

Supporting Information

Self-healing and mechanically robust poly(thiourea-disulfide) elastomers based on synergistic tri-dynamic bondings

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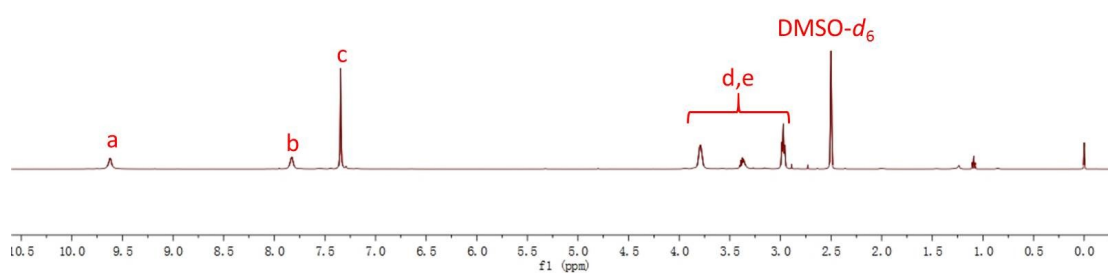
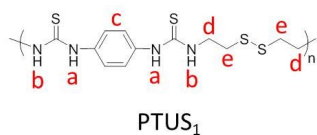


Fig. S1 ¹H NMR of PTUS₁.

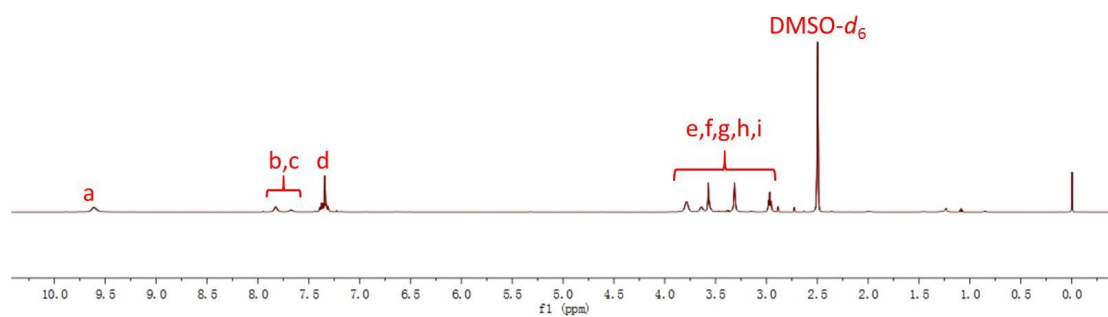
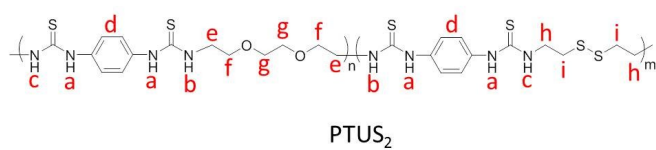


Fig. S2 ¹H NMR of PTUS₂.

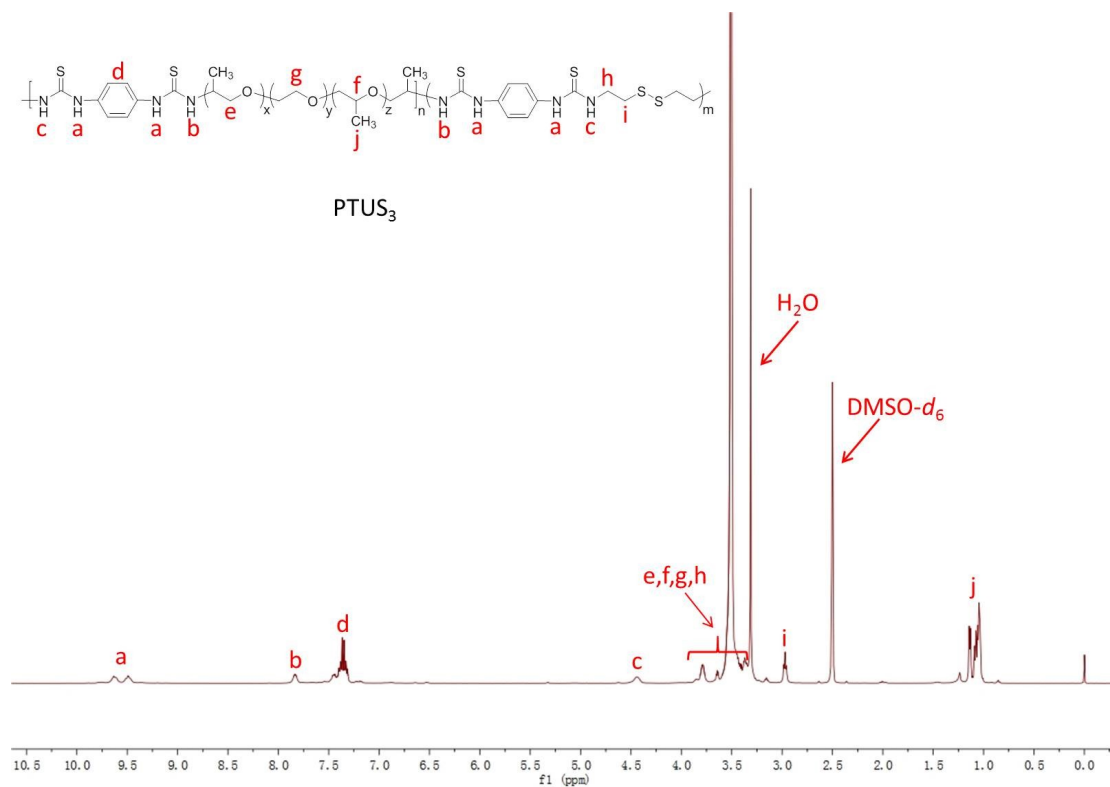


Fig. S3 ¹H NMR of PTUS₃.

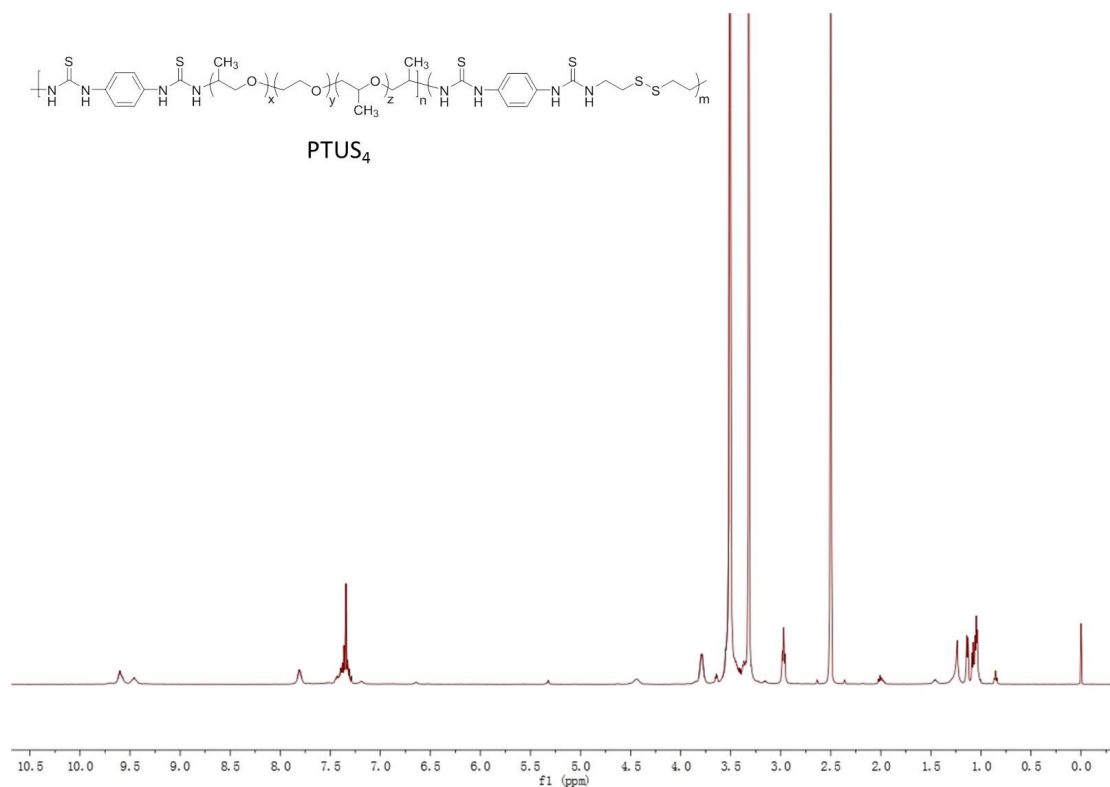


Fig. S4 ¹H NMR of PTUS₄.

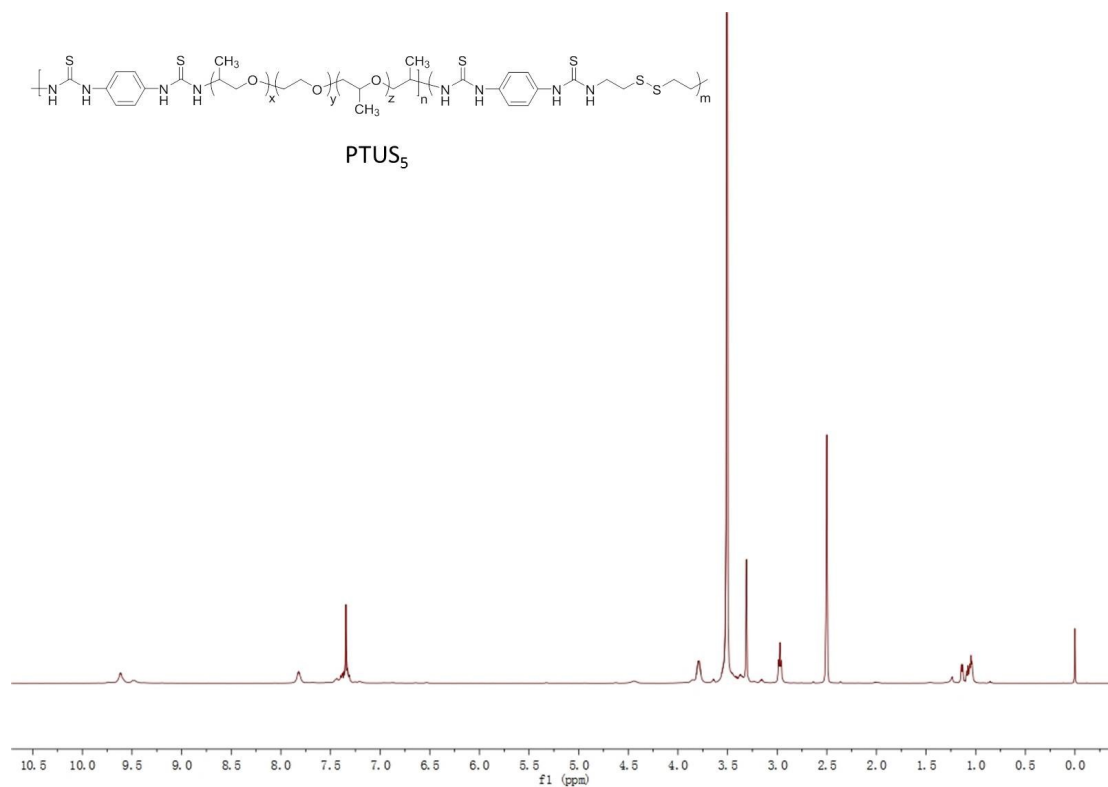


Fig. S5 ¹H NMR of PTUS₅.

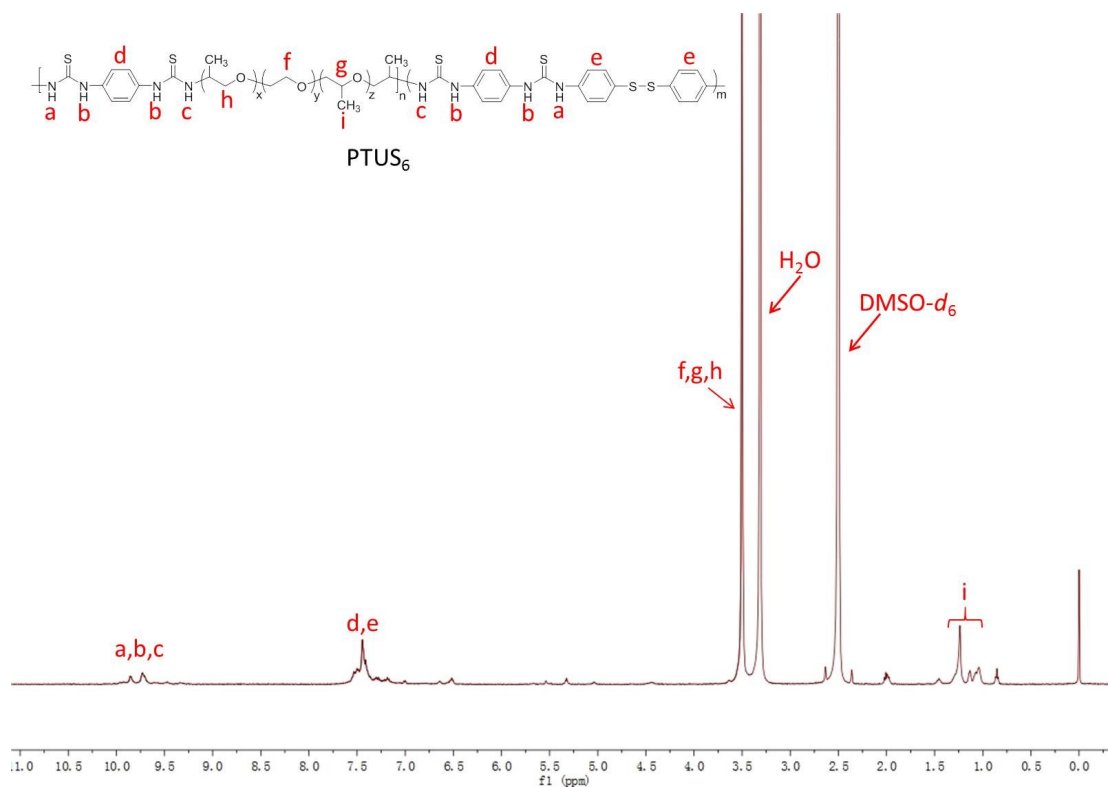


Fig. S6 ¹H NMR of PTUS₆.

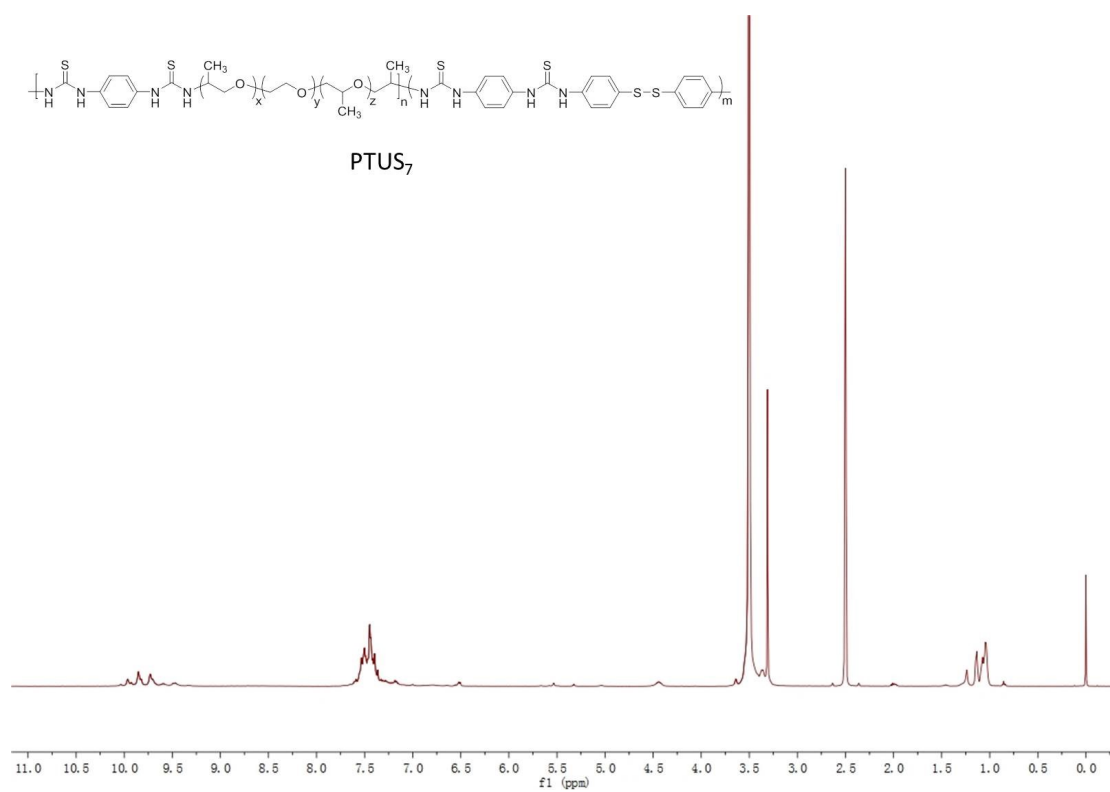


Fig. S7 ¹H NMR of PTUS₇.

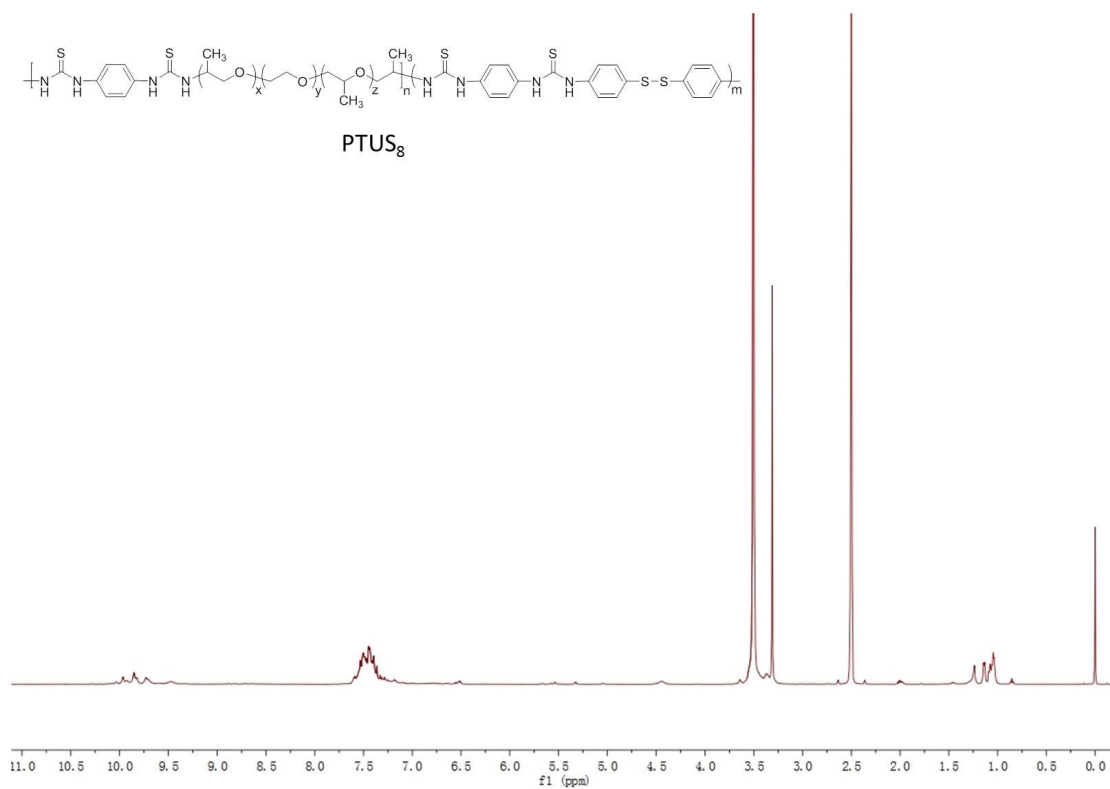


Fig. S8 ¹H NMR of PTUS₈.

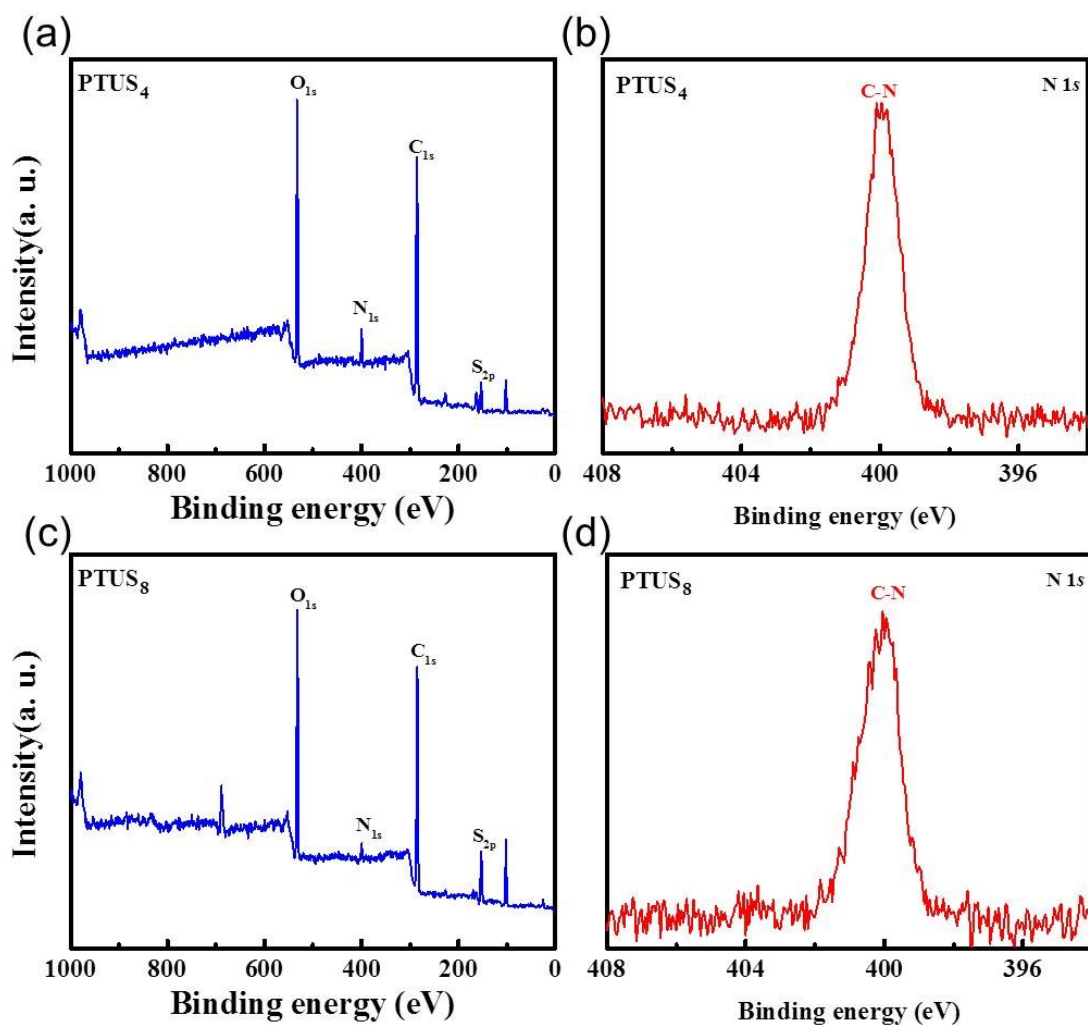


Fig. S9 XPS spectra of PTUS₄ and PTUS₈: (a) XPS survey scan of PTUS₄, (b) C-N of PTUS₄, (c) XPS survey scan of PTUS₈, (d) C-N of PTUS₈.

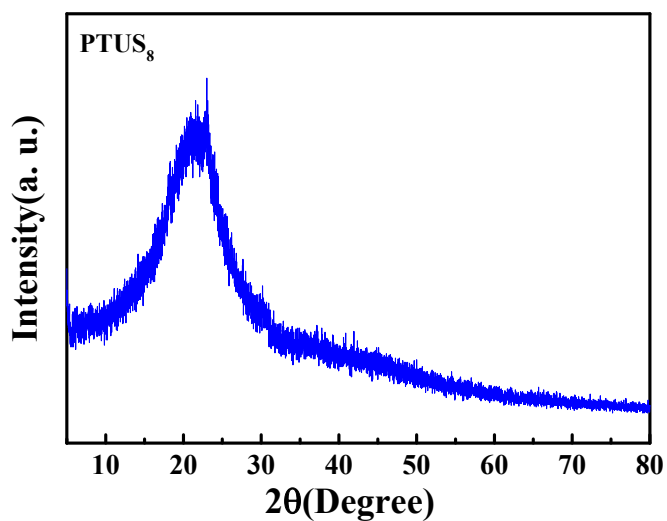


Fig. S10 XRD curve of PTUS₈.

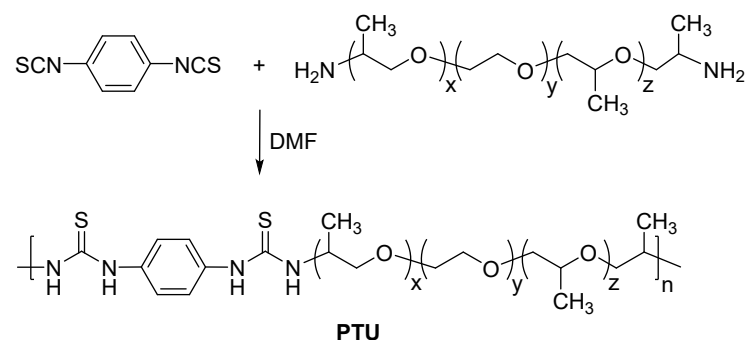
Table S1. Summary of mechanical properties of PTUSx.

Samples	Tensile strength (MPa)	Elongation at break (%)	Young's modulus (MPa)	Toughness (MJ m ⁻³)
PTUS ₄	0.33	167	1.37	47.17
PTUS ₅	0.12	114	0.70	4.54
PTUS ₇	2.55	767	3.57	507.05
PTUS ₈	12.9	800	26.7	884.6

Table S2. Summary of self-healing and mechanical properties of polyurethane elastomers reported in the literatures.

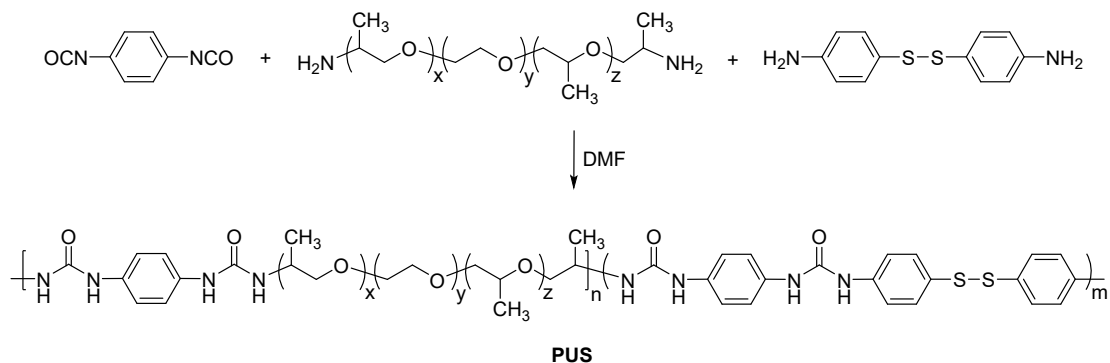
	Tensile strength (MPa)	Healing efficiency (%)	Elongation at break (%)	Ref.
1	12.9	96 (48h)	800	This work
2	4.54	59.1 (24h)	1586	1
3	0.1	93 (2h)	14000	2
4	0.22	86 (48h)	1880	3
5	24.81	93 (7h)	860	4
6	16	100 (1h)	126	5
7	40	97 (24h)	1072	6
8	0.93	87 (12h)	300	7
9	25	86.4 (6h/70 °C)	1600	8

Synthesis of PTU:



The synthesis process of PTU: PDIT (384.8 mg, 2.0 mmol) was slowly added to a Jeffamine solution (3.8 g, 2.0 mmol in 5 mL DMF) under N₂ atmosphere. After stirred at 60 °C for 48 h, the mixture was cooled to room temperature and slowly dropped into ice ethyl ether (200 mL). The precipitate was collected and dissolved with dichloromethane (DCM, 50 mL) and then reprecipitated twice in ice ethyl ether. The final product was dried in an oven at 80 °C for 24 h to obtain PTU.

Synthesis of PUS:



The synthesis process of PUS: ADS (745.1 mg, 3.0 mmol) and Jeffamine (1.9 g, 1.0 mmol) were firstly dissolved in acetone (15 mL) in a 100 mL three-necked flask. Next, 1,4-phenylene diisocyanate (640.5 mg, 4.0 mmol) was added to the three-necked flask under N₂ atmosphere. The mixture was stirred at 40 °C for 48 h. Finally, the solution was casted into a teflon mold and volatilized at 60 °C for 24 h. After vacuum drying at 60 °C for 12 h to obtain PUS.

References

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