Supporting Information

Mechanically enhanced healable and recyclable silicone with dynamic hindered urea bond for flexible electronics

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Fig. S1. The possible chemical reaction during PDMS-SPUA preparation.

Chemical structure of compound with hindered urea bond

Synthesis of hindered urea: 0.168 g HDI was added slowly into the solution of PA/ acetone (0.086 g/2 g) in a beaker. The reaction was carried out 1 h at 50 °C until the white solid appeared. And then the solid was dried at 50 °C for 1 h under vacuum.



Fig. S2. FTIR spectra of compound with hindered urea bond and PDMS-SPUA-31.



Fig. S3. (a)Tan δ curves and (b) storage modulus curves of PDMS-SPUA-xx.



Fig. S4. Swelling test of samples, take PDMS-SPUA-21 as an example.

Calculation of crosslink density

The rubber molecular chains are connected into a denser three-dimensional network structure as the cross-linking density increases according to the cross-linking network theory, which limits the movement of the molecular chains. Therefore, the smaller the deformation, the greater the modulus. According to the theory of rubber elasticity, the cross-linking density v_e can be calculated using Eq. (S1).

$$v_{\rm e} = \frac{E}{3RT}$$
(S1)

Whereby v_e represents cross-linking density (in mol·m⁻³), *E* is storage modulus of rubber platform area above T_g (in 0.1Pa), *R* is the gas constant ($R = 8.314 \text{ J} \cdot \text{mol}^{-1}\text{K}^{-1}$), and *T* is absolute temperature (in K). Table S1 gives the results of calculation.

Sample	Crosslinking density (mol·m ⁻³)	$\sigma({ m MPa})^{ m a}$	ε (%) ^b	E (MPa) ^c
PDMS-SPUA-01	151.00	1.9 ± 0.1	79.4 ± 3.6	69.9 ± 3.4
PDMS-SPUA-11	93.20	1.9 ± 0.5	155.8 ± 24.8	49.5 ± 10.7
PDMS-SPUA-21	94.74	2.0 ± 0.4	232.2 ± 7.9	51.9 ± 15.9
PDMS-SPUA-31	95.78	2.9 ± 0.1	297.7 ± 1.1	53.2 ± 0.3
PDMS-SPUA-12	102.58	2.9 ± 0.1	247.8 ± 14.5	55.3 ± 1.2
PDMS-SPUA-13	108.18	2.6 ± 0.5	225.2 ± 21.2	58.1 ± 2.8

Table S1. The crosslinking density and the mechanical properties of different samples

^aMaximum tensile strength. ^bElongation at break. ^cElastic modulus.



Fig. S5. TGA curves and DTG curves of PDMS-SPUA-xx.



Fig. S6. Stress-strain curves of different samples before and after cutting healing.



Fig. S7. Stress-strain curves of different samples before and after recycling.



Fig. S8. FTIR spectra of the original and recycled PDMS-SPUA-xx samples.



Fig. S9. Cut-healing efficiency of PDMS-SPUA/graphene under different healing temperature.