Supporting information

Cross-Linked Polyurethane with Dynamic Phenol–Carbamate Bonds: Properties Affected by the Chemical Structure of Isocyanate

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Fig. S2 FTIR spectroscopy of (a)PU-H, (b)PU-IP, (c)PU-X, (d)PU-HX and (e)PU-



Fig. S3 Dynamic properties of model compounds. (a) Reaction of model compounds. (b) After different periods of time, the characteristic peaks of (b1) AF-CHI (b2) AF-BI (b3) AF-HI in ¹H NMR spectra. The calculation method of reactant consumption ratio is: $(A_b+A_*)/(A_a+A_b+A_*)$, where A is the integrated area of the characteristic peaks.



Fig. S4 Swelling of the PU samples.

Sample	Original mass (g)	Mass after swelling test (g)	Gel fraction (%)
PU-H	1.361	1.342	98.6
PU-IP	1.528	1.458	95.4
PU-X	1.442	1.433	99.4
PU-HX	1.353	1.317	97.3

Table S1 Swelling and Soxhlet extraction results of the cross-linked PU same	oles
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Fig. S5 Stress-strain curve of the five PU samples in the 350% strain cyclic tensile

test with 350% maximum strain.



Fig. S6 The relationship between phenol-carbamate dissociation ratio and time of five PU samples at 120°C.

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Sample	$\Delta H_r^0 (kJ \cdot mol^{-1})$	$\Delta S_r^0 (J \cdot mol^{-1} \cdot K^{-1})$
PU-H	207	476
PU-IP	150	343
PU-X	117	248
PU-HX	166	374

Table S2 Thermodynamic parameters of compound reaction in polymer bulk



Scheme S1 Synthesis of linear polyurethane samples.



Fig. S7 Stress relaxation curves at 50°C of linear polyurethane samples under a 5% strain.



Fig. S8 Stress relaxation curves of PU-IP at 90°C under different strains.



Fig. S9 Maxwell plots of stress relaxation curve of the PU samples at different

temperatures.



Fig. S10 Optical images of repairing behavior of scratched PUs healed at 100 °C.



Fig. S11 TGA curves of the five PU samples. Experiments were conducted using a nitrogen atmosphere with a heating rate of 20 °C/min



Fig. S12 Stress-strain curve of five PU samples before and after thermal aging in air



Fig. S13 Time sweep of five PU samples in air at 120 °C for 8h.



Fig. S14 Stress-strain curve of control PU samples (without dynamic covalent bonds) before and after heat treatment.