20	1.7 ± 0.2	0.5 ± 0.1	375 ± 15
PDA(42)/PCA(68)[28/22/36]	37.2 ± 1.6	-	-	8.0 ± 0.4	450 ± 40	3.4 ± 0.9	1.6 ± 0.1	375 ± 45
PDA(42)/PCA(68)[34/14/40]	38.4 ± 4.1	-	-	6.9 ± 0.3	305 ± 20	6.8 ± 1.5	2.5 ± 0.5	255 ± 30
PDA(50)/PCA(47)[25/24/37]	38.0 ± 4.5	-	-	12.4 ± 0.8	690 ± 50	8.1 ± 4.1	2.9 ± 0.5	320 ± 95
PDA(50)/PCA(47)[33/16/37]	39.4 ± 4.6	-	-	14.8 ± 0.8	820 ± 40	7.3 ± 0.6	3.0 ± 0.2	465 ± 55
PDA(50)/PCA(47)[40/9/37]	54.1 ± 4.0	-	-	13.1 ± 0.4	605 ± 40	12.7 ± 0.7	5.0 ± 0.3	525 ± 10
PDA(50)/PCA(68)[17/45/25]	40.5 ± 9.4	4.1 ± 0.6	29 ± 3	16.7 ± 0.7	780 ± 35	4.1 ± 1.3	1.2 ± 0.6	310 ± 125
PDA(50)/PCA(68)[25/34/28]	45.2 ± 4.9	5.2 ± 0.2	35 ± 3	17.6 ± 0.4	755 ± 15	4.7 ± 0.7	2.0 ± 0.1	535 ± 115
PDA(50)/PCA(68)[34/22/31]	49.6 ± 3.1	6.0 ± 0.1	37 ± 7	13.5 ± 1.0	615 ± 40	9.0 ± 1.4	3.0 ± 0.2	410 ± 65
PDA(50)/PCA(68)[40/14/34]	60.3 ± 5.9	6.7 ± 0.02	33 ± 6	13.7 ± 0.9	640 ± 1	15.4 ± 4.2	5.4 ± 0.1	590 ± 135

a) The three numbers in squared brackets of the sample ID give the content in wt% of PPDO, PCL and PADOH in the polymer blends.
b) determined at 20 °C
c) determined at 50 °C

Table S2. Shape-memory properties of binary polymer blends obtained by co-precipitation and co-extrusion method determined in strain-controlled cyclic, thermomechanical tests. T_{low} = 0 °C, T_{high} = 50 °C, ε_m = 100%, $R_f(N)$: strain fixity rate, $R_r(N)$: strain recovery rate, N: cycle number, $\overline{R}_{f,1\text{-}5}$: average of R_f from 1st to 5th cycle, $\overline{R}_{r,2\text{-}4}$: average of R_r from 2nd to 4th cycle.

	co-precipitation				co-extrusion			
Polymer blend ID [a)]	$\overline{R}_{f,1-5}$ (%)	$R_r(1)$ (%)	$R_r(2)$ (%)	$\overline{R}_{r,2-4}$ (%)	$\overline{R}_{f,1-5}$ (%)	$R_r(1)$ (%)	$R_r(2)$ (%)	$\overline{R}_{r,2-4}$ (%)
PDA(42)/PCA(47)[21/24/42]	81.9 ± 0.8	80.3	97.1	97.3 ± 0.2	86.6 ± 0.5	63.8	91.7	95.2 ± 2.8
PDA(42)/PCA(47)[28/16/42]	68.1 ± 0.3	73.9	96.3	98.1 ± 1.6	75.8 ± 3.1	67.7	93.9	95.1 ± 1.3
PDA(42)/PCA(47)[34/10/44]	66.9 ± 1.0	84.7	96.8	98.9 ± 2.0	73.2 ± 2.7	70.1	92.8	95.2 ± 2.1
PDA(42)/PCA(68)[14/45/28]	93.7 ± 0.5	76.6	94.5	97.4 ± 3.9	98.4 ± 0.4	59.5	82.2	84.0 ± 3.1
PDA(42)/PCA(68)[21/34/32]	92.2 ± 0.3	73.3	96.6	98.6 ± 3.6	95.7 ± 0.3	58.9	90.1	92.4 ± 2.0
PDA(42)/PCA(68)[28/22/36]	84.5 ± 0.3	72.8	97.2	98.2 ± 0.9	89.8 ± 1.2	68.2	92.2	96.1 ± 3.5
PDA(42)/PCA(68)[34/14/40]	72.8 ± 0.3	76.8	95.9	97.2 ± 1.6	87.4 ± 0.3	63.9	94.5	96.6 ± 3.0
PDA(50)/PCA(47)[25/24/37]	85.4 ± 2.1	63.8	95.1	95.1 ± 3.0	86.3 ± 5.1	62.5	96.2	96.6 ± 0.9
PDA(50)/PCA(47)[33/16/37]	80.9 ± 0.3	55.0	91.0	94.6 ± 3.2	79.6 ± 0.6	71.5	97.3	98.7 ± 3.6
PDA(50)/PCA(47)[40/9/37]	77.8 ± 2.9	63.8	92.0	95.1 ± 3.0	73.8 ± 5.4	67.7	93.9	95.8 ± 3.5
PDA(50)/PCA(68)[17/45/25]	96.3 ± 0.8	58.6	95.7	94.6 ± 3.3	99.1 ± 1.2	58.6	83.4	88.9 ± 4.8
PDA(50)/PCA(68)[25/34/28]	90.8 ± 0.6	55.3	94.4	94.0 ± 1.5	92.9 ± 0.1	61.6	93.6	95.0 ± 1.7
PDA(50)/PCA(68)[34/22/31]	86.4 ± 1.1	57.1	91.8	96.5 ± 5.4	89.7 ± 1.8	60.0	94.2	96.0 ± 2.3

a) The three numbers in squared brackets of the sample ID give the content in wt% of PPDO, PCL and PADOH in the polymer blends according to calculation.

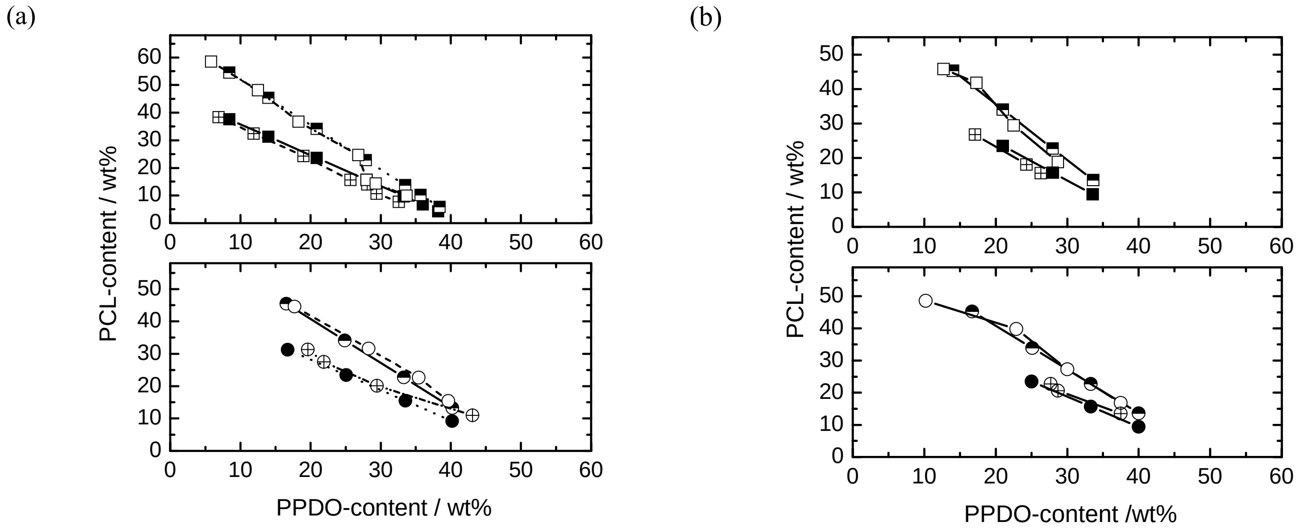

Fig. S1. Comparison of the initially weighted quantity and the composition determined by [1]H-NMR-spectroscopy of polymer blends. The initially weighted quantity of the PDA-polymer and the PCA-polymer vary between 10:1, 6:1, 4:1, 2:1, 1:1, 1:2 and 1:4. (a) Polymer blends from solution and (b) polymer blends from extrusion.

! PDA(42)/PCA(47) (weight in quantity), ∃ PDA(42)/PCA(47) ([1]H-NMR-spectroscopy),

(PDA(42)/PCA(68) (weight in quantity), ∀ PDA(42)/PCA(68) ([1]H-NMR-spectroscopy),

, PDA(50)/PCA(47) (weight in quantity), / PDA(50)/PCA(47) ([1]H-NMR-spectroscopy),

3 PDA(50)/PCA(68) (weight in quantity), – PDA(50)/PCA(68) ([1]H-NMR-spectroscopy)

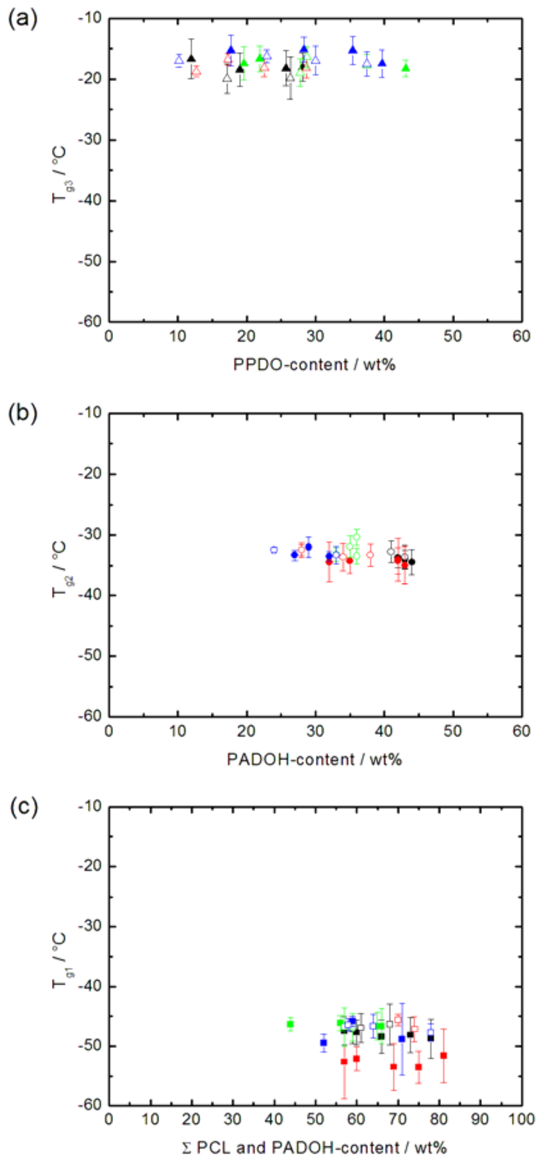

Fig. S2. Glass transition temperatures T_g of binary polymer blends from PDA and PCA as a function of weight content in blends. Filled symbols: polymer blends from co-precipitation: ! T_{g1}; , T_{g2}; 7 T_{g3}; hollow symbols: polymer blends from co-extrusion ∀ T_{g1}; − T_{g2}; 8 T_{g3}. The error bars show the range of T_g. The color indexes the different series: black PDA(42)/PCA(47), red PDA(42)/PCA(68), green PDA(50)/PCA(47), blue PDA(50)/PCA(68). a) T_{g1} denoted to PPDO; b) T_{g2} denoted to PADOH; T_{g3} denoted to PCL

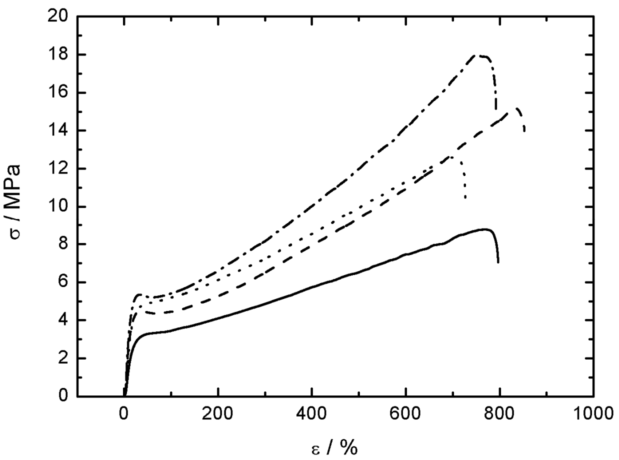

Fig. S3. Stress-Strain diagrams of extruded polymer blends at room temperature: solid line = PDA(42)/PCA(47)[21/24/42], dashed line = PDA(42)/PCA(68)[14/45/28], dotted line = PDA(50)/PCA(47)[25/24/37], dash-dotted line = PDA(50)/PCA(68)[25/34/28].

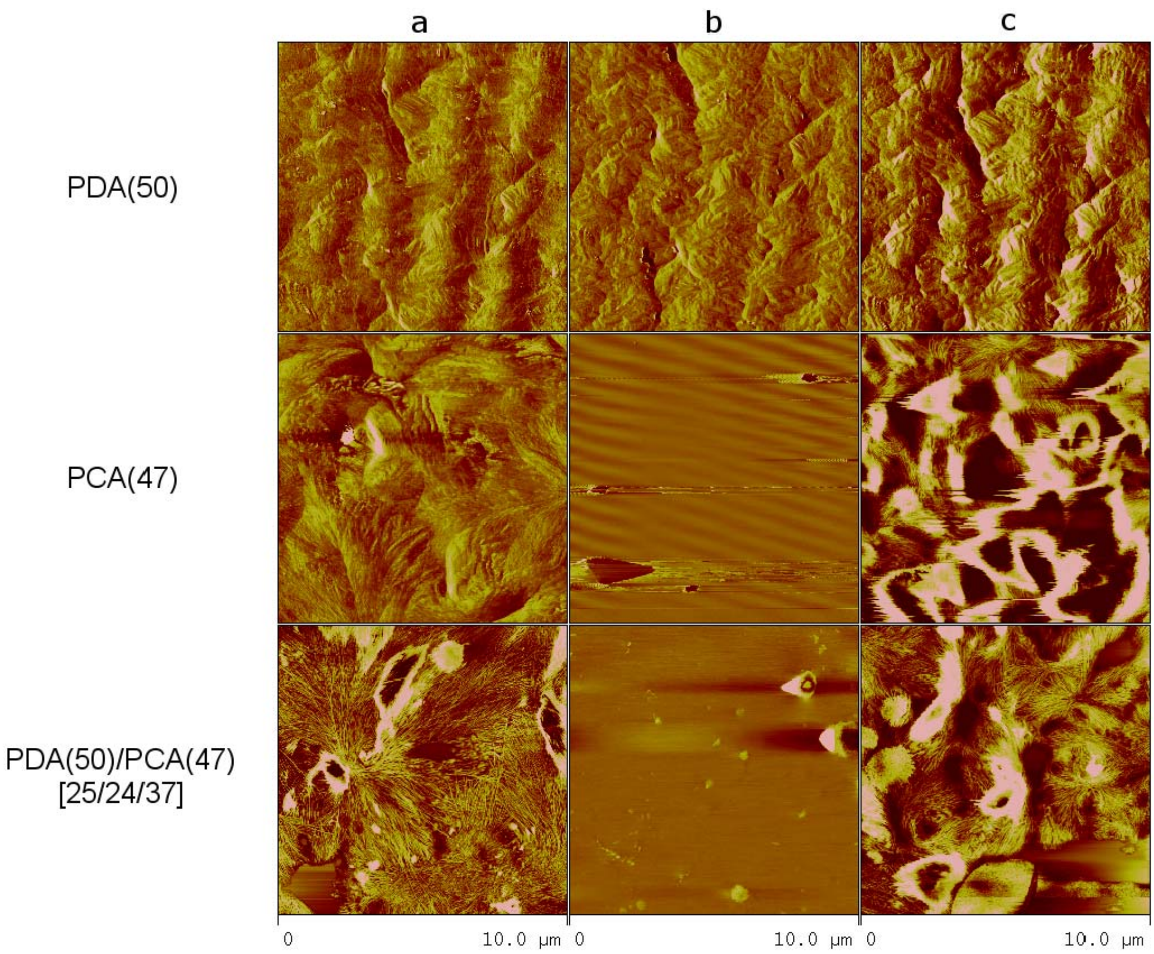

Fig. S4. AFM phase image of multiblock copolymers PDA(50) and PCA(47) as well as polymer blend PDA(50)/PCA(47)[25/24/37] surfaces at different temperatures. a: room temperature, b: 60 °C, c: room temperature after cooling from 60 °C. Dark area = soft phase, and bright area = stiff phase. (phase image, scan size 10.0 µm, scan rate 1 Hz, samples per line 512, z range 60°)

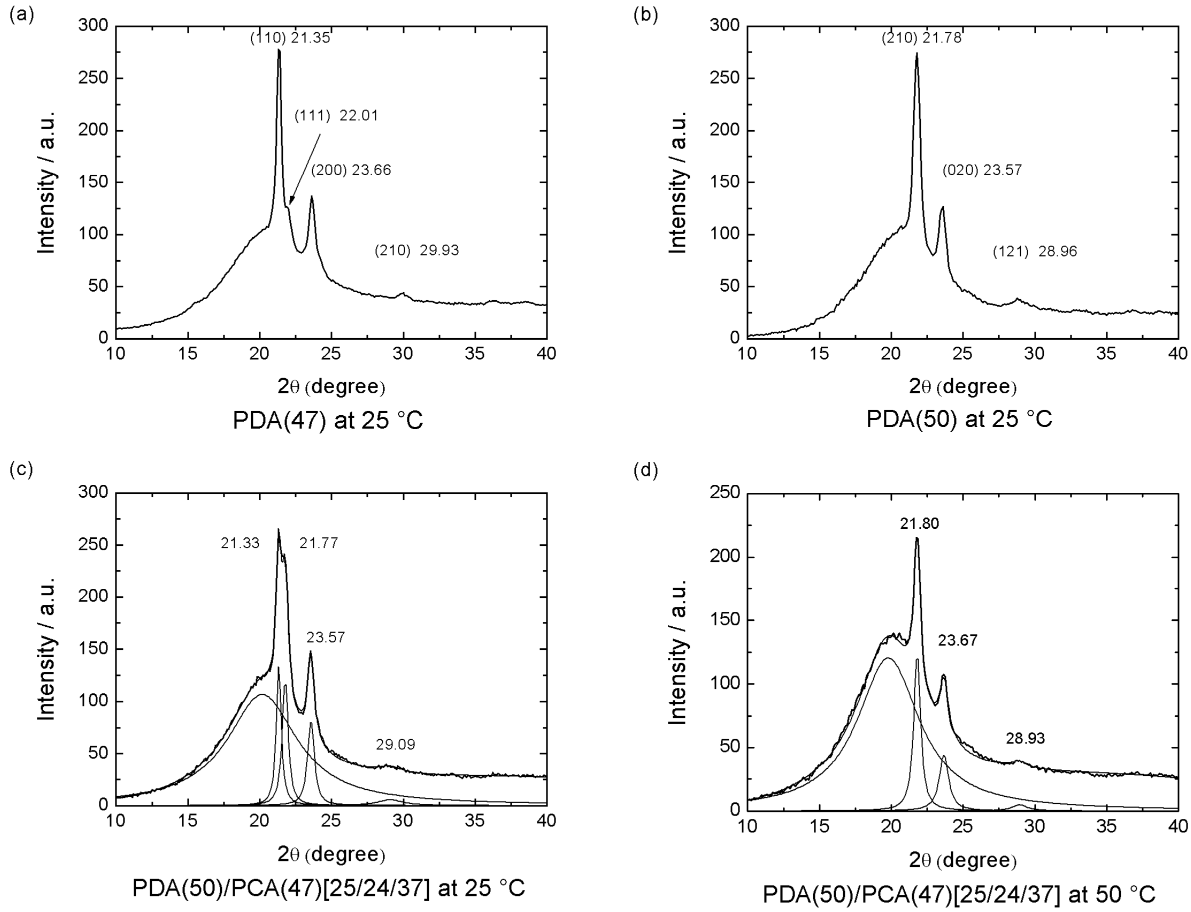

Fig. S5. WAXS diffractogram of multiblock copolymers (a) PCA(47) and (b) PDA(50) as well as polymer blend (c,d) PDA(50)/PCA(47)[25/24/37].

Video S1

The video displays programming and recovery applied to two different multi-block copolymers and a binary polymer blend. Left: the PCL-based multiblock copolymer PCA(47); middle: binary blend PDA(50)/PCA(47)[25/24/37] and right: the PPDO-based multi-block copolymer PDA(50). In part 1 of the video the permanent shape at 25 °C is shown. In part 2 the temporary shape was programmed to each sample at 50 °C and is shown at an environmental temperature of 25 °C (part 2). In part 3 the thermally-induced shape-memory effect was triggered by heating to 50 °C (part 3). Only the binary blend of the multi-block copolymers shows a shape-memory effect (sample in the middle).