

ADVANCED FUNCTIONAL MATERIALS

Supporting Information

for *Adv. Funct. Mater.*, DOI: 10.1002/adfm.202207317

Peptide-Reinforced Amphiphilic Polymer Conetworks

*Sara T. R. Velasquez, Daseul Jang, Peter Jenkins, Peng Liu, Liu Yang, LaShanda T. J. Korley, and Nico Bruns**

Supporting Information

Peptide-Reinforced Amphiphilic Polymer Conetworks

*Sara T. R. Velasquez, Daseul Jang, Peter Jenkins, Peng Liu, Liu Yang, LaShanda T. J. Korley, Nico Bruns**

S. T. R. Velasquez, Prof. N. Bruns
Department of Pure and Applied Chemistry, University of Strathclyde, Thomas Graham Building, 295 Cathedral Street, Glasgow G1 1XL, United Kingdom.

Department of Chemistry, Technical University of Darmstadt, Alarich-Weiss-Straße 4, 64287 Darmstadt, Germany.

E-mail: nico.bruns@tu-darmstadt.de

Dr. P. Liu
Adolphe Merkle Institute, University of Fribourg, Chemin des Verdiers 4, 1700 Fribourg, Switzerland.

D. Jang
Department of Materials Science and Engineering, University of Delaware, 127 The Green, 209 DuPont Hall, Newark, DE, USA, 19716.

Prof. L. T. J. Korley
Department of Materials Science and Engineering, University of Delaware, 127 The Green, 209 DuPont Hall, Newark, DE, USA, 19716.

Department of Chemical and Biomolecular Engineering, University of Delaware, 150 Academy Street, Newark, DE, USA, 19716.

Dr. P. Jenkins, Dr. L. Yang
Department of Mechanical and Aerospace Engineering, University of Strathclyde, James Weir Building, 75 Montrose Street, Glasgow G1 1XJ, United Kingdom.

Table S1. Relative amount of peptide secondary structure, β -sheet and α -helix, as a function of PBLA repeat length and content and percentage of hydrophilic phase. Calculated from the IR spectroscopy data shown in Figure 2 and S1.

	α -helix (%)	β -sheet (%)
PBLA00_30	0	0
PBLA05_30	0	100
PBLA20_30	48	52
PBLA00_50	0	0
PBLA05_50	0	100
PBLA20_50	50	50
PBLA00_70	0	0
PBLA05_70	0	100
PBLA20_70	31	69

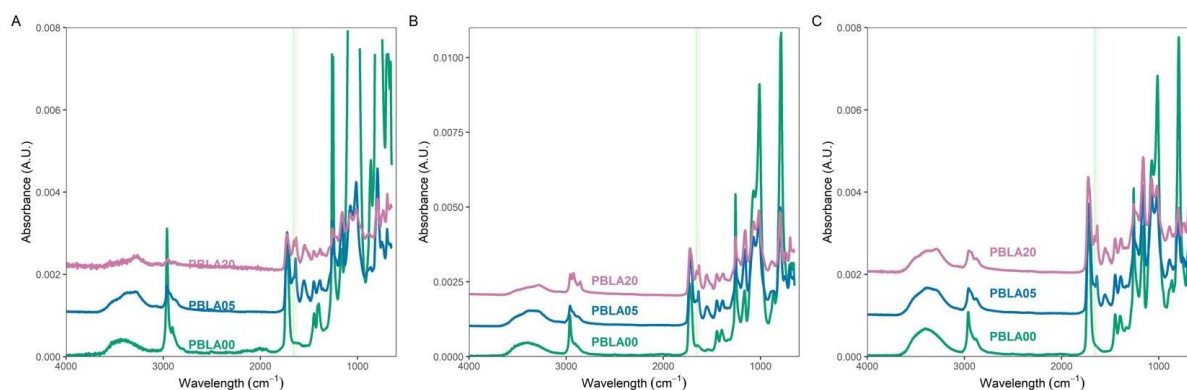


Figure S1. Full ATR-FTIR spectra of PHEA-*l*-PDMS and PHEA-*l*-(PBLA_x-*b*-PDMS-*b*-PBLA_x) APCNs. A) APCNs with 30 wt% PHEA and different amounts of PBLA. B) APCNs with 50 wt% PHEA and different amounts of PBLA. C) APCNs with 70 wt% PHEA and different amounts of PBLA.

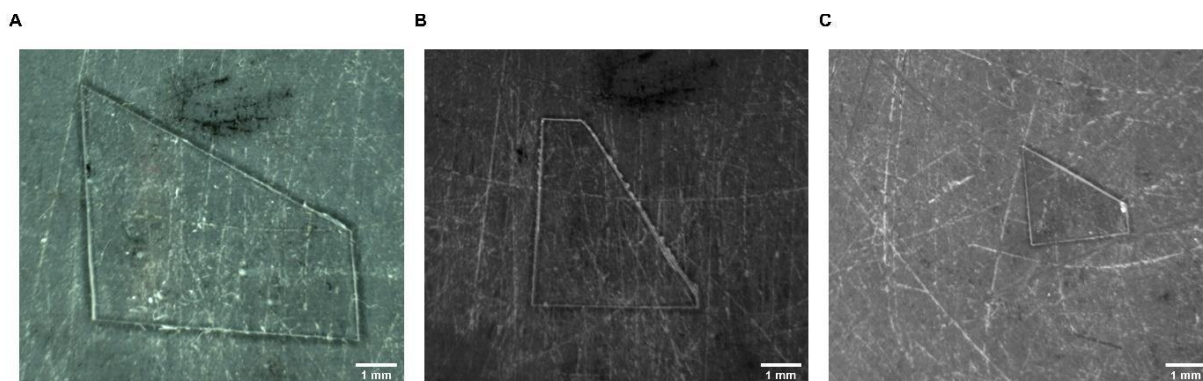


Figure S2. Optical microscope image of APCNs, on a scratched surface, showing the transparency of the samples. A) PBLA00_50, B) PBLA05_50 and C) PBLA20_50. All images are in greyscale. Scale bars are 1 mm.

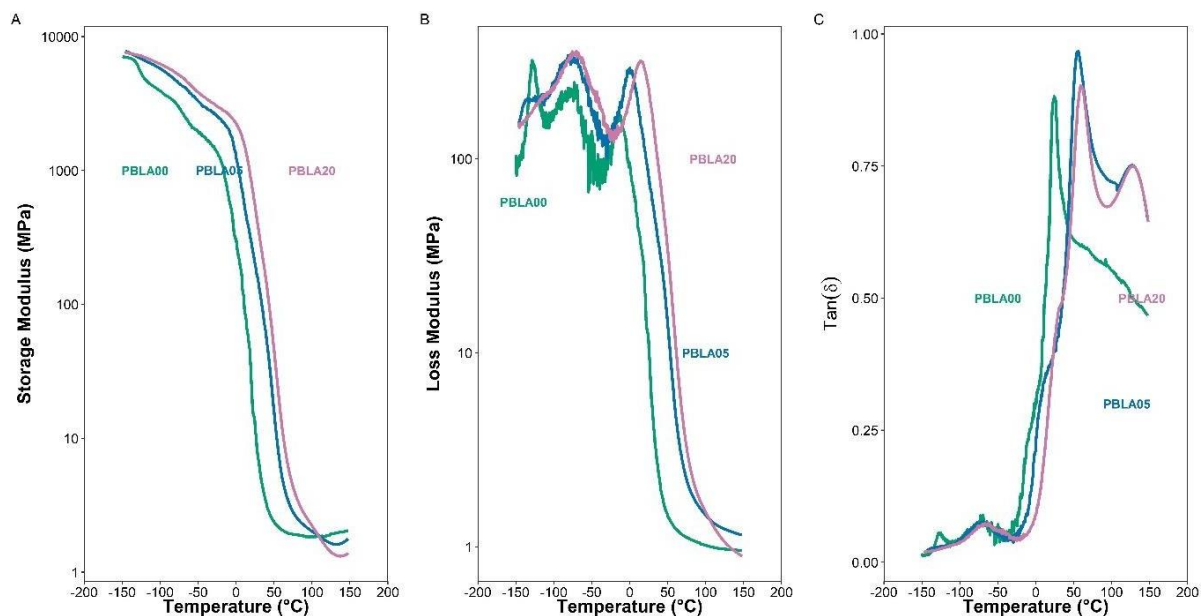


Figure S3. DMA temperature sweep of PBLA00_70, PBLA05_70 and PBLA20_70 samples. A) Storage modulus, B) Loss modulus, and C) Loss factor $\tan \delta$.

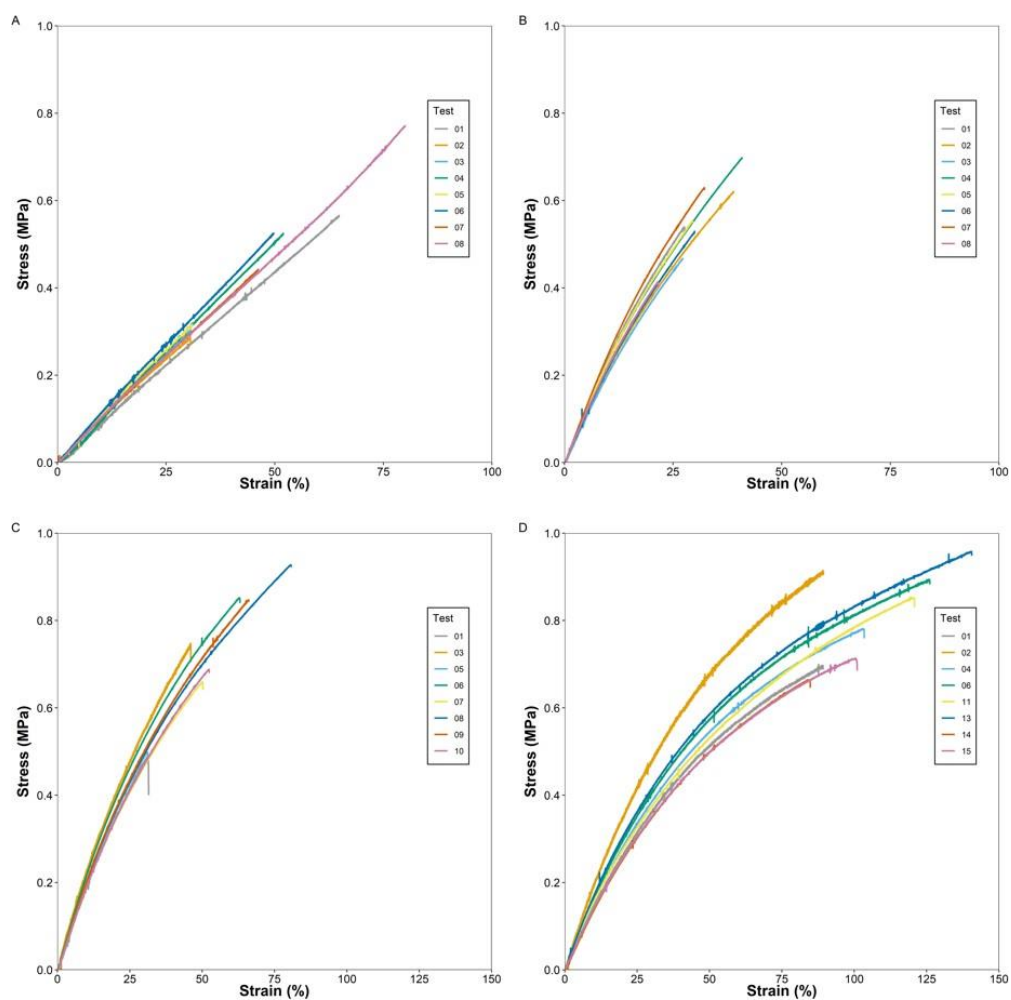


Figure S4. Stress strain curves of the PBLA00 samples with different PHEA content measured at a strain rate of 1 mm min^{-1} . A) PBLA00_00; B) PBLA00_30; C) PBLA00_50; D) PBLA00_70.

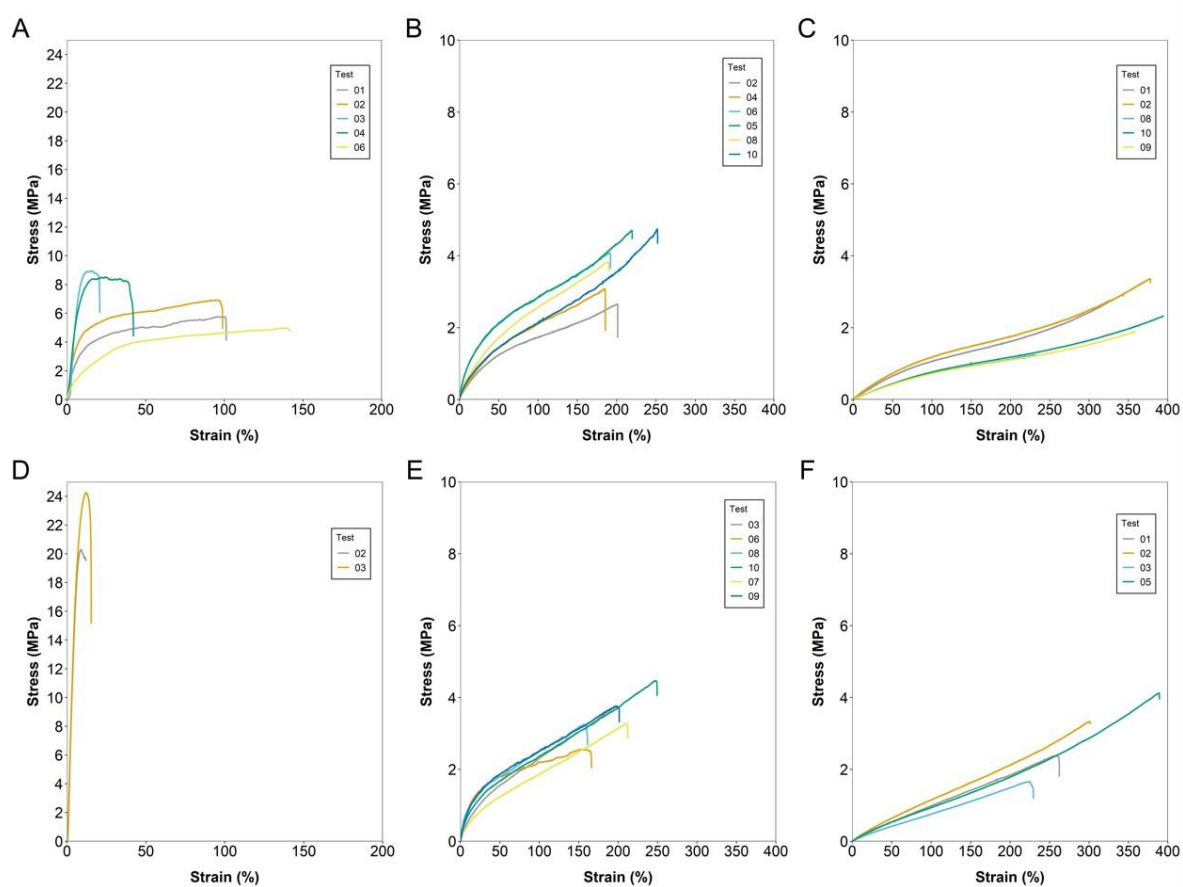


Figure S5. Stress-strain curves for PBLA05 and PBLA20 samples with different PHEA content measured at a strain rate of 1 mm min^{-1} . A) PBLA05_30; B) PBLA05_50; C) PBLA05_70; D) PBLA20_30; E) PBLA20_50; F) PBLA20_70.

Table S2. Summary of maximum stress, strain at break, Young's modulus and toughness for the APCNs measured through stress-strain curves at 1 mm min⁻¹. (Mean values of n= 2 – 5 samples, error = SD)

APCN	Stress (MPa)	Strain (%)	Young's Modulus (MPa)	Toughness (MJ m ⁻³)
PBLA00_00	0.5 ± 0.2	48 ± 17	1.01 ± 0.09	0.13 ± 0.09
PBLA00_30	0.6 ± 0.1	31 ± 6	2.25 ± 0.15	0.10 ± 0.04
PBLA00_50	0.7 ± 0.2	53 ± 17	2.02 ± 0.13	0.2 ± 0.1
PBLA00_70	0.8 ± 0.1	107 ± 20	1.56 ± 0.19	0.8 ± 0.2
PBLA05_30	6.7 ± 2.1	73 ± 42	43 ± 26	3.4 ± 1.9
PBLA05_50	3.9 ± 0.9	206 ± 25	6.2 ± 2.2	4.9 ± 1.3
PBLA05_70	2.4 ± 0.8	341 ± 64	1.3 ± 0.4	4.3 ± 1.8
PBLA20_30	22 ± 3	14 ± 2	217 ± 25	2.2 ± 0.7
PBLA20_50	3.5 ± 0.7	198 ± 32	7.4 ± 1.9	4.5 ± 1.2
PBLA20_70	2.9 ± 1.1	296 ± 69	1.4 ± 0.3	4.4 ± 2.3

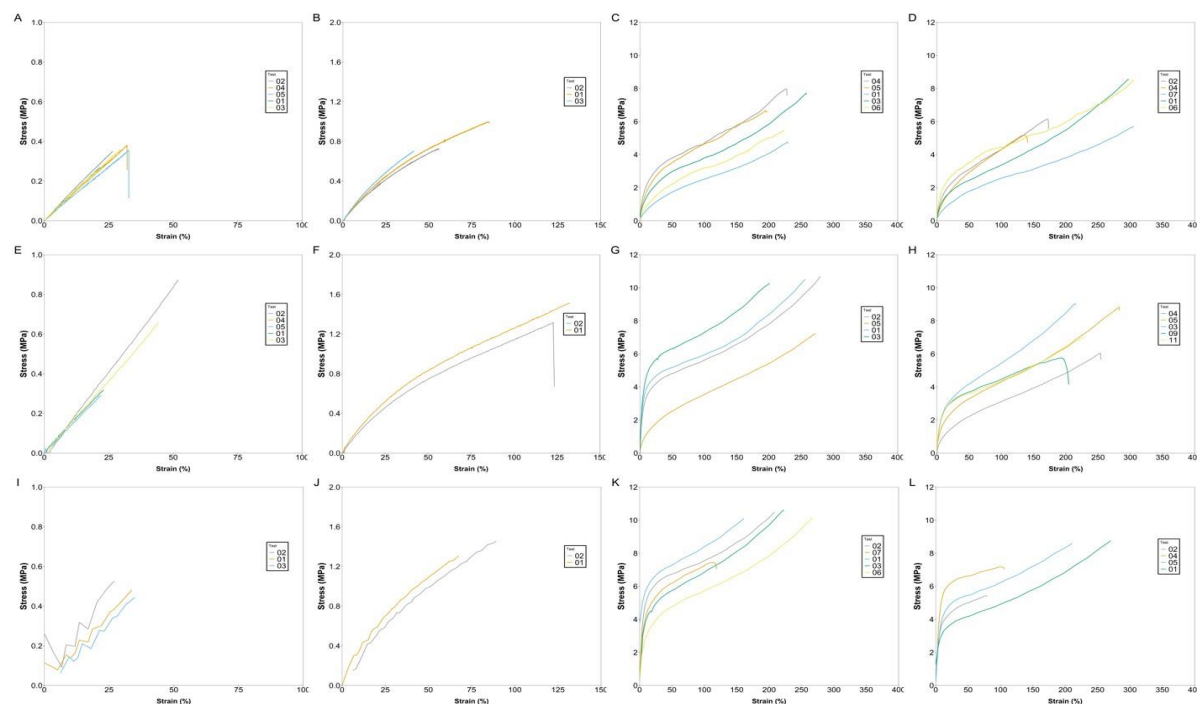


Figure S6. Stress-strain curves for PBLA00, PBLA05 and PBLA20 samples measured at strain rates of 10, 100, and 500 mm min⁻¹. A) PBLA00_00, 10 mm min⁻¹; B) PBLA00_50, 10 mm min⁻¹; C) PBLA05_50, 10 mm min⁻¹; D) PBLA20_50, 10 mm min⁻¹; E) PBLA00_00, 100 mm min⁻¹; F) PBLA00_50, 100 mm min⁻¹; G) PBLA05_50, 100 mm min⁻¹; H) PBLA20_50, 100 mm min⁻¹; I) PBLA00_00, 500 mm min⁻¹; J) PBLA00_50, 500 mm min⁻¹; K) PBLA05_50, 500 mm min⁻¹; L) PBLA20_50, 500 mm min⁻¹.

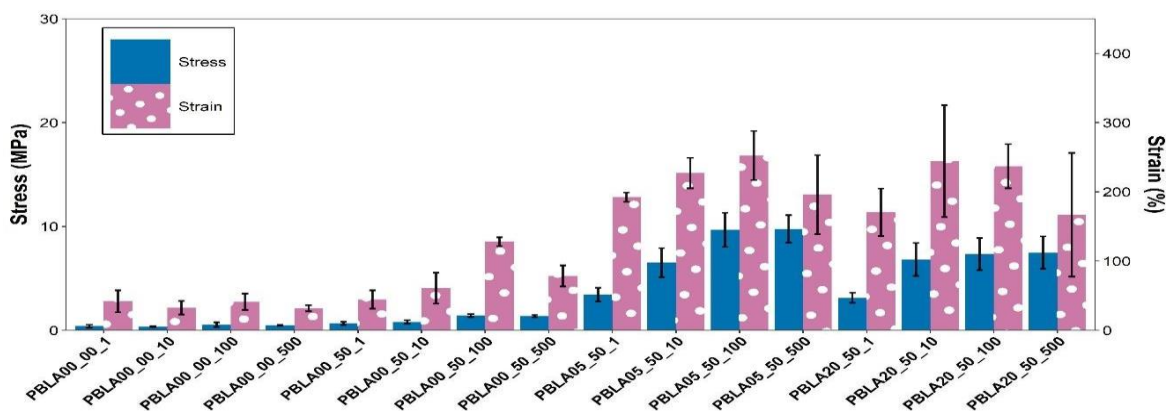


Figure S7. Summary of maximum stress and strain measured for PBLA00_00, PBLA00_50, PBLA05_50, and PBLA20_50 at strain rates of 1, 10, 100 and 500 mm min⁻¹. (Mean values of n= 2 – 5 samples), error = SD.

Table S3. Summary of maximum stress, maximum strain, Young’s modulus and toughness for PBLA00_00, PBLA00_50, PBLA05_50, and PBLA20_50 measured through stress-strain curves at 1, 10, 100, 500 mm min⁻¹. (Mean values of n= 2 – 5 samples), error = SD)

APCN	Strain rate (mm min ⁻¹)	Stress (MPa)	Strain (%)	Young's Modulus (MPa)	Toughness (MJ m ⁻³)
PBLA00_00	1	0.4 ± 0.1	42 ± 16	0.98 ± 0.10	0.09 ± 0.06
PBLA00_00	10	0.35 ± 0.04	32 ± 10	1.9 ± 1.5	0.05 ± 0.01
PBLA00_00	100	0.5 ± 0.2	41 ± 12	2.5 ± 2.2	0.09 ± 0.09
PBLA00_00	500	0.48 ± 0.04	32 ± 4	1.8 ± 1.5	0.08 ± 0.01
PBLA00_50	1	0.7 ± 0.2	44 ± 13	2.1 ± 0.2	0.2 ± 0.1
PBLA00_50	10	0.8 ± 0.2	61 ± 22	2.1 ± 0.2	0.3 ± 0.2
PBLA00_50	100	1.4 ± 0.1	128 ± 6	2.5 ± 0.4	1.1 ± 0.2
PBLA00_50	500	1.4 ± 0.1	78 ± 15	3.9 ± 1.6	0.7 ± 0.2
PBLA05_50	1	3.4 ± 0.7	192 ± 7	5.8 ± 2.3	4.1 ± 0.9
PBLA05_50	10	6.5 ± 1.4	227 ± 22	13.4 ± 5.9	9 ± 2
PBLA05_50	100	9.7 ± 1.6	252 ± 36	29.6 ± 13.5	15 ± 3
PBLA05_50	500	9.7 ± 1.3	196 ± 57	22.3 ± 6.3	13 ± 4
PBLA20_50	1	3.1 ± 0.5	170 ± 34	6.4 ± 2.2	3.4 ± 0.9
PBLA20_50	10	6.8 ± 1.6	244 ± 80	13.4 ± 4.9	10.2 ± 4.7
PBLA20_50	100	7.3 ± 1.5	237 ± 32	17.6 ± 4.8	11.1 ± 2.4
PBLA20_50	500	7.5 ± 1.6	167 ± 89	29.1 ± 13.1	9.8 ± 5.5

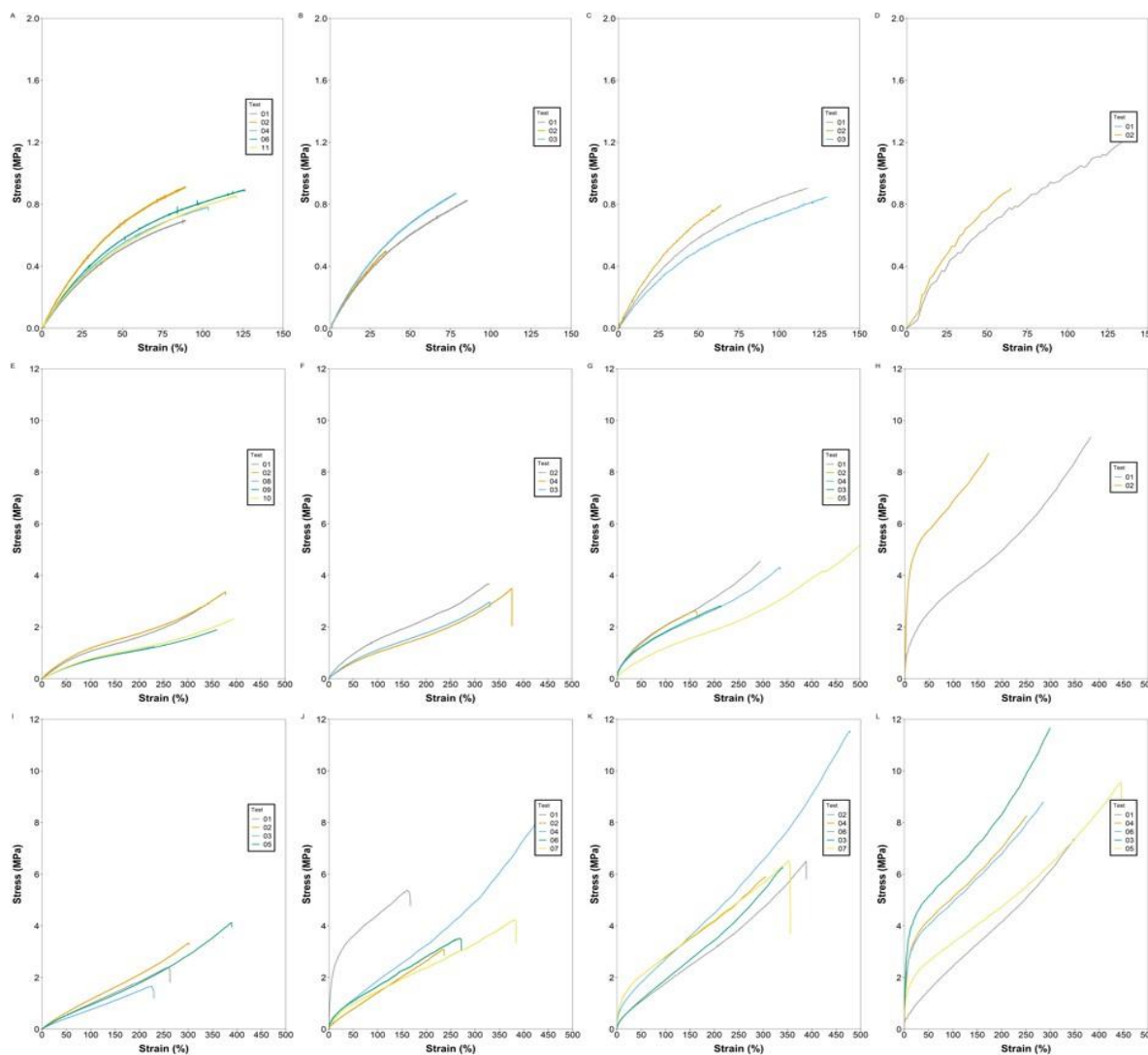


Figure S8. Stress-strain curves for PBLA00_70, PBLA05_70 and PBLA20_70 samples measured at strain rates of 1, 10, 100, and 500 mm min⁻¹. A) PBLA00_70, 1 mm min⁻¹; B) PBLA00_70, 10 mm min⁻¹; C) PBLA00_70, 100 mm min⁻¹; D) PBLA00_70, 500 mm min⁻¹; E) PBLA05_70, 1 mm min⁻¹; F) PBLA05_70, 10 mm min⁻¹; G) PBLA05_70, 100 mm min⁻¹; H) PBLA05_70, 500 mm min⁻¹; I) PBLA20_70, 1 mm min⁻¹; J) PBLA20_70, 10 mm min⁻¹; K) PBLA20_70, 100 mm min⁻¹; L) PBLA20_70, 500 mm min⁻¹.

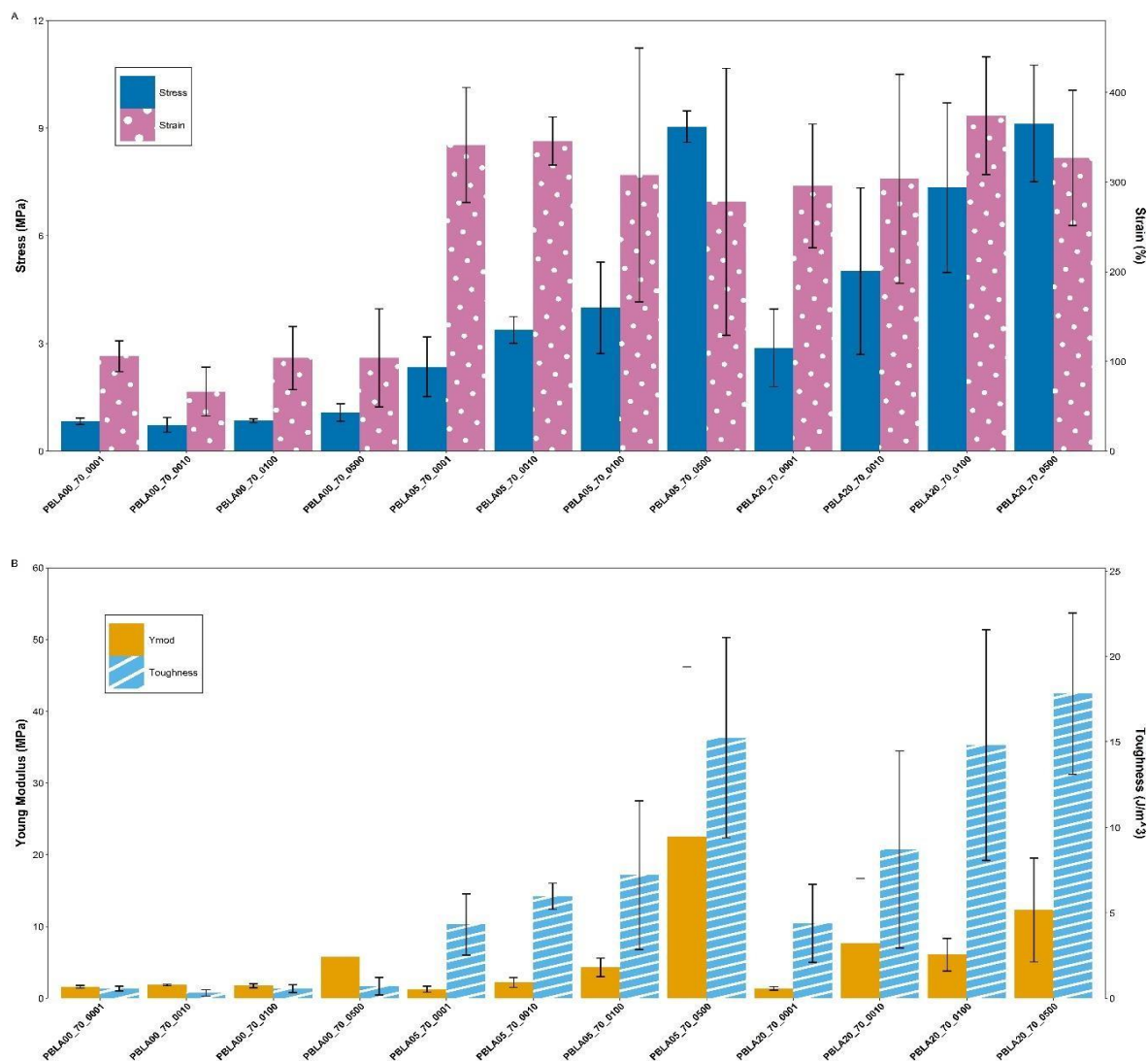


Figure S9. Summary of mechanical properties of PBLA00_70, PBLA05_70, and PBLA20_70 at strain rates of 1, 10, 100 and 500 mm min⁻¹. A) Maximum stress and strain; B) Young's modulus and toughness. (Mean values of n= 5 samples, error = SD)

Table S4. Summary of maximum stress, maximum strain, Young's modulus and toughness for PBLA00_70, PBLA05_70, and PBLA20_70 measured through stress-strain curves at 1, 10, 100, 500 mm min⁻¹.

APCN	Strain rate (mm min ⁻¹)	Stress (MPa)	Strain (%)	Young's Modulus (MPa)	Toughness (MJ m ⁻³)
PBLA00_70	1	0.83 ± 0.09	106 ± 17	1.6 ± 0.2	0.6 ± 0.1
PBLA00_70	10	0.7 ± 0.2	66 ± 27	1.9 ± 0.1	0.3 ± 0.2
PBLA00_70	100	0.7 ± 0.1	104 ± 35	1.8 ± 0.3	0.6 ± 0.3
PBLA00_70	500	1.1 ± 0.2	103 ± 55	- ^a	0.7 ± 0.5
PBLA05_70	1	2.4 ± 0.8	341 ± 64	1.3 ± 0.4	4.3 ± 1.8
PBLA05_70	10	3.4 ± 0.4	346 ± 27	2.2 ± 0.7	5.9 ± 0.8
PBLA05_70	100	4 ± 1	308 ± 141	4.3 ± 1.3	7.2 ± 4.4
PBLA05_70	500	9.0 ± 0.4	278 ± 149	22.6 ± 23.6	15.3 ± 5.9
PBLA20_70	1	2.9 ± 1.1	296 ± 69	1.4 ± 0.3	4.4 ± 2.3
PBLA20_70	10	5.0 ± 2.3	304 ± 116	7 ± 9	8.7 ± 5.8
PBLA20_70	100	7.4 ± 2.4	374 ± 66	6.1 ± 2.3	14.8 ± 6.8
PBLA20_70	500	9.1 ± 1.6	327 ± 75	12.3 ± 7.2	17.8 ± 4.7

^a Young's modulus for sample PBLA00_70 not measured due to noise on the measurement of this sample at 500 mm min⁻¹

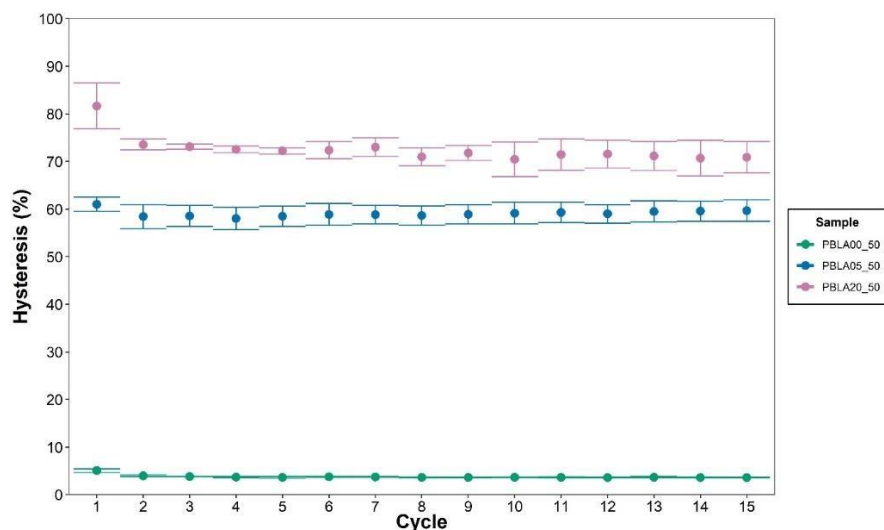


Figure S10. Hysteresis (%) for cyclic test up to 40% strain for PBLA00_50, PBLA05_50, and PBLA20_50 samples. (Mean values of n= 4 samples, error = SD)

Table S5. Hysteresis for cyclic tests up to 40% strain for PBLA00_50, PBLA05_50, and PBLA20_50 samples. (Mean values of n= 4 samples, error = SD)

Cycle	Strain (%)	PBLA00_50		PBLA05_50		PBLA20_50	
		Hysteresis (%)	Max. Stress (Mpa)	Hysteresis (%)	Max. Stress (Mpa)	Hysteresis (%)	Max. Stress (Mpa)
1	40	5.1 ± 0.4	0.699 ± 0.003	61.00 ± 1.5	1.7 ± 0.1	82 ± 5	3.5 ± 0.3
2	40	3.9 ± 0.2	0.696 ± 0.002	58.4 ± 2.5	1.7 ± 0.1	74 ± 1	3.4 ± 0.2
3	40	3.8 ± 0.1	0.694 ± 0.002	58.5 ± 2.2	1.7 ± 0.1	73 ± 1	3.5 ± 0.1
4	40	3.7 ± 0.1	0.695 ± 0.004	58.0 ± 2.3	1.7 ± 0.1	73 ± 1	3.4 ± 0.1
5	40	3.6 ± 0.2	0.694 ± 0.002	58.5 ± 2.1	1.7 ± 0.1	72 ± 1	3.4 ± 0.1
6	40	3.8 ± 0.1	0.693 ± 0.001	58.9 ± 2.3	1.7 ± 0.1	72 ± 2	3.3 ± 0.1
7	40	3.7 ± 0.1	0.695 ± 0.003	59 ± 2	1.7 ± 0.1	73 ± 2	3.4 ± 0.1
8	40	3.6 ± 0.1	0.692 ± 0.001	58 ± 2	1.7 ± 0.1	71 ± 2	3.4 ± 0.1
9	40	3.6 ± 0.1	0.696 ± 0.002	59 ± 2	1.7 ± 0.1	72 ± 2	3.4 ± 0.1
10	40	3.7 ± 0.1	0.693 ± 0.001	59 ± 2	1.7 ± 0.1	70 ± 4	3.4 ± 0.1
11	40	3.6 ± 0.1	0.692 ± 0.001	59 ± 2	1.7 ± 0.1	71 ± 3	3.4 ± 0.1
12	40	3.6 ± 0.1	0.692 ± 0.002	59 ± 2	1.7 ± 0.1	72 ± 3	3.4 ± 0.2
13	40	3.7 ± 0.1	0.691 ± 0.001	59 ± 2	1.7 ± 0.1	71 ± 3	3.5 ± 0.1
14	40	3.6 ± 0.1	0.692 ± 0.002	59.6 ± 2.1	1.7 ± 0.1	71 ± 4	3.4 ± 0.2
15	40	3.6 ± 0.1	0.692 ± 0.001	59.7 ± 2.2	1.7 ± 0.1	71 ± 3	3.4 ± 0.1

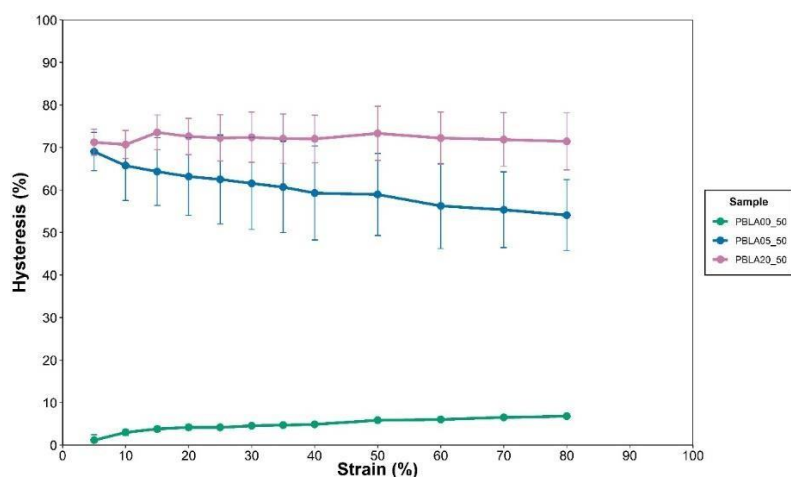


Figure S11. Hysteresis for the elastic cyclic tests for PBLA00_50, PBLA05_50, and PBLA20_50 samples. (Mean values of n= 4 samples, error = SD)

Table S6. Hysteresis for elastic cyclic tests for PBLA00_50, PBLA05_50, and PBLA20_50 samples. (Mean values of n= 4 samples, error = SD)

Cycle	PBLA00_50			PBLA05_50		PBLA20_50	
	Strain (%)	Hysteresis (%)	Max. Stress (Mpa)	Hysteresis (%)	Max. Stress (Mpa)	Hysteresis (%)	Max. Stress (Mpa)
1	5	1.2 ± 1.2	0.12 ± 0.01	69 ± 5	0.8 ± 0.5	71 ± 3	5 ± 1
2	10	3.0 ± 0.7	0.22 ± 0.01	66 ± 8	1.2 ± 0.6	70.7 ± 3.3	2.1 ± 1.2
3	15	3.8 ± 0.8	0.31 ± 0.02	64 ± 8	1.5 ± 0.6	74 ± 4	2.4 ± 1.3
4	20	4.2 ± 0.6	0.39 ± 0.03	63 ± 9	1.7 ± 0.7	72.6 ± 4.3	2.6 ± 1.4
5	25	4.2 ± 0.5	0.46 ± 0.03	63 ± 10	1.9 ± 0.7	72.3 ± 5.4	2.7 ± 1.3
6	30	4.6 ± 0.2	0.53 ± 0.04	62 ± 11	2.1 ± 0.7	72.4 ± 5.9	2.9 ± 1.3
7	35	4.7 ± 0.4	0.60 ± 0.04	61 ± 11	2.3 ± 0.8	72.1 ± 5.8	3 ± 1.3
8	40	4.9 ± 0.4	0.66 ± 0.05	59 ± 11	2.4 ± 0.8	72.0 ± 5.6	3.1 ± 1.3
9	50	5.8 ± 0.4	0.76 ± 0.05	59 ± 10	2.7 ± 0.8	73.3 ± 6.4	3.3 ± 1.3
10	60	6.1 ± 0.4	0.85 ± 0.05	56 ± 10	2.9 ± 0.9	72.2 ± 6.1	3.4 ± 1.2
11	70	6.5 ± 0.4	0.9 ± 0.1	55.4 ± 8.9	3 ± 1	71.9 ± 6.4	3.6 ± 1.2
12	80	6.8 ± 0.4	1.01 ± 0.06	54.1 ± 8.3	3.4 ± 1.1	71.5 ± 6.7	3.7 ± 1.2

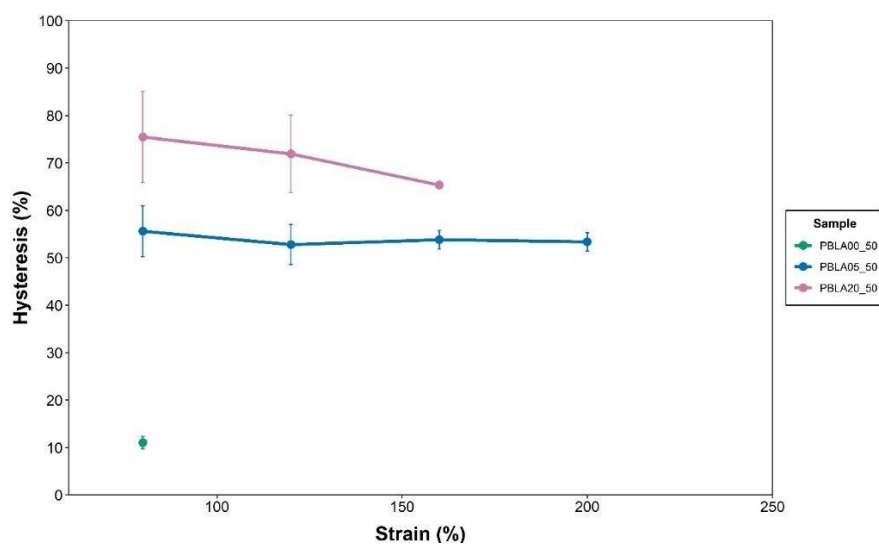


Figure S12. Hysteresis for the plastic cyclic tests for PBLA00_50, PBLA05_50, and PBLA20_50 samples. (Mean values of n= 4 samples, error = SD)

Table S7. Hysteresis and maximum stress for plastic cyclic tests for PBLA00_50, PBLA05_50, and PBLA20_50 samples. (Mean values of n= 4 samples, error = SD)

Cycle	Strain (%)	PBLA00_50		PBLA05_50		PBLA20_50	
		Hysteresis (%)	Max. Stress (Mpa)	Hysteresis (%)	Max. Stress (Mpa)	Hysteresis (%)	Max. Stress (Mpa)
1	80	11.1 ± 1.3	1.00 ± 0.09	56 ± 5	2.6 ± 0.2	75.48 ± 9.64	3.63 ± 0.59
2	120	- ^a	-	53 ± 4	3.3 ± 0.3	71.90 ± 8.19	4.26 ± 0.62
3	160	-	-	54 ± 2	4.1 ± 0.1	65.32 ± - ^b	4.41 ± -
4	200	-	-	53 ± 2	4.8 ± 0.3	-	-
5	240	-	-	-	-	-	-
6	280	-	-	-	-	-	-
7	320	-	-	-	-	-	-
8	360	-	-	-	-	-	-

^aData not available, all samples were measured until the sample breaks

^bSD error not available as only one sample was measured for this cycle.

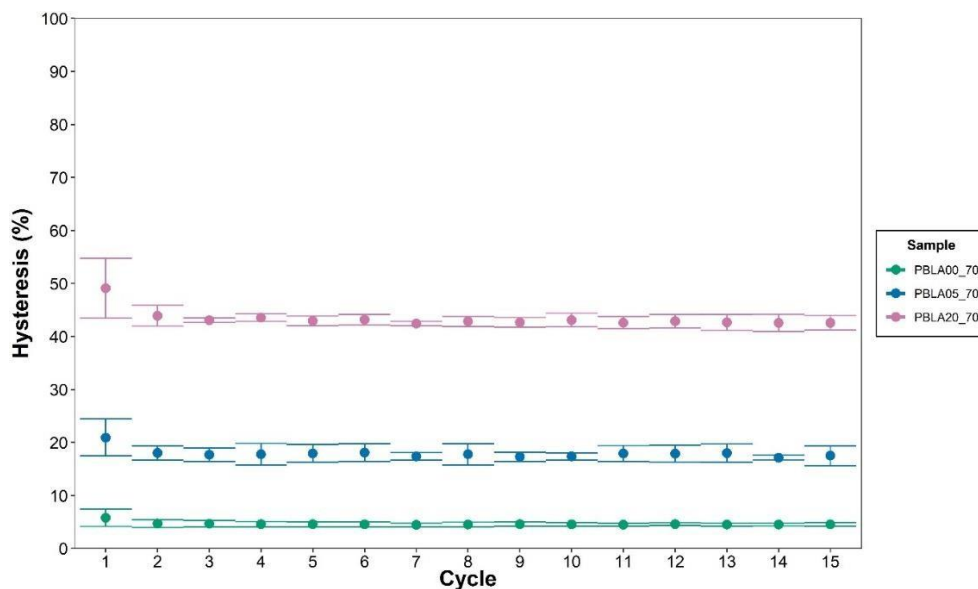


Figure S13. Hysteresis for cyclic test up to 40% strain for PBLA00_70, PBLA05_70, and PBLA20_70 samples. (Mean values of n= 4 samples, error = SD)

Table S8. Hysteresis and maximum stress for 40 % strain cyclic tests for PBLA00_70, PBLA05_70, and PBLA20_70 samples. (Mean values of n= 4 samples, error = SD)

Cycle	Strain (%)	PBLA00_70		PBLA05_70		PBLA20_70	
		Hysteresis (%)	Max. Stress (Mpa)	Hysteresis (%)	Max. Stress (Mpa)	Hysteresis (%)	Max. Stress (Mpa)
1	40	5.78 ± 1.64	0.54 ± 0.01	20.92 ± 3.48	0.45 ± 0.00	49.09 ± 5.66	0.95 ± 0.02
2	40	4.70 ± 0.71	0.54 ± 0.02	18.04 ± 1.34	0.44 ± 0.01	43.89 ± 1.95	0.94 ± 0.04
3	40	4.67 ± 0.60	0.54 ± 0.02	17.70 ± 1.28	0.44 ± 0.01	43.06 ± 0.44	0.93 ± 0.05
4	40	4.60 ± 0.47	0.54 ± 0.02	17.79 ± 2.04	0.44 ± 0.01	43.56 ± 0.68	0.93 ± 0.06
5	40	4.59 ± 0.46	0.54 ± 0.02	17.95 ± 1.67	0.44 ± 0.01	42.95 ± 0.90	0.92 ± 0.05
6	40	4.56 ± 0.47	0.54 ± 0.02	18.10 ± 1.67	0.44 ± 0.01	43.17 ± 0.99	0.92 ± 0.06
7	40	4.46 ± 0.34	0.54 ± 0.02	17.37 ± 0.74	0.44 ± 0.02	42.44 ± 0.44	0.91 ± 0.06
8	40	4.52 ± 0.42	0.54 ± 0.02	17.77 ± 2.00	0.44 ± 0.02	42.84 ± 0.95	0.92 ± 0.06
9	40	4.58 ± 0.40	0.54 ± 0.02	17.30 ± 0.89	0.44 ± 0.02	42.67 ± 0.90	0.91 ± 0.06
10	40	4.55 ± 0.32	0.54 ± 0.02	17.36 ± 0.66	0.43 ± 0.02	43.09 ± 1.27	0.91 ± 0.07
11	40	4.48 ± 0.31	0.54 ± 0.02	17.92 ± 1.50	0.43 ± 0.02	42.61 ± 1.15	0.91 ± 0.07
12	40	4.58 ± 0.26	0.54 ± 0.02	17.91 ± 1.60	0.43 ± 0.02	42.87 ± 1.24	0.91 ± 0.07
13	40	4.51 ± 0.29	0.54 ± 0.02	18.00 ± 1.69	0.43 ± 0.02	42.65 ± 1.51	0.91 ± 0.07
14	40	4.53 ± 0.27	0.54 ± 0.02	17.14 ± 0.42	0.43 ± 0.02	42.57 ± 1.65	0.91 ± 0.08
15	40	4.55 ± 0.35	0.54 ± 0.02	17.52 ± 1.89	0.43 ± 0.02	42.58 ± 1.40	0.91 ± 0.08

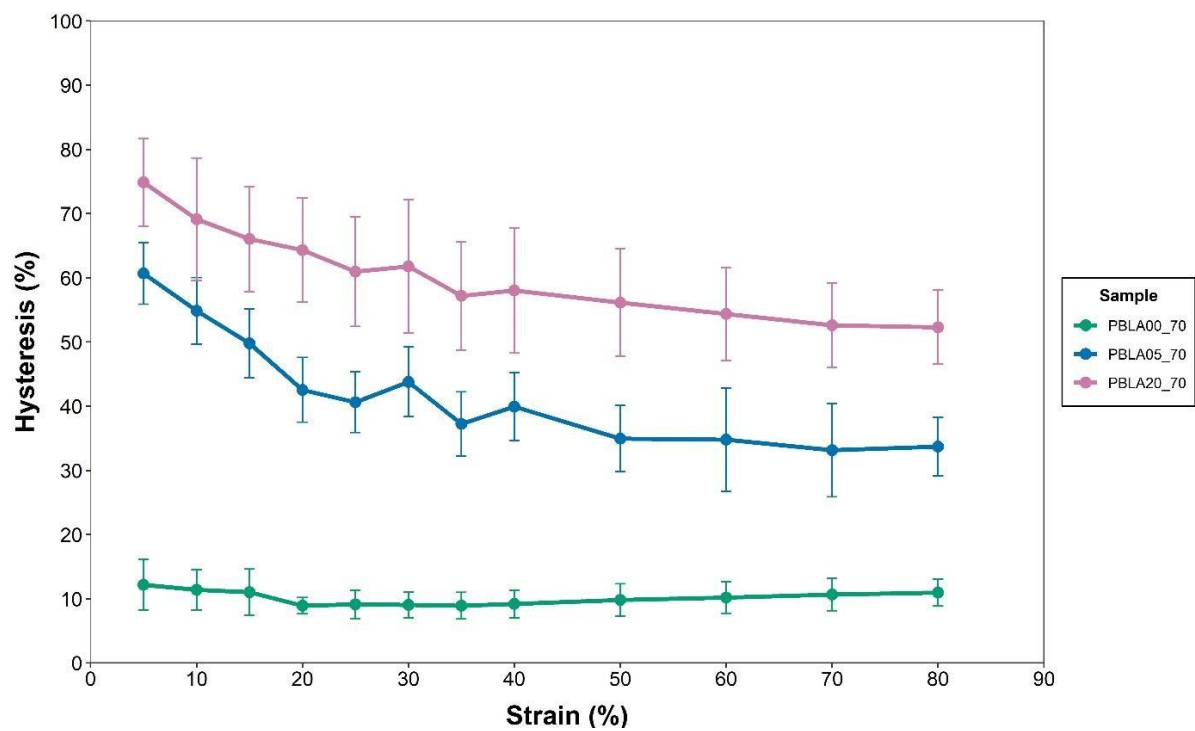


Figure S14. Hysteresis for the elastic cyclic tests for PBLA00_70, PBLA05_70, and PBLA20_70 samples. (Mean values of $n=4$ samples, error = SD)

Table S9. Hysteresis for elastic strain cyclic test for PBLA00_70, PBLA05_70, and PBLA20_70 samples. (Mean values of n= 4 samples, error = SD).

Cycle	Strain (%)	PBLA00_70		PBLA05_70		PBLA20_70	
		Hysteresis (%)	Max. Stress (Mpa)	Hysteresis (%)	Max. Stress (Mpa)	Hysteresis (%)	Max. Stress (Mpa)
1	5	12.18 ± 3.92	0.08 ± 0.01	60.69 ± 4.80	0.13 ± 0.02	74.87 ± 6.83	0.22 ± 0.02
2	10	11.39 ± 3.14	0.15 ± 0.02	54.86 ± 5.18	0.20 ± 0.03	69.12 ± 9.56	0.33 ± 0.03
3	15	11.02 ± 3.59	0.22 ± 0.03	49.78 ± 5.38	0.26 ± 0.04	66.05 ± 8.18	0.42 ± 0.05
4	20	8.92 ± 1.26	0.27 ± 0.04	42.55 ± 5.05	0.32 ± 0.05	64.33 ± 8.06	0.48 ± 0.06
5	25	9.14 ± 2.19	0.32 ± 0.04	40.61 ± 4.71	0.37 ± 0.05	60.97 ± 8.56	0.56 ± 0.07
6	30	9.04 ± 1.98	0.37 ± 0.05	43.78 ± 5.44	0.42 ± 0.06	61.78 ± 10.38	0.63 ± 0.09
7	35	8.93 ± 2.08	0.41 ± 0.06	37.23 ± 4.98	0.48 ± 0.07	57.18 ± 8.44	0.70 ± 0.10
8	40	9.19 ± 2.14	0.46 ± 0.06	39.92 ± 5.31	0.52 ± 0.07	58.03 ± 9.76	0.77 ± 0.12
9	50	9.82 ± 2.50	0.53 ± 0.07	34.95 ± 5.16	0.62 ± 0.09	56.12 ± 8.37	0.91 ± 0.15
10	60	10.19 ± 2.46	0.59 ± 0.07	34.80 ± 8.06	0.71 ± 0.10	54.36 ± 7.25	1.04 ± 0.18
11	70	10.65 ± 2.50	0.64 ± 0.08	33.13 ± 7.28	0.79 ± 0.11	52.62 ± 6.55	1.17 ± 0.19
12	80	10.97 ± 2.11	0.69 ± 0.08	33.72 ± 4.57	0.85 ± 0.11	52.31 ± 5.77	1.27 ± 0.21

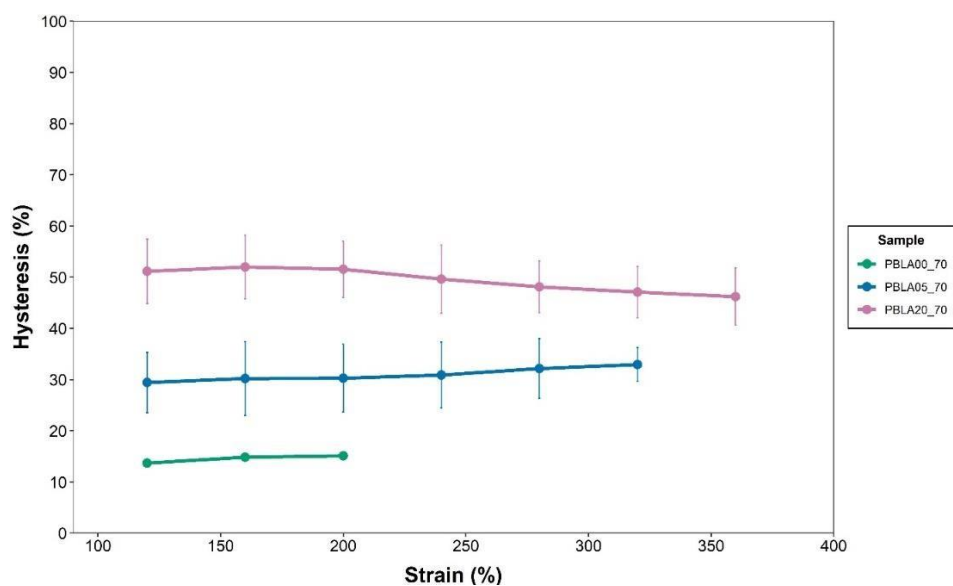


Figure S15. Hysteresis for the plastic cyclic tests for PBLA00_70, PBLA05_70, and PBLA20_70 samples. (Mean values of n= 4 samples, error = SD)

Table S10. Hysteresis for plastic strain cyclic tests for PBLA00_70, PBLA05_70, and PBLA20_70 samples. (Mean values of n= 4 samples, error = SD).

Cycle	Strain (%)	PBLA00_70		PBLA05_70		PBLA20_70	
		Hysteresis (%)	Max. Stress (Mpa)	Hysteresis (%)	Max. Stress (Mpa)	Hysteresis (%)	Max. Stress (Mpa)
1	80	9.07 ± 0.33	0.79 ± 0.03	33.58 ± 2.52	0.83 ± 0.32	53.20 ± 5.32	2.60 ± 0.66
2	120	13.68 ± 0.11	0.96 ± 0.04	29.43 ± 5.92	1.12 ± 0.38	51.13 ± 6.33	3.52 ± 0.67
3	160	14.83 ± NA	1.07 ± - ^b	30.19 ± 7.20	1.37 ± 0.36	51.97 ± 6.22	4.31 ± 0.63
4	200	15.11 ± NA	1.22 ± -	30.26 ± 6.60	1.66 ± 0.31	51.54 ± 5.47	5.08 ± 0.63
5	240	- ^a	-	30.89 ± 6.41	2.01 ± 0.17	49.62 ± 6.68	5.69 ± 0.76
6	280	-	-	32.17 ± 5.80	2.40 ± 0.01	48.11 ± 5.07	6.58 ± 1.07
7	320	-	-	32.93 ± 3.33	2.84 ± 0.10	47.08 ± 5.00	7.71 ± 1.25
8	360	-	-	-	-	46.20 ± 5.59	8.92 ± 1.39

^aData not available, all samples were measured until the sample breaks

^bSD error not available as only one sample was measured for this cycle.

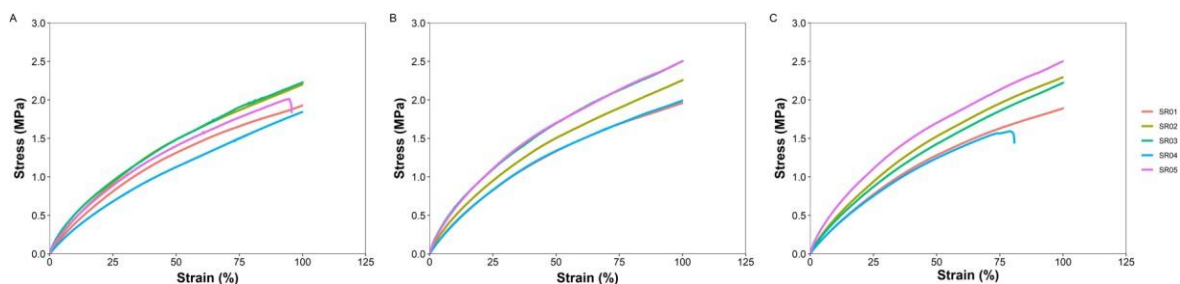


Figure S16. Stress strain curves for self-recovery test for the different cycles. In the legend SRXX refers to each cycle. Results for each tested sample, A) Sample 1, B) Sample 2, C) Sample 3.

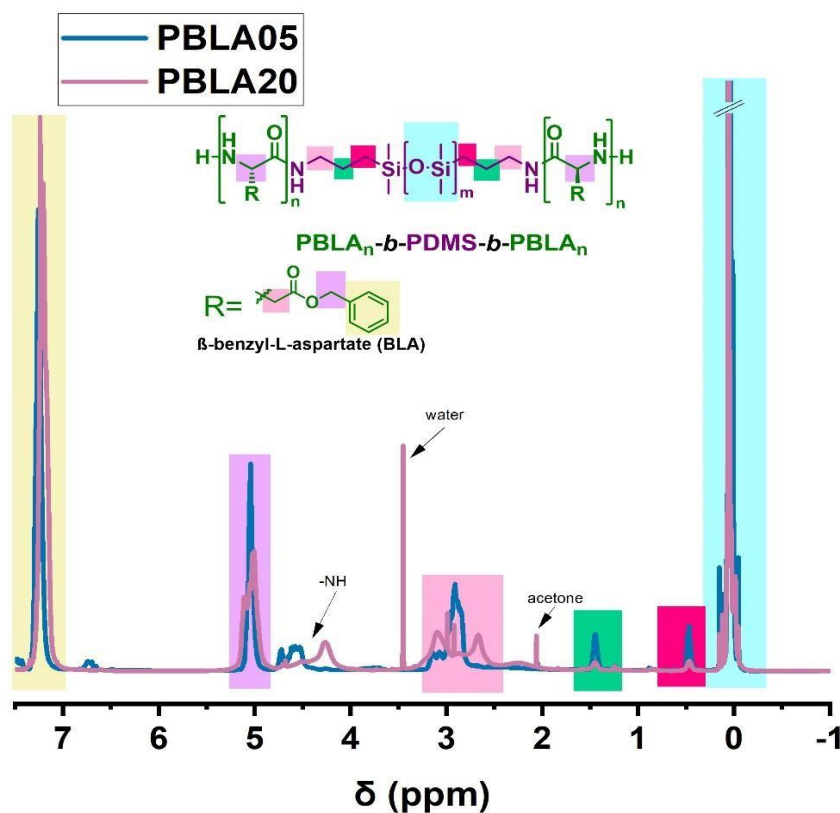


Figure S17. Zoomed-in ^1H -NMR spectra of $\text{PBLA}_x\text{-}b\text{-PDMS-}b\text{-PBLA}_x$ triblock copolymers before functionalization with methacrylate endgroups measured in CDCl_3 . These NMR spectra were used to calculate the number average molecular weight of the PBLA blocks, taking into consideration the known PDMS block size. The peak at δ 4.8-5.3 ppm was integrated. The peak at δ 0 ppm (PDMS methyl groups, m, 254 H) was used as the reference peak. 24H and 96H were obtained for PBLA05 and for PBLA20, respectively. The actual repeat length is 6 and 24 for PBLA05 and PBLA20 triblock copolymers, respectively

Figure S18. GPC elograms of PBLA_x-*b*-PDMS-*b*-PBLA_x triblock copolymers before functionalization with methacrylate endgroups. The measurements were performed in DMAc/LiBr (0.5 wt%) as eluent and PMMA as standard. A molecular weight of 11.7 and 3.7 kg mol⁻¹ was obtained for the PBLA05 and PBLA20 triblock copolymers, respectively.

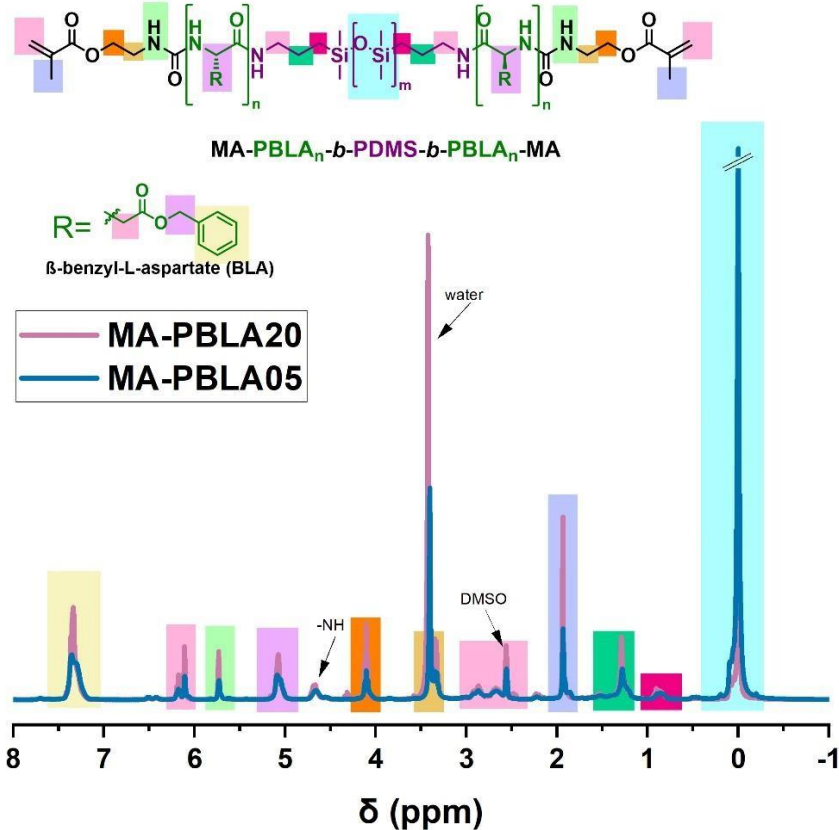


Figure S19. Zoomed-in ¹H-NMR spectra of PBLA_x-*b*-PDMS-*b*-PBLA_x triblock copolymers functionalized with methacrylate endgroups, measured in (CD₃)₂SO.