Table S1. Molar ratio of components in PU-x.

Samples	PCL	PDMS	IPDI	HMBA	DPS
PU-1	0.9	0.1	2	0.5	0.5
PU-2	0.8	0.2	2	0.5	0.5
PU-3	0.7	0.3	2	0.5	0.5
PU-4	0.5	0.5	2	0.5	0.5

Table S2. Molar ratio of components in PUBS-x.

Samples	PCL	PDMS	IPDI	HMBA	DPS
PUBS-1	0.5	0.5	2	0	1
PUBS-2	0.5	0.5	2	0.25	0.75
PUBS-3	0.5	0.5	2	0.5	0.5
PUBS-4	0.5	0.5	2	0.75	0.25

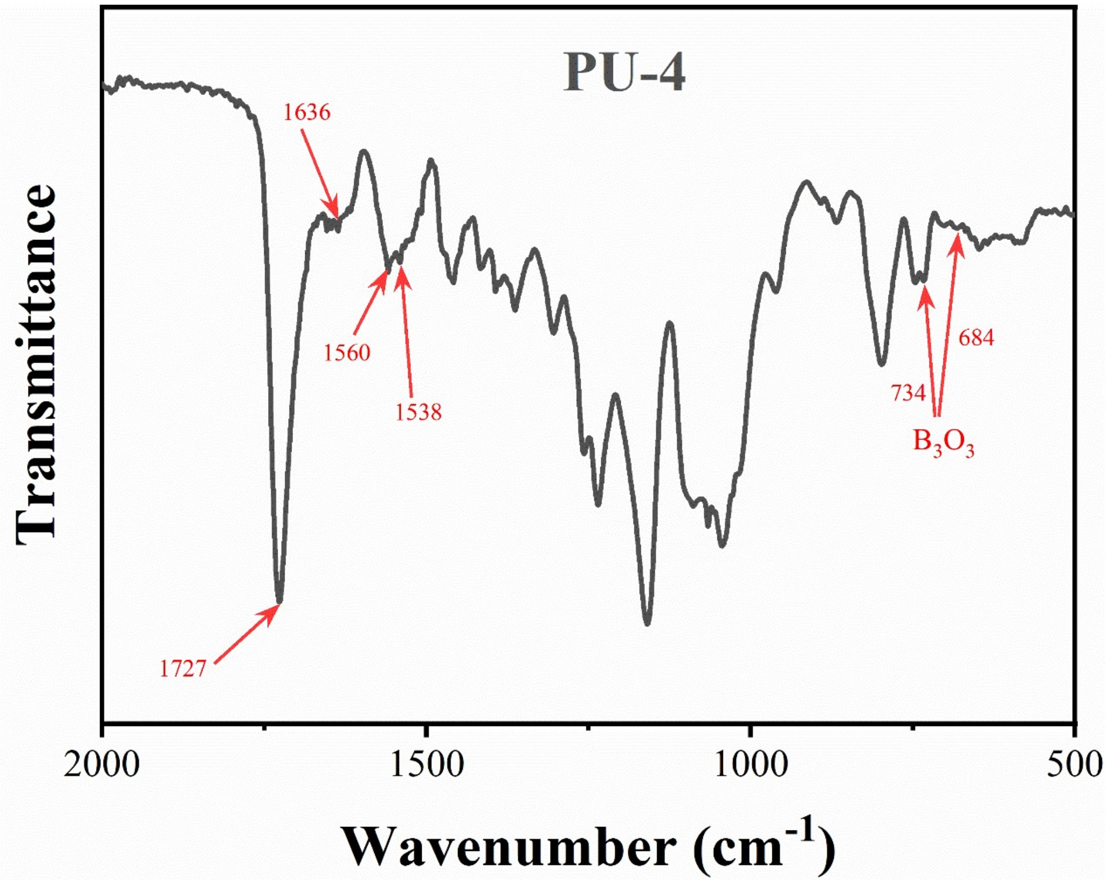

Fig. S1. FTIR spectrum of PU-4.

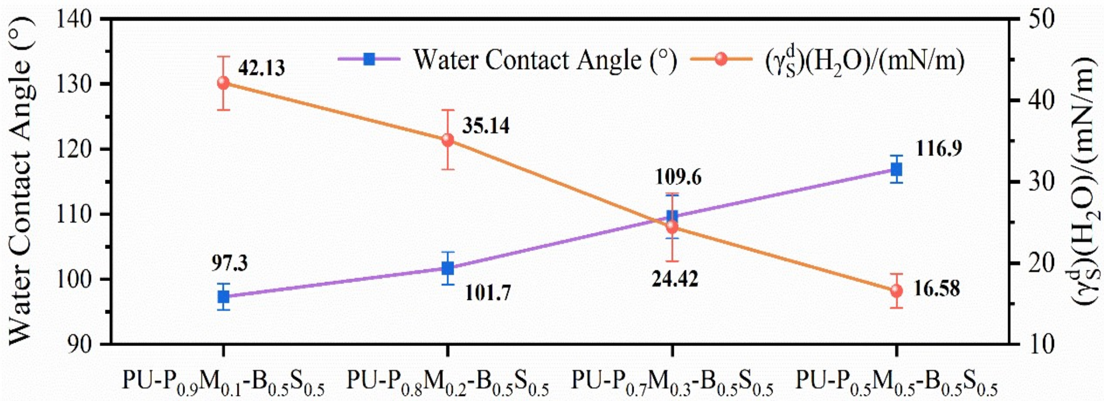

Fig. S2. The contact angle and dispersion surface energy of PU-x elastomers under static contact angle.

Fig. S3. Dynamic contact angle of PU-1.

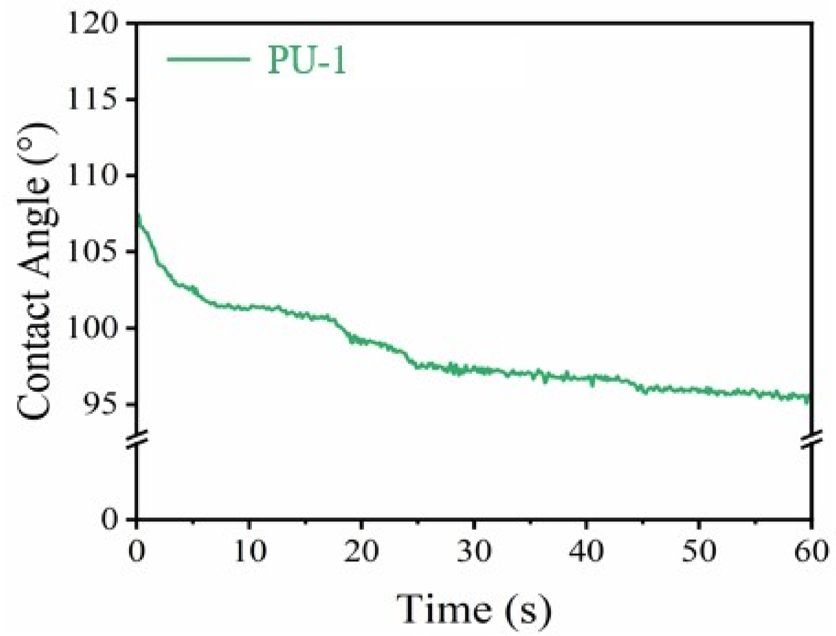

Fig. S4. Image of PU-1 contact angle magnitude as a function of time.

Fig. S5. Dynamic contact angle of PU-3

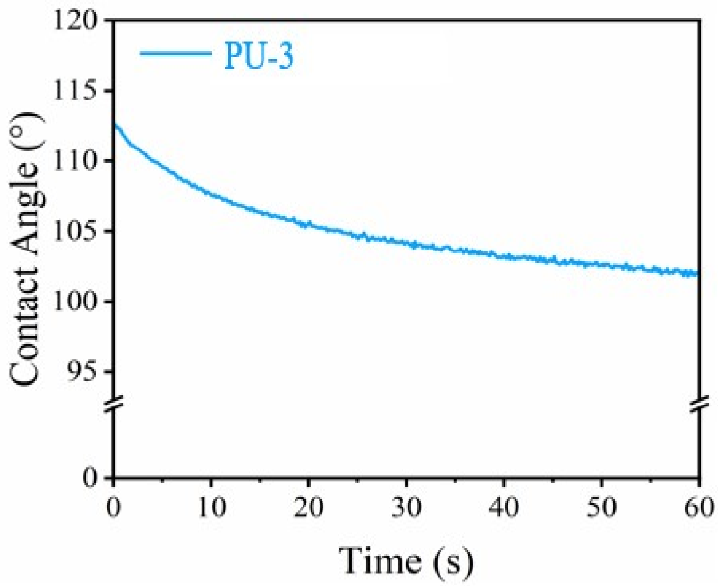

Fig. S6. Image of PU-3 contact angle magnitude as a function of time

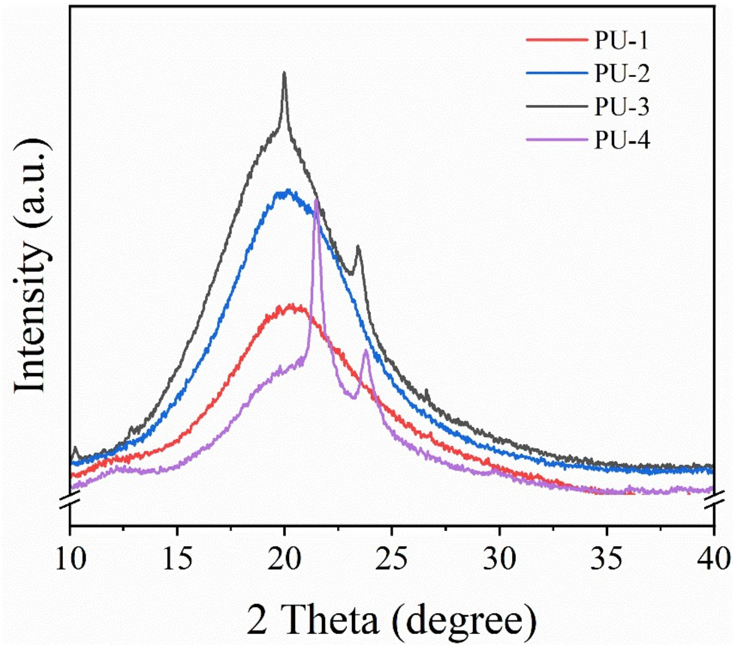

Fig. S7. XRD pattern of PU-x with PCL and PDMS at different ratios.

Table S3.

The melting and crystallization enthalpy (ΔHm, ΔHc) and melting and crystallization temperature (Tm, Tc) and crystallinity (χ_c) of PUs elastomers obtained from DSC.

	ΔH_m(J/g)	ΔH_c(J/g)	T_m (℃)	T_c(℃)	χ_c(%)
PU-1	59.2458	48.8812	51.74	-40.77	45.06
PU-2	49.409815	34.9461	57.53	-43.9	39.86

PU-3	36.51396	28.0878	60.12	-46.3	31.35
PU-4	21.96147	17.204816	64.74	-53.77	21.66

Table S4.
Mechanical property data of PUs elastomers with different ratios of cross-linked PCL and PDMS.

Sample	Tensile strength (MPa)	Elongation at break (%)	Fracture toughness (MJ · m^{-3})
PU-1	9.36	1932.52	60.28
PU-2	7.61	2533.86	101.76
PU-3	10.60	2770.15	142.02
PU-4	16.26	3300.84	278.82

Table S5.
Mechanical property data of PUs elastomers with different ratios of cross-linked structures PCL and PDMS.

Sample	Tensile strength (MPa)	Elongation at break (%)	Fracture toughness (MJ · m^{-3})
PUBS-1	12.05	3793.12	227.54
PUBS-2	14.19	3497.24	263.71

PUBS-3	16.26	3302.31	280.31
PUBS-4	16.90	2779.06	295.63

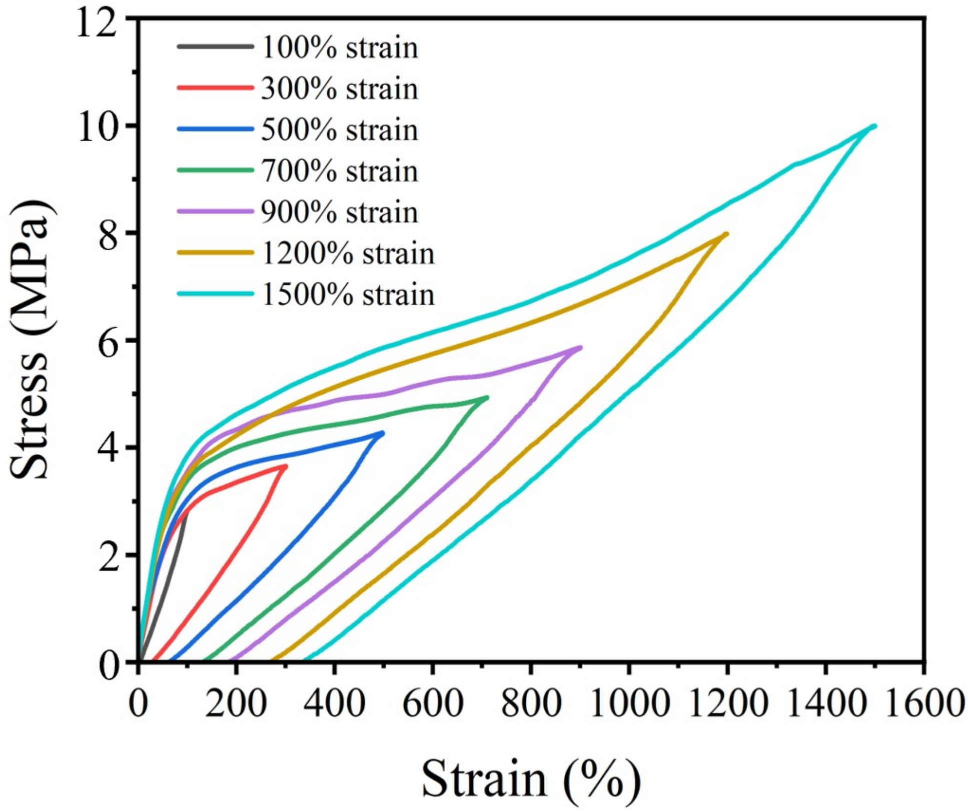

Fig.S8. Cyclic tensile curves of PU-4 with increasing strain in loading-unloading cycle.

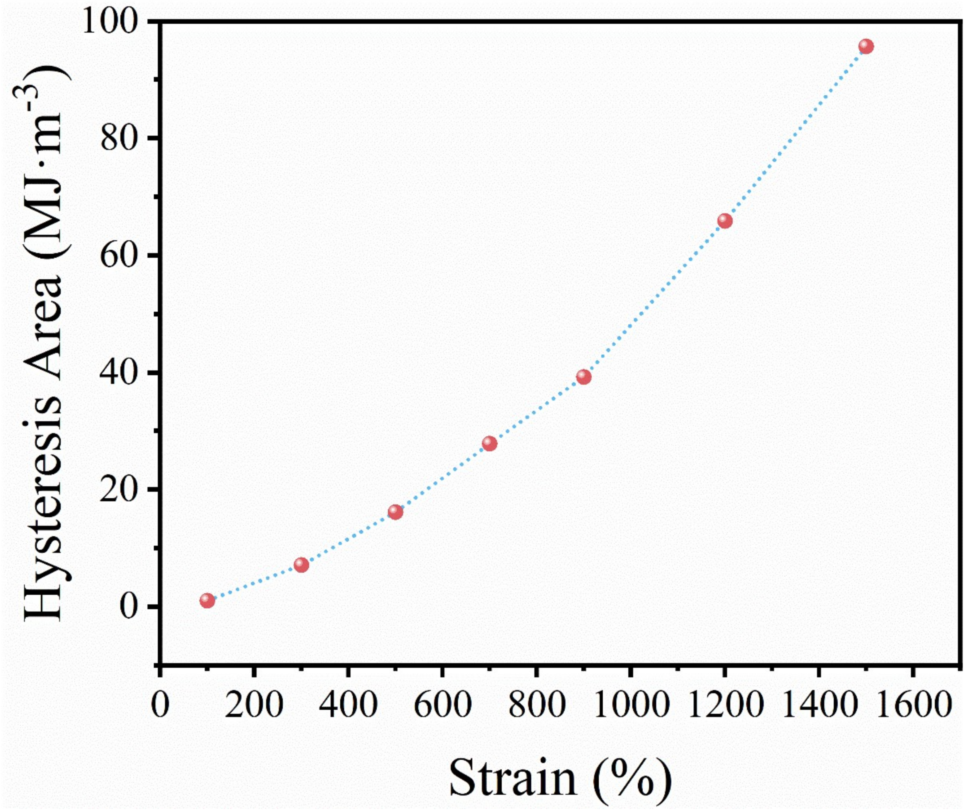

Fig. S9. The hysteresis area corresponding to the cycle period under different strains

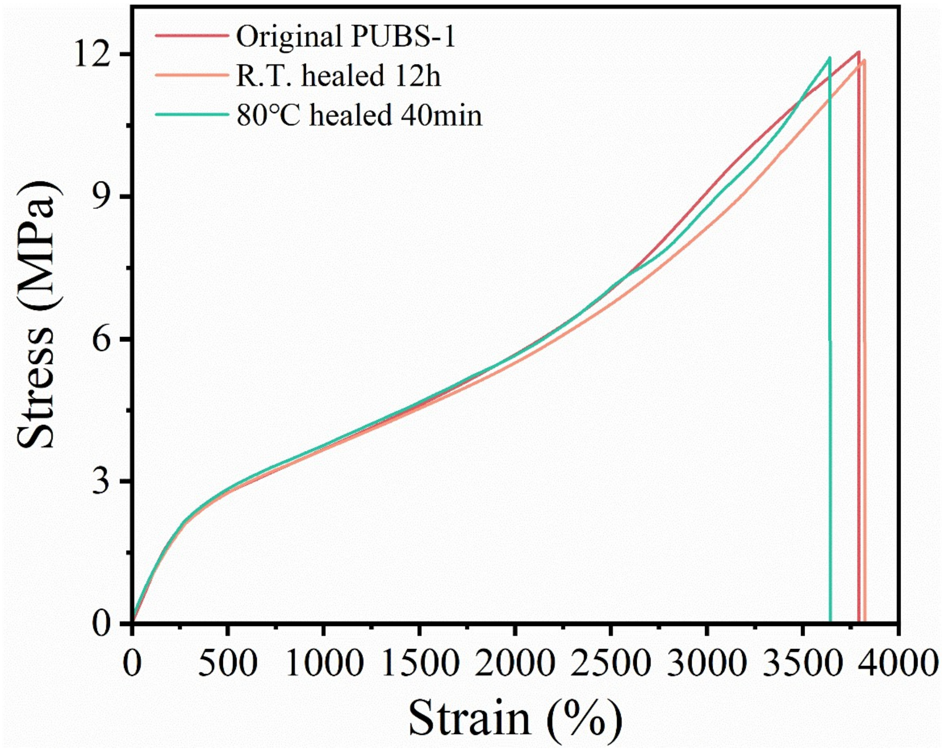

Fig. S10. Stress-strain curves of PUBS-1 after self-healing at room temperature and 80°C.

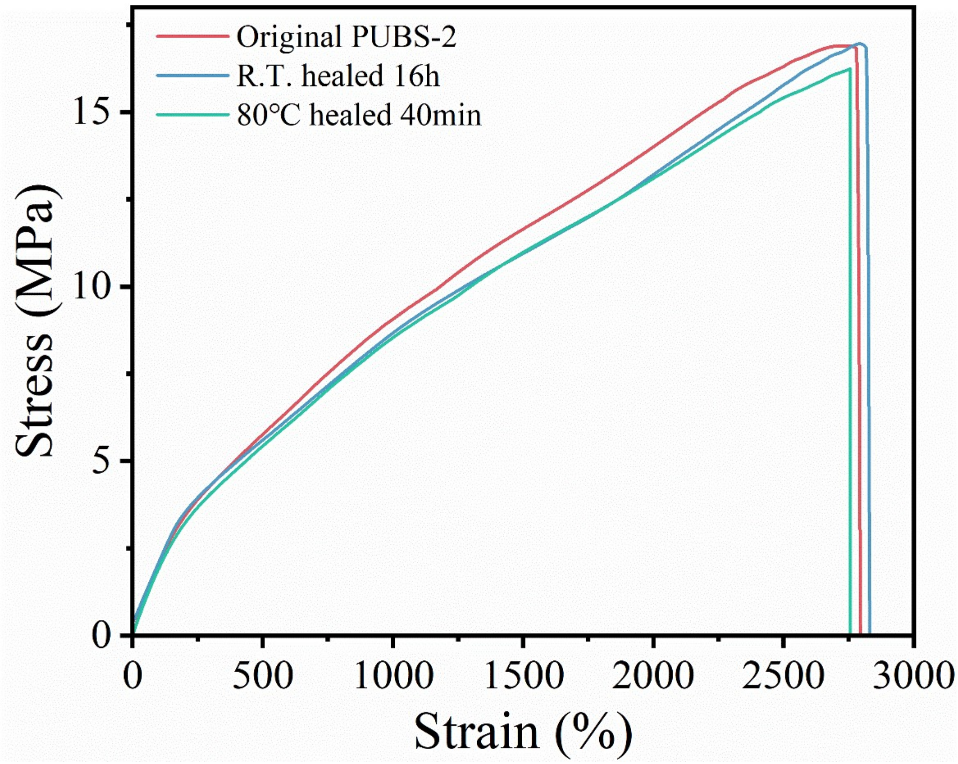

Fig. S11. Stress-strain curves of PUBS-2 after self-healing at room temperature and 80°C.

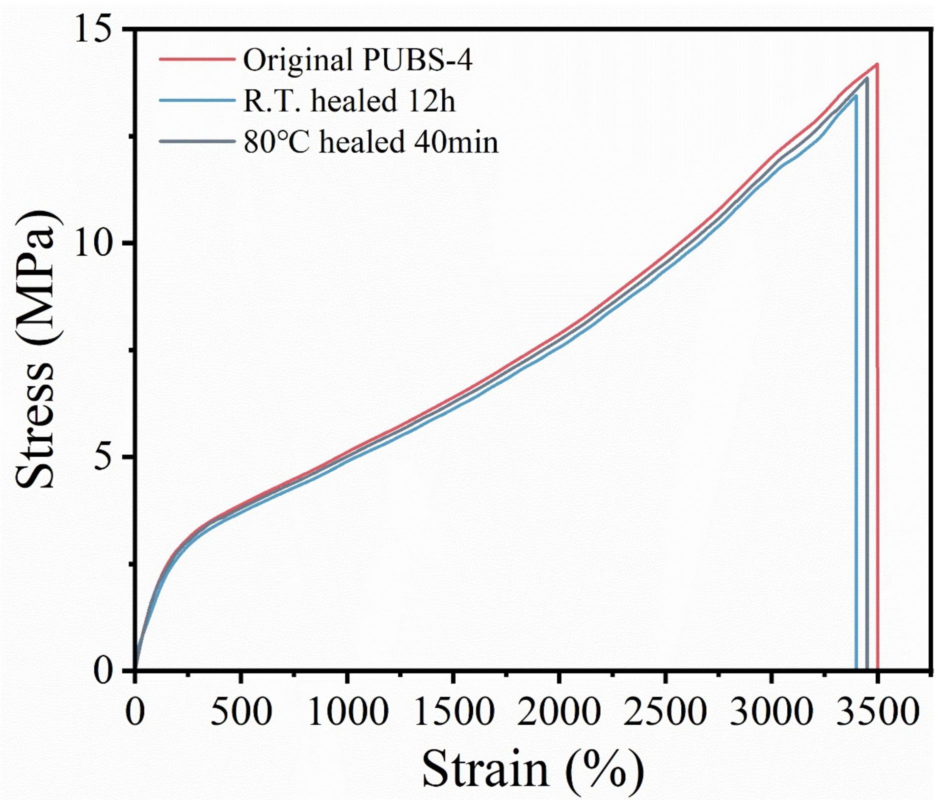

Fig. S12. Stress-strain curves of PUBS-4 after self-healing at room temperature and 80°C.

Table S6.
Mechanical properties and healing efficiency of PUBS-1 after self-healing under different conditions.

Sample	Self-healing condition	Tensile strength (MPa)	Elongation at break (%)	Self-healing efficiency (%)
PUBS-1	Original	12.05	3793.12	-
	R.T. healed 3h	7.61	2857.34	63.12
	R.T. healed 6h	9.13	3182.76	75.77
	R.T. healed 12h	11.42	3821.32	94.78
	80°C healed 20min	10.02	3440.21	83.40
	80°C healed 40min	11.92	3696.33	98.92

Table S7.
Mechanical properties and healing efficiency of PUBS-2 after self-healing under different conditions.

Sample	Self-healing condition	Tensile strength (MPa)	Elongation at break (%)	Self-healing efficiency (%)
	Original	14.19	3497.12	-
	R.T. healed 3h	8.46	2239.89	59.59
	R.T. healed 6h	12.10	2977.48	85.27

	R.T. healed 12h	13.98	3396.60	98.64
	80℃ healed 20min	9.98	2629.03	70.33
	80℃ healed 40min	14.12	3448.68	99.54

Table S8.
Mechanical properties and healing efficiency of PUBS-3 after self-healing under different conditions.

Sample	Self-healing condition	Tensile strength (MPa)	Elongation at break (%)	Self-healing efficiency (%)
PUBS-3	Original	16.26	3302.31	-
	R.T. healed 3h	9.09	2635.16	55.91
	R.T. healed 6h	12.38	2781.81	76.13
	R.T. healed 12h	16.27	3414.08	100
	80℃healed 20min	12.41	2974.97	76.32
	80℃ healed 40min	16.30	3388.38	100

Table S9.
Mechanical properties and healing efficiency of PUBS-4 after self-healing under different conditions.

Sample	Self-healing condition	Tensile strength (MPa)	Elongation at break (%)	Self-healing efficiency (%)
PUBS-4	Original	16.90	2779.06	-
	R.T. healed 4h	9.48	1491.17	56.13
	R.T. healed 8h	12.70	2084.12	75.15
	R.T. healed 16h	16.92	2821.12	100
	80℃ healed 20min	11.20	1896.19	66.27
	80℃ healed 40min	16.24	2755.50	96.09

Table S10.
Comparison of mechanical properties and self-healing efficiency of PUBS-3 with the reported values of the PU as materials in references.

No.	Tensile strength (MPa)	Elongation at break(%)	Strength self-healing efficiency (%)	Self-healing temperature/time	Resource
1	16.26	3302.31	100	25 ºC/12 h	This work
2	4.8	2100	~100	25 ºC/48 h	1

No.	Tensile strength (MPa)	Elongation at break(%)	Strength self-healing efficiency (%)	Self-healing temperature/time	Resource
3	4.5	912	97	60 ºC/2 h	2
4	~1.7	700	95	25 ºC/24 h	3
5	52.4	2250	99	100 ºC/3 h	4
6	29	1806	81.3	25 ºC/24 h	5
7	3.64	2800	99	25 ºC/12 h	6
8	25.0	1600	85	70 ºC/24 h	7
9	14.2	~500	95	25 ºC/24 h	8
10	21.8	1600	~100	25 ºC/24 h	9
11	24.8	2144	94	40 ºC/24 h	10
12	~1.0	14000	93	25 ºC/2 h	11
13	34.1	2014	83	25°C/48h	12
14	10.5	3120	82	25°C/36h	13
15	25.1	710	97.2	90°C/1h	14

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