Supporting Information

Ultra-Robust, Highly Stretchable and Ambient Temperature Rapid Self-Healing Polyurethane/Graphene Elastomers Enabled by Multi-Type Hydrogen Bonds

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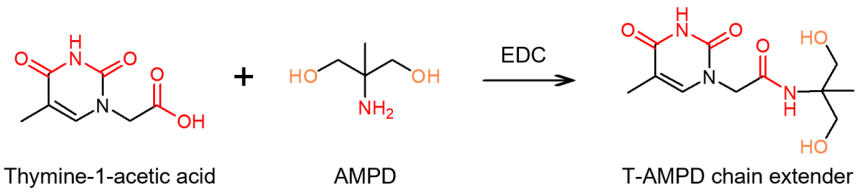

Figure S1. Synthesis route of T-AMPD chain extender.

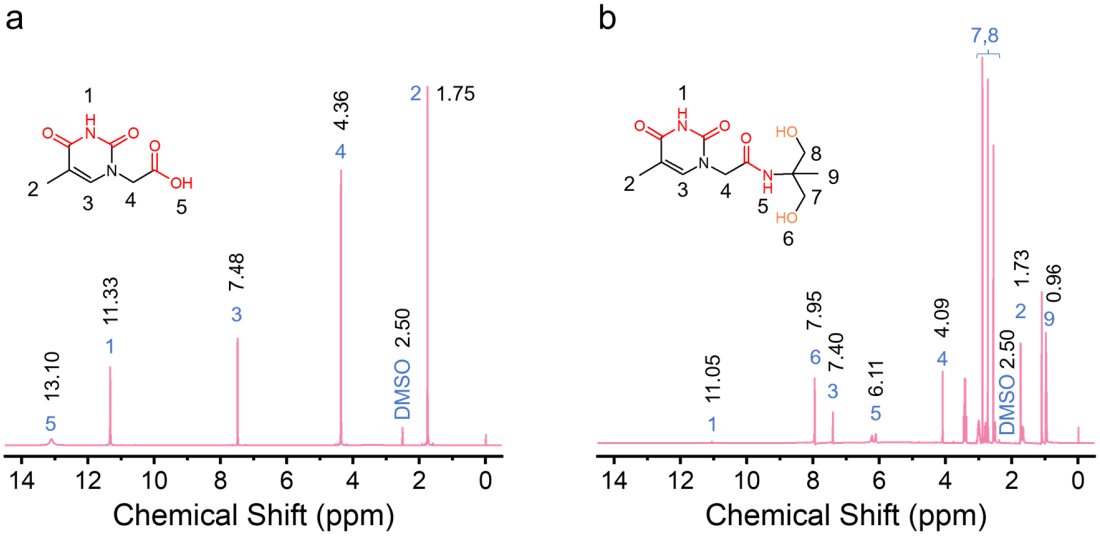

Figure S2. ^{1}H NMR spectra (400 MHz, DMSO): (a) Thymine-1-acetic acid and (b) T-AMPD chain extender.

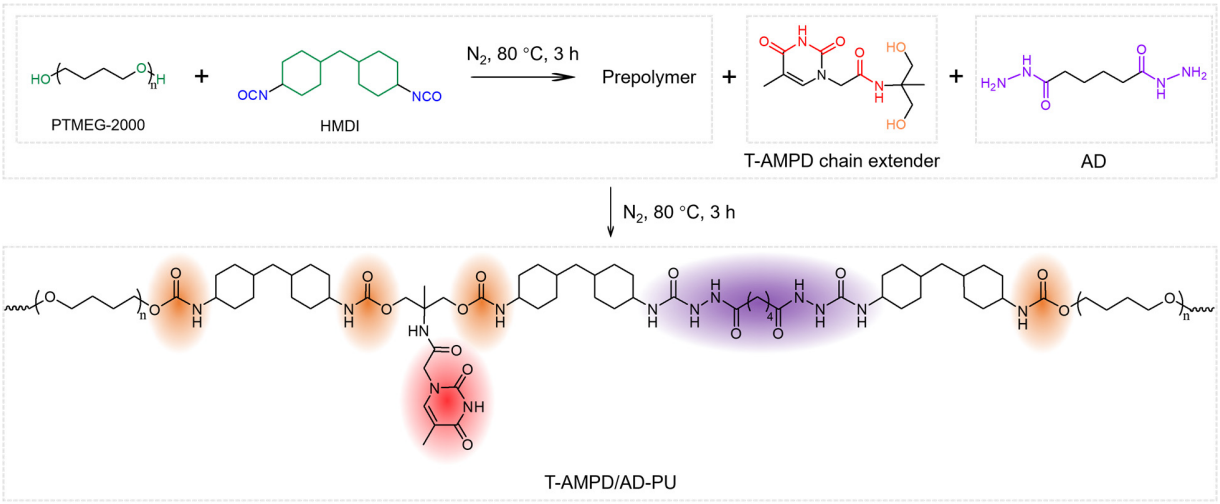

Figure S3. Synthesis route of T-AMPD/AD-PU.

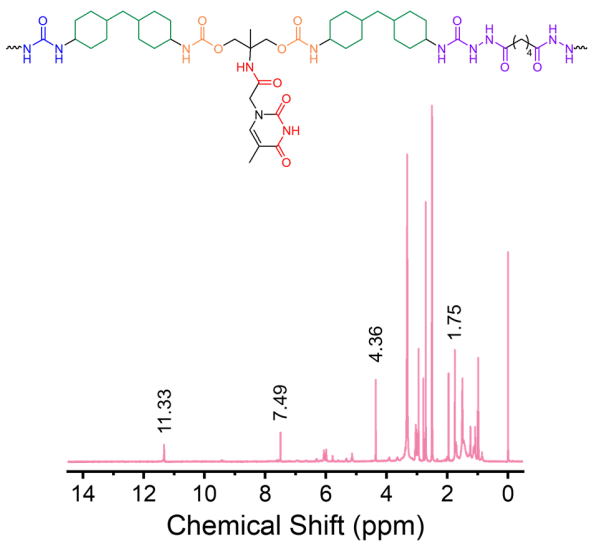

Figure S4. ^{1}H NMR spectra (400 MHz, DMSO) of T-AMPD/AD-PU.

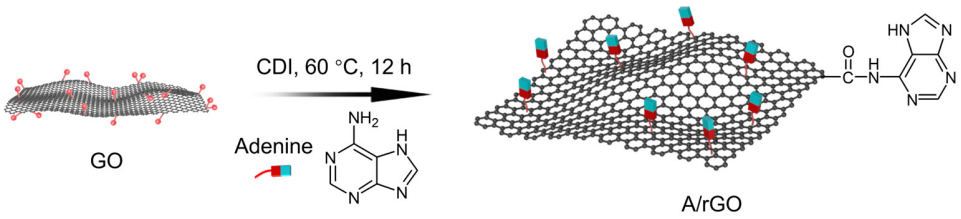

Figure S5. Schematic diagram of the preparation process of A/rGO nanosheets.

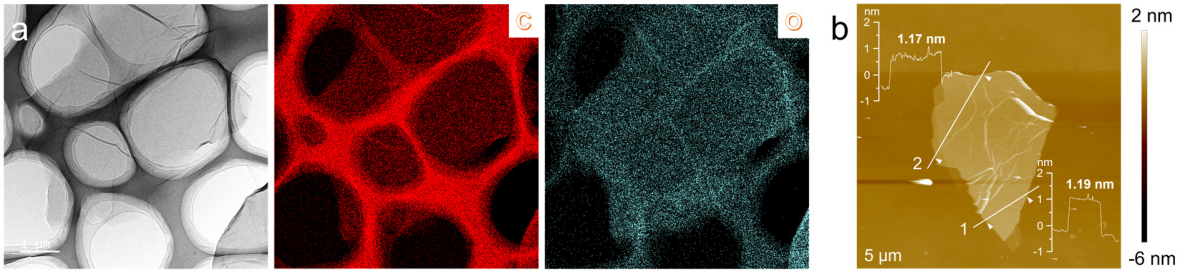

Figure S6. (a) TEM-EDS and (b) SPM images of GO nanosheet.

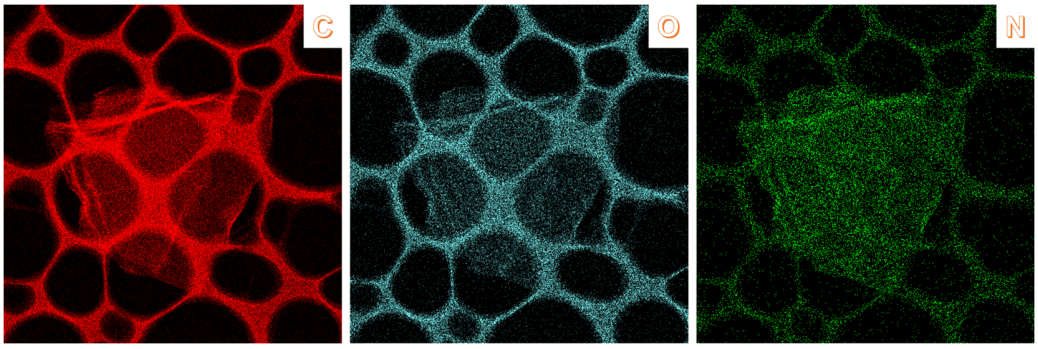

Figure S7. TEM-EDS images of A/rGO nanosheet.

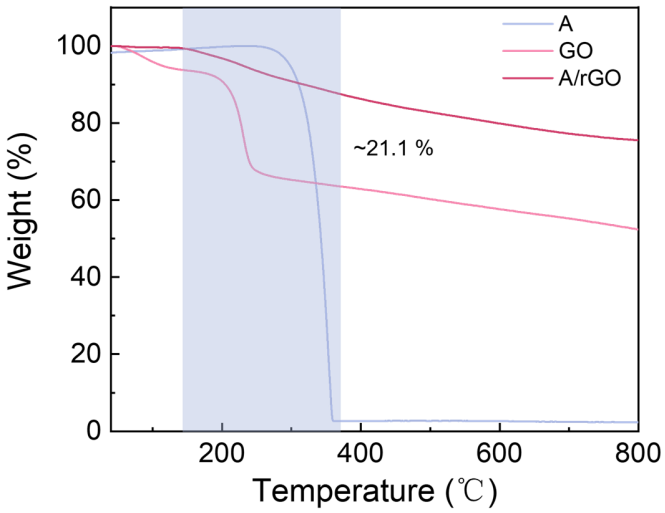

Figure S8. TG curves of samples.

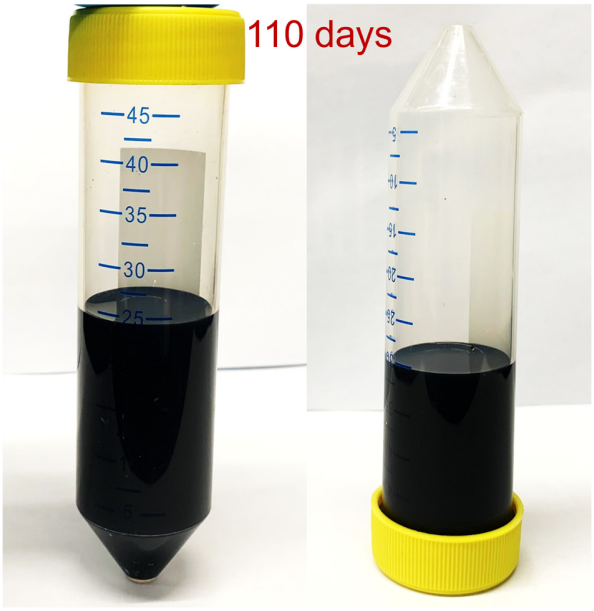

Figure S9. Optical photographs of A/rGO nanosheets dispersed in water for 110 days

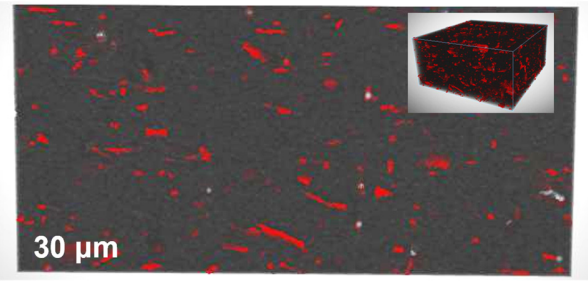

Figure S10. Micro-CT images of T/AGs-0.5%-PU.

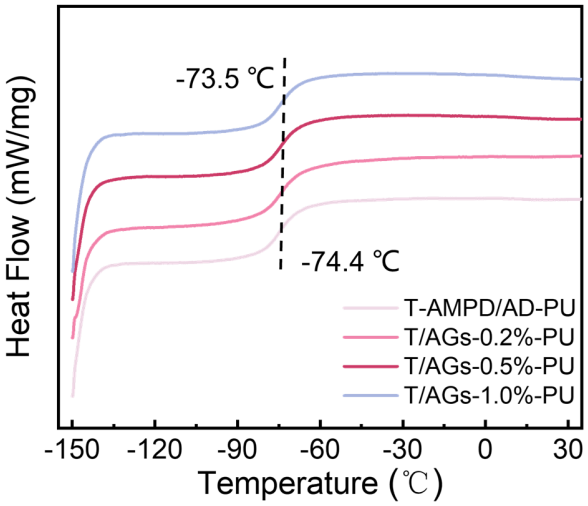

Figure S11. DSC curves of PU composites.

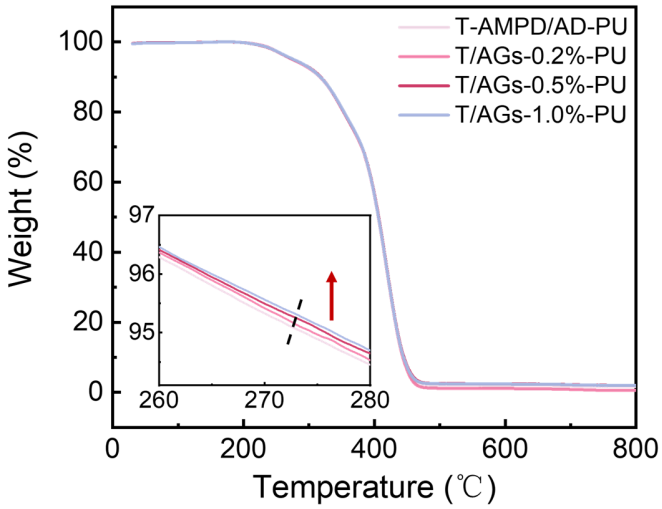

Figure S12. TG curves of PU composites.

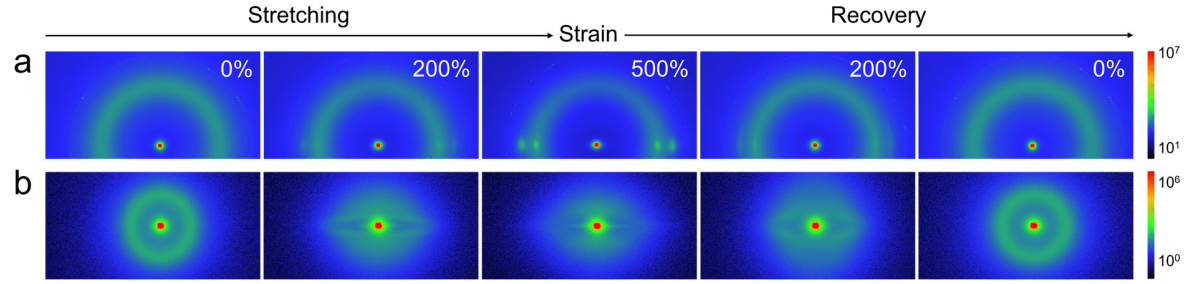

Figure S13. (a) 2D WAXS and (b) 2D SAXS patterns for T/AGs-0.5%-PU at different stretch-release strains.

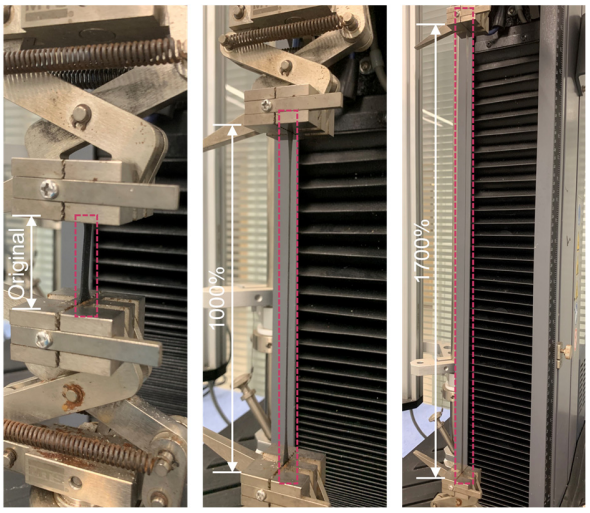

Figure S14. Stretch testing photographs of T/AGs-0.5%-PU.

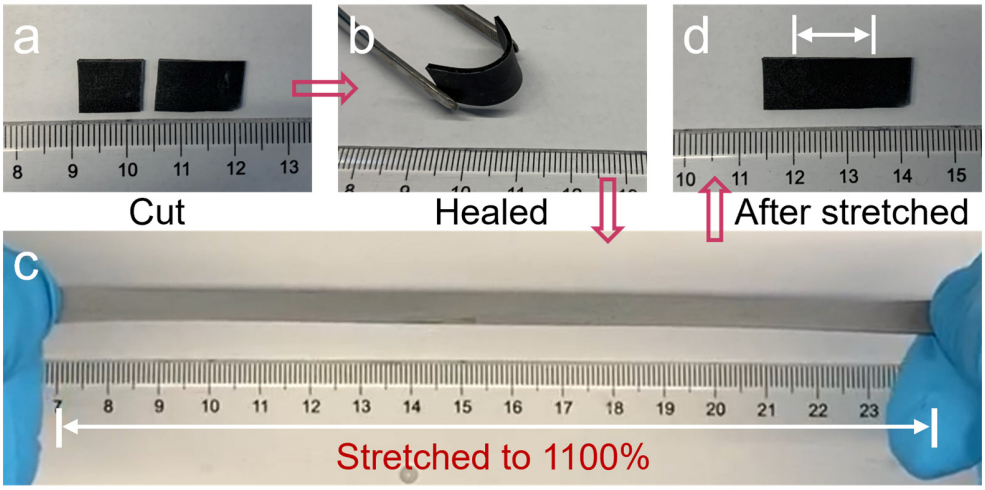

Figure S15. Photographs showing the detailed self-healing process of T/AGs-0.5%-PU: (a) cut, (b) healed, (c) stretched to 1100%, and (d) after stretched.

Table S1 Comparison of ultimate tensile strength, elongation at break, toughness, and Young's modulus of PU composites.

Samples	Ultimate tensile strength (MPa)	Elongation at break (%)	Toughness (MJ m^{-3})	Young's modulus (MPa)
T-AMPD/AD-PU	30.29 ± 2.41	1631.39 ± 17.24	215.31 ± 28.23	17.42 ± 0.72
T/AGs-0.2%-PU	36.79 ± 1.84	1653.82 ± 20.13	245.34 ± 27.81	19.82 ± 0.68
T/AGs-0.5%-PU	46.60 ± 2.15	1736.89 ± 18.17	337.19 ± 30.64	24.40 ± 1.15
T/AGs-1.0%-PU	41.89 ± 2.67	1536.73 ± 19.58	292.59 ± 30.92	25.64 ± 1.16

Table S2 Comparison of ultimate tensile strength, elongation at break, self-healing efficiency, and self-healing conditions of PU composites under different self-healing times.

Samples	Ultimate tensile strength (MPa)	Elongation at break (%)	Self-healing efficiency (%)	Self-healing times
T-AMPD/AD-PU	26.40 ± 1.42	1407.68 ± 12.14	87.16 ± 1.84	RT, 2 h
T/AGs-0.2%-PU	31.82 ± 1.67	1427.84 ± 16.87	86.49 ± 1.31	RT, 2 h
T/AGs-0.5%-PU	39.90 ± 1.61	1484.36 ± 17.62	85.62 ± 1.16	RT, 2 h
T/AGs-1.0%-PU	32.79 ± 1.77	1203.47 ± 18.43	78.28 ± 1.48	RT, 2 h
T/AGs-0.5%-PU-0.5 h	18.70 ± 1.14	698.79 ± 10.83	40.13 ± 1.62	RT, 0.5 h
T/AGs-0.5%-PU-1 h	28.75 ± 0.97	1069.21 ± 10.24	61.70 ± 1.87	RT, 1 h
T/AGs-0.5%-PU-1.5 h	35.59 ± 1.79	1322.79 ± 13.47	76.37 ± 1.44	RT, 1.5 h

Table S3 Comparison of the proposed T/AGs-0.5%-PU elastomer with other reported polymers with self-healing ability within 2 h at room temperature.

Ref.	Self-healing motif	Ultimate tensile strength (MPa)	Elongation at break (%)	Toughness (MJ m^{-3})	Young's modulus (MPa)	Self-healing efficiency (%)	Function healing	Self-healing times
1	Disulfides	6.8	923	26.9	1.5	88	Yes	RT. 2 h
2	Disulfides	0.81	3100	13.0	N/A	80	N/A	RT. 2 h
3	Metal-ligand	10	607	46.1	N/A	92	Yes	RT. 5 min
4	H-bond	10.3	596.2	37.8	31.5	90	N/A	RT. 1 h
5	H-bond	1.6	1400	N/A	N/A	88	N/A	RT. 1.5 h
6	H-bond	12.86	1.26	N/A	1560	99	N/A	RT. 1 h
7	Metal-ligand	3.36	623	10.4	30.7	73.2	N/A	RT. 2 h
8	H-bond+ Disulfides	0.096	1320	N/A	N/A	93	Yes	RT. 2 h
9	H-bond+ O-B bond	5.5	2000	30	N/A	97.6	Yes	RT. 2 h
This Work	H-bond	46.60	1736.89	337.19	24.40	85.62	Yes	RT. 2 h

Table S4 Oxygen transmission rates of the PU composites.

Samples	Oxygen transmission rates (m^3 m^{-2} d^{-1})
T-AMPD/AD-PU	$8.39 \times 10^{-3} \pm 3.18 \times 10^{-3}$
T/AGs-0.2%-PU	$9.57 \times 10^{-5} \pm 2.60 \times 10^{-5}$
T/AGs-0.5%-PU	$3.45 \times 10^{-5} \pm 8.94 \times 10^{-6}$
T/AGs-1.0%-PU	$2.23 \times 10^{-5} \pm 4.27 \times 10^{-6}$

References

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