

Support Information

## A self-healing elastomer with outstanding mechanical properties designed based on urea bonds

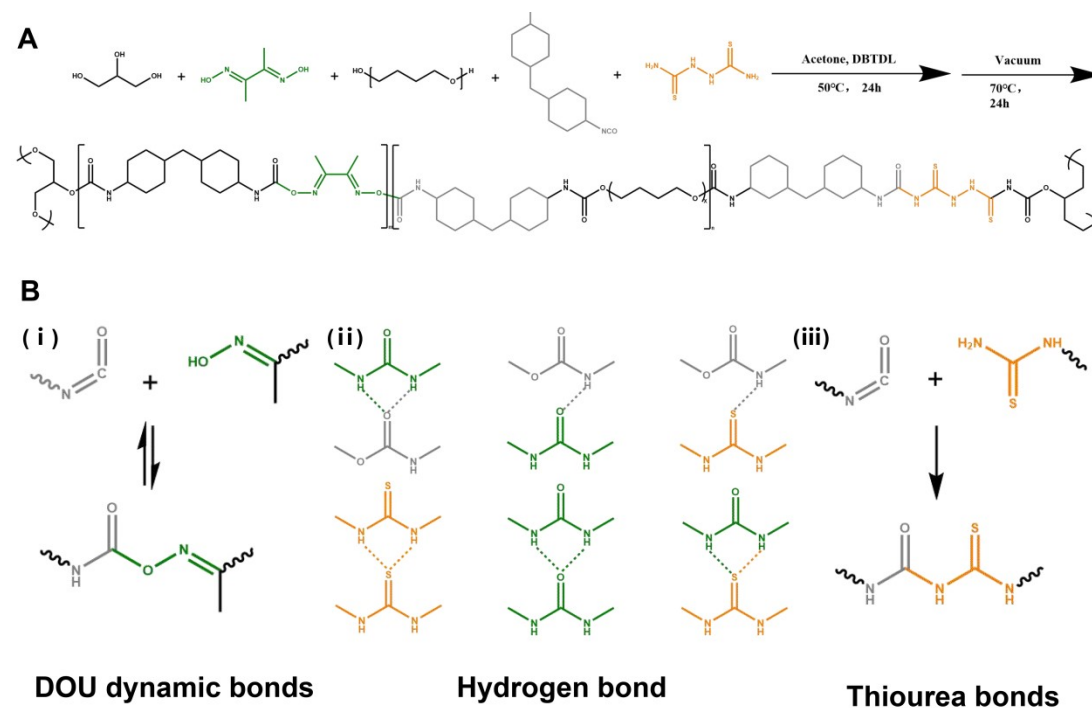
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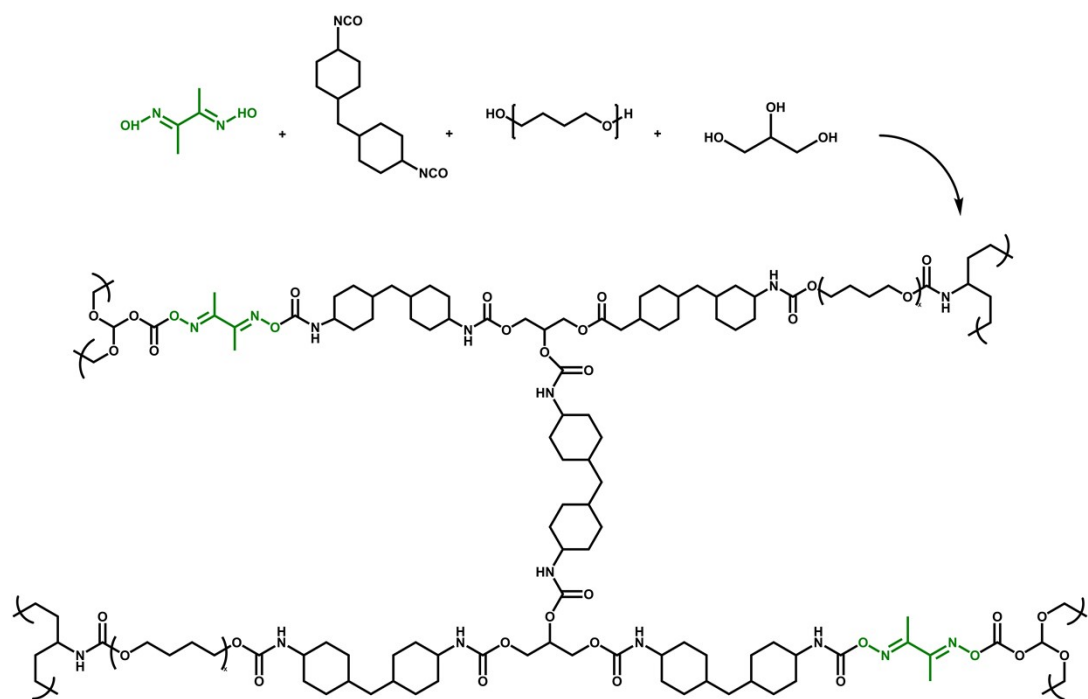
<sup>b</sup> Shaoxing Institute of Technology, Shanghai University, Shaoxing 312000, PR China

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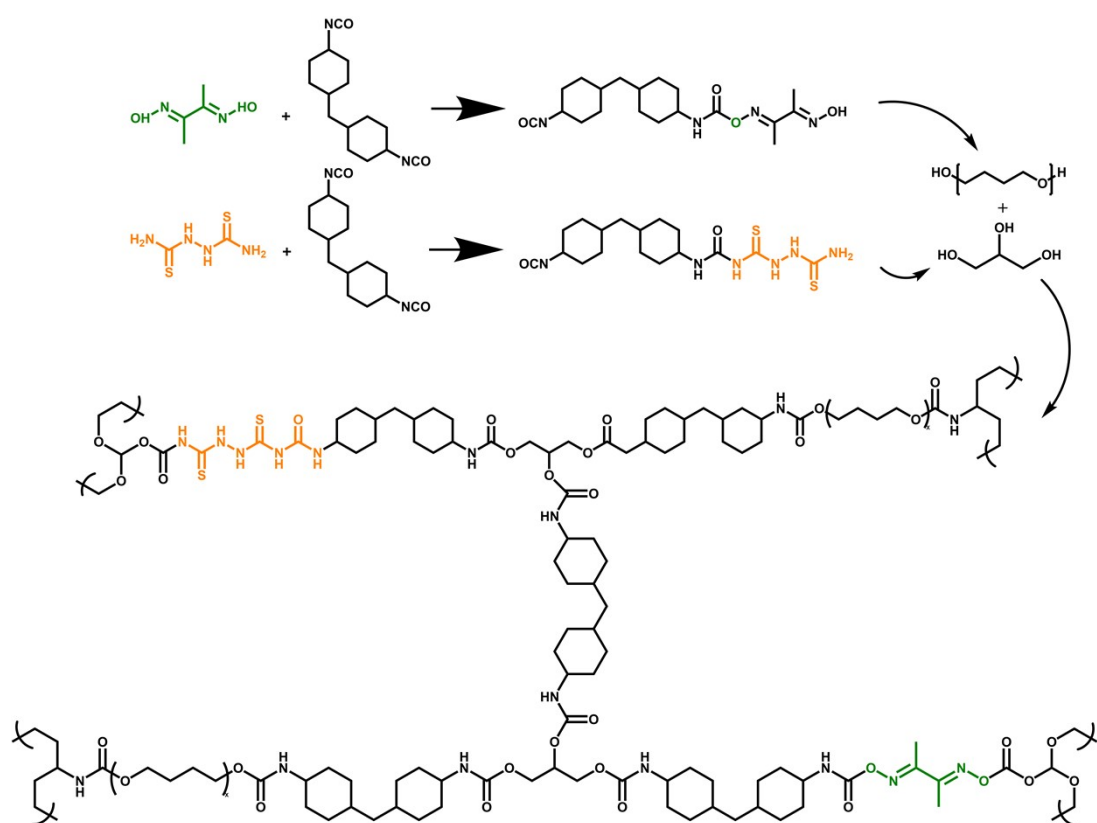
<sup>d</sup> Zhejiang Institute of Advanced Materials, Shanghai University, Jiashan 314113, PR China



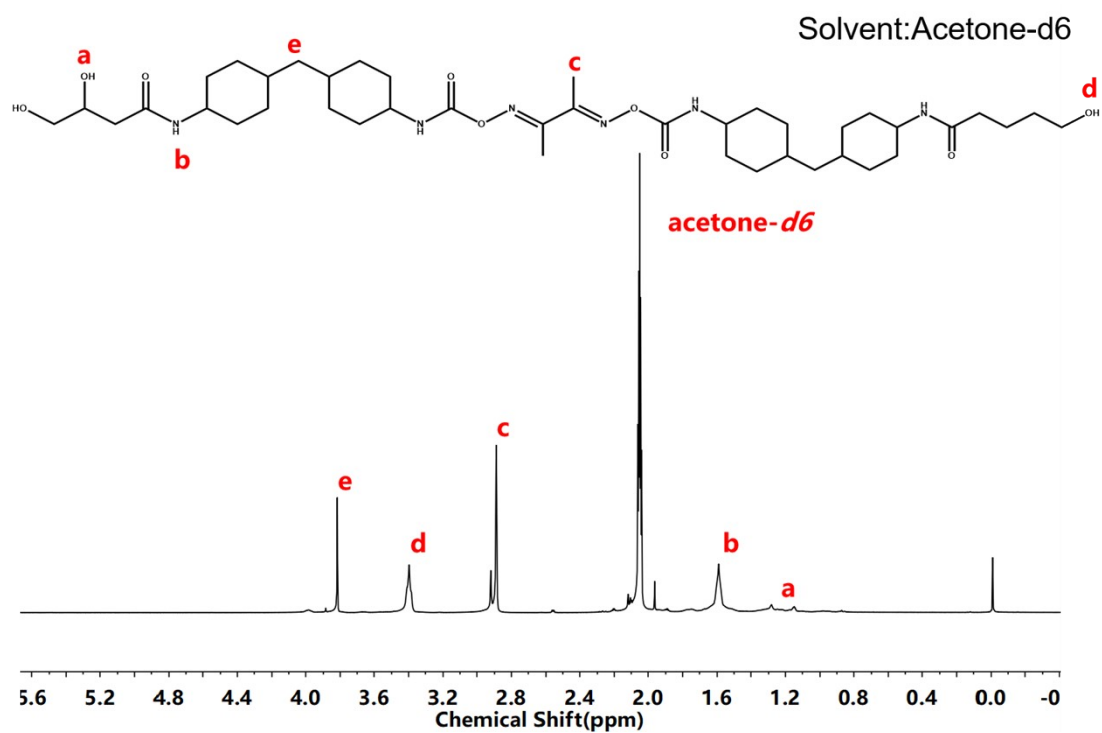
**Figure S1** The route for the synthesis of DDPU.



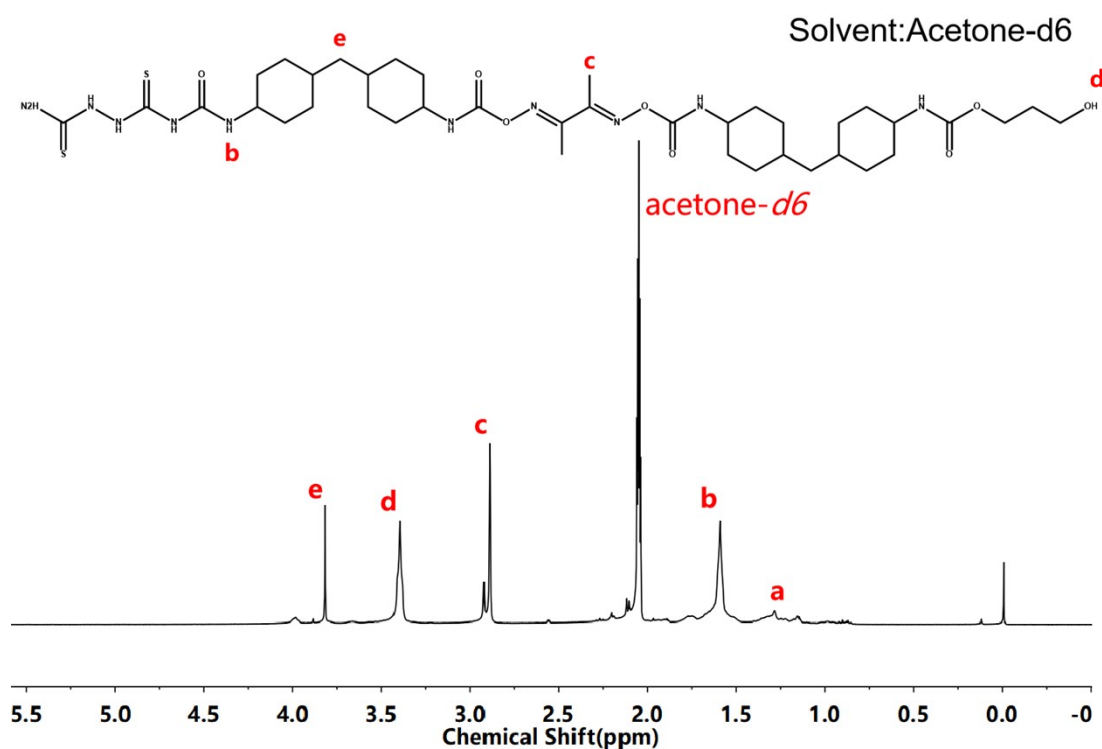
**Figure S2** DOU-PU synthesis roadmap.



**Figure S3** DDPU synthesis roadmap.



**Figure S4**  $^1\text{H-NMR}$  spectrum (400 MHz, Acetone-*d*6) of Br-thiolactone



**Figure S5**  $^1\text{H-NMR}$  spectrum (400 MHz, Acetone-*d*6) of DDPU

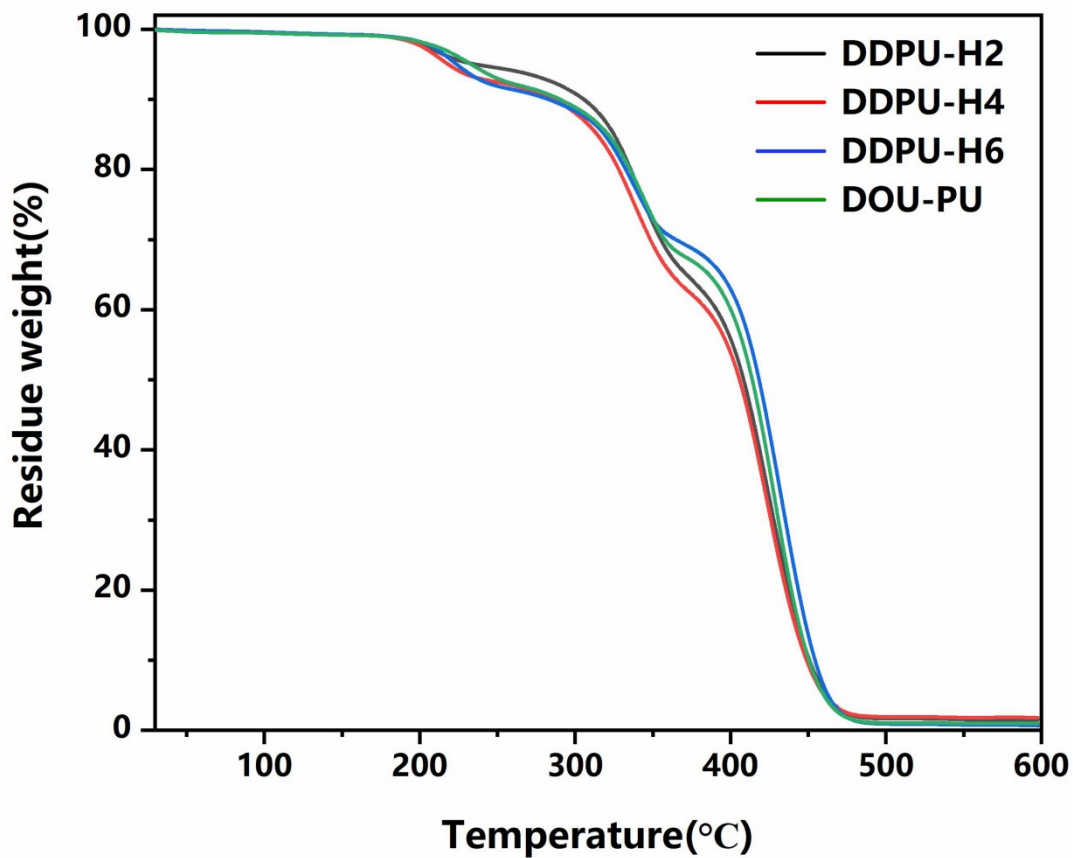


Figure S6 TGA curves of DDPU-H2, DDPU-H4, DDPU-H6 and DOU-PU elastomers

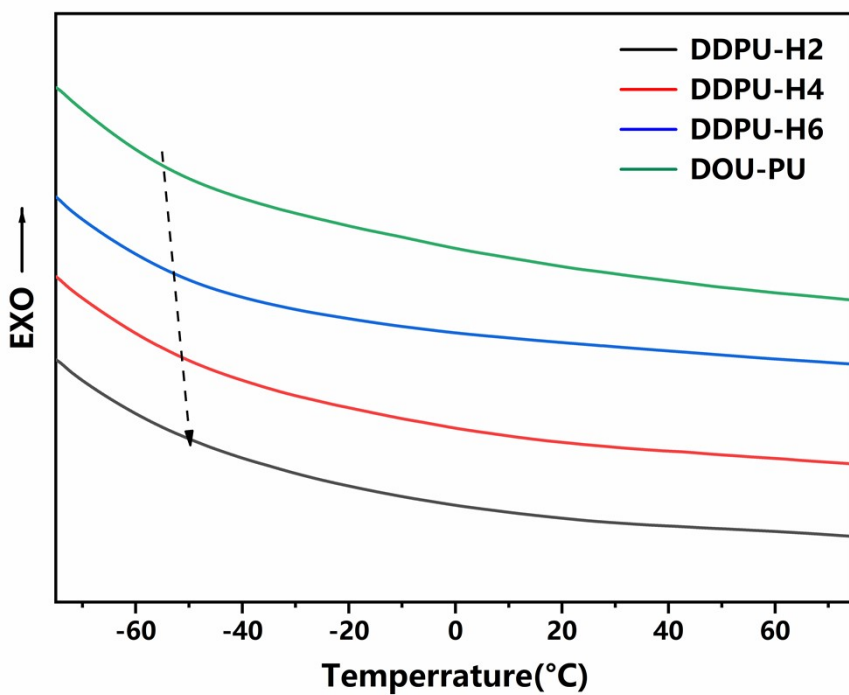


Figure S7 DSC curves of DDPU-H2, DDPU-H4, DDPU-H6 and DOU-PU elastomers

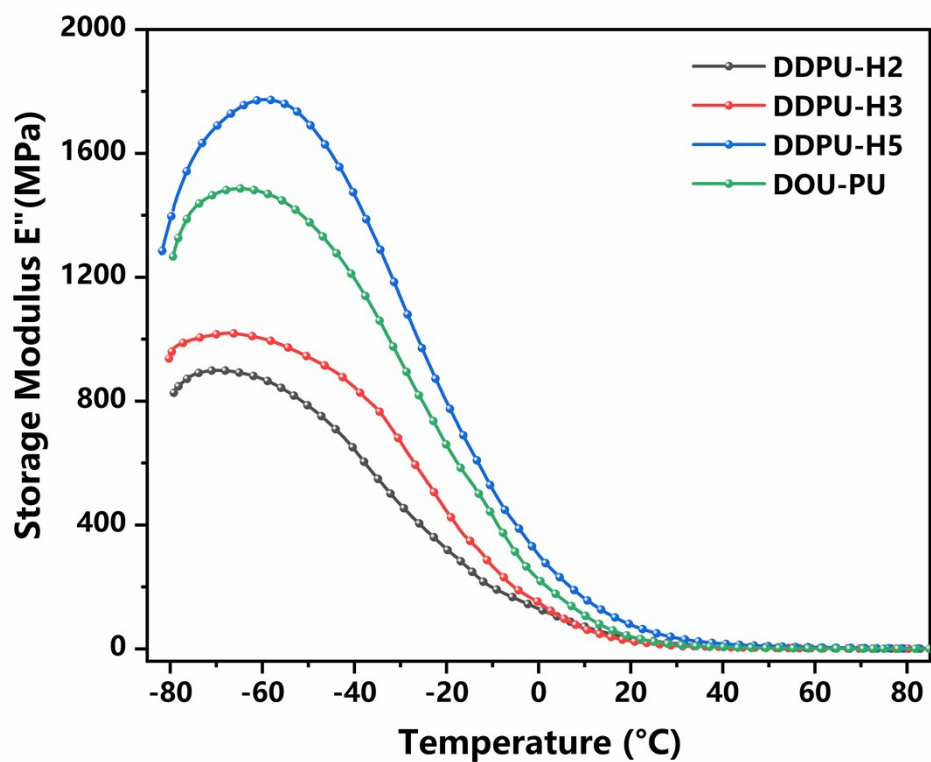


Figure S8 Variation of storage modulus ( $E'$ ) of DDPU and DOU-PU elastomers with temperature.

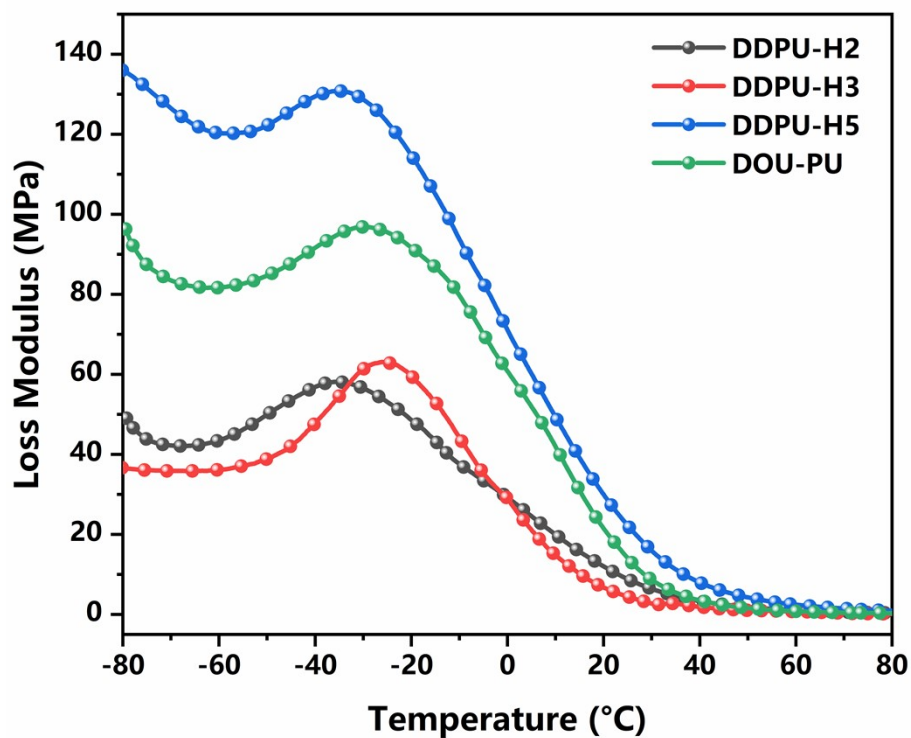
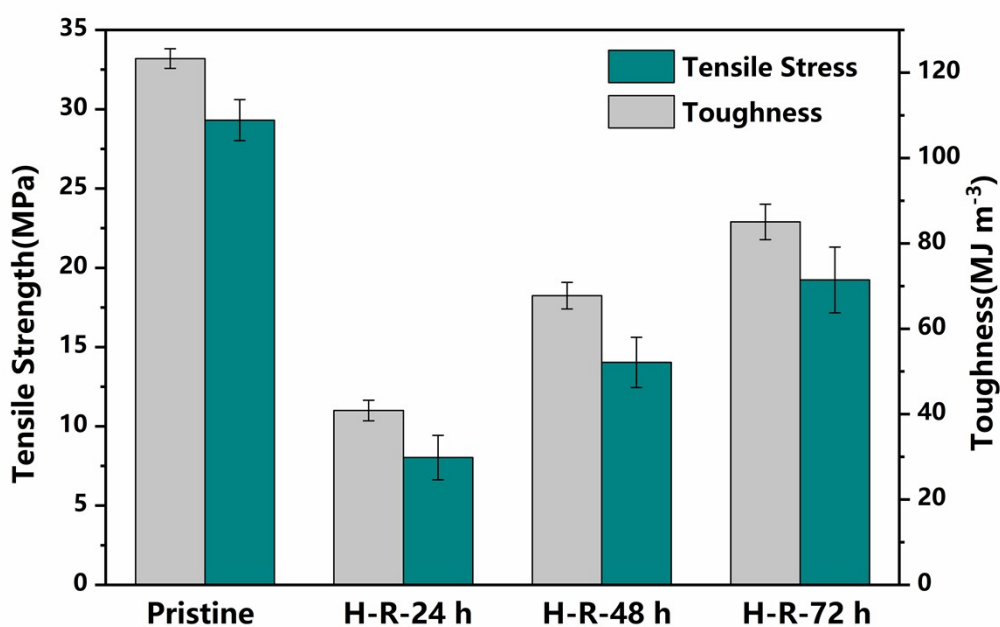


Figure S9 Variation of loss modulus ( $E''$ ) of DDPU and DOU-PU elastomers with temperature.

Table S1 Mechanical properties of DOU-PU, DDPU-H2, DDPU-H3 and DDPU-H4 elastomers

Sample	Breaking strength (MPa)	Toughness (MJ m <sup>-3</sup> )	Breaking elongation (%)
DOU-PU	7.17±0.36	32.52±3.1	1052.44
DDPU-H3	35.99±0.86	93.17±4.1	811.36
DDPU-H4	19.59±0.58	71.9±4.2	636.25
DDPU-H5	23.39±0.76	60.77±3.3	794.28
DDPU-H7	33.20±0.62	108.95±4.8	581.26

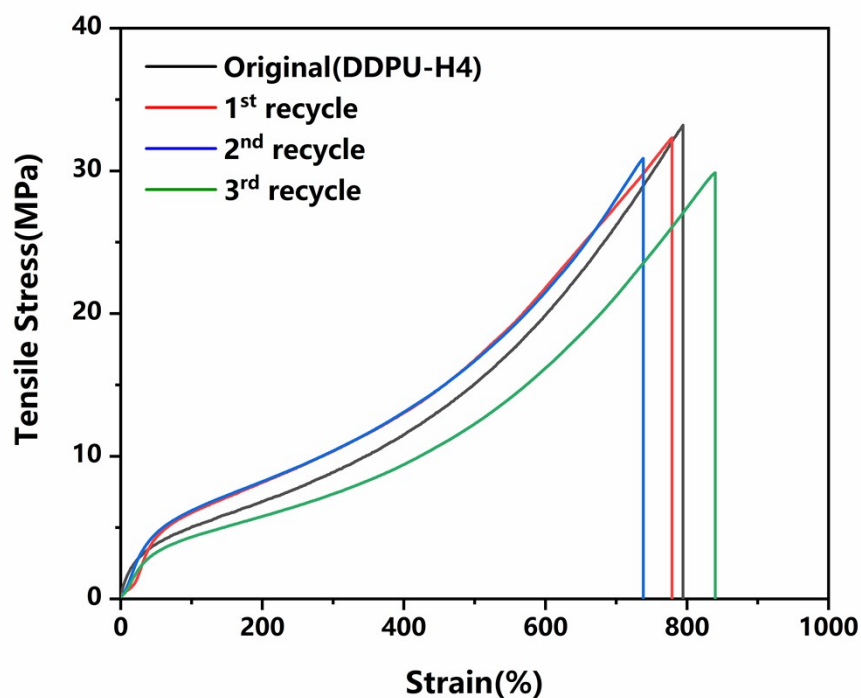


**Figure S10** Tensile strength and toughness of DDPU elastomers healed for different times at room temperature.

**Table S2** Mechanical properties of DDPU elastomer after healing at room temperature for different times.

Sample	Breaking strength (MPa)	Toughness (MJ m <sup>-3</sup> )	Breaking elongation (%)
DDPU-H4	33.20±0.6	108.95±4.8	581.26
H-R-24h	11.07±1.5	29.85±5.2	794.28

H-R-48h	18.25 ± 1.9	52.14 ± 6.6	636.25
H-R-72h	22.92 ± 2.8	71.47 ± 8.7	811.36



**Figure S11** Stress-strain curve of DDPU-H4 elastomer recovered by hot pressing method.

**Table S3** Mechanical properties of DDPU-H4 elastomer restored by hot pressing method.

Sample	Breaking strength (MPa)	Toughness (MJ m <sup>-3</sup> )	Breaking elongation (%)
Original	33.20	108.95	794.27
1 <sup>st</sup> recycle	32.33	113.89	778.90
2 <sup>nd</sup> recycle	30.88	101.88	738.20
3 <sup>rd</sup> recycle	29.87	102.06	839.85

Tensile mechanical properties were carried out on the AGS-X (Shimadzu) tensile machine (test speed: 100 mm min<sup>-1</sup>, gauge length: 15 mm). The toughness is calculated using eq 1.<sup>1,2</sup>

$$Toughness = \int_0^{fracture} \sigma d\varepsilon \quad (1)$$

Here,  $\varepsilon$  represents the strain during the tensile test and  $\sigma$  represents the tensile stress.

The definition of self-healing efficiency ( $\eta$ ) is based on the integral area under the stress-strain curve, which is calculated using the formula below.

$$\eta = \frac{\sigma_{healed}}{\sigma_{original}} \quad (2)$$

In the formula,  $\eta$ —self-healing efficiency (%);

$\sigma_{healed}$ —The toughness of the pattern after healing;

$\sigma_{original}$ —The toughness of original style.

**Table S4** Reagent usage of DOU-PU and DDPU elastomers

	PTMEG	HMDI	Glycerol	DMG	DBTU	DBTDL
DOU-PU	1	0.622	0.023	0.116	0.000	0.02g
DDPU-H2	1	0.622	0.023	0.058	0.044	0.02g
DDPU-H3	1	0.622	0.023	0.070	0.035	0.02g
DDPU-H4	1	0.622	0.023	0.081	0.026	0.02g
DDPU-H5	1	0.622	0.023	0.093	0.018	0.02g
DDPU-H6	1	0.622	0.023	0.104	0.009	0.02g