SUPPORTING INFORMATION

Synthesis of Self-Healing Waterborne Polyurethane Systems

Chain Extended with Chitosan.

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Figure 1. FT-IR spectra of waterborne polyurethanes (WPU) during the chain extension. Peaks due to isocyanate groups at 2259 cm⁻¹ disappeared after 3 h.



Figure S2. Images of WPU emulsions.



Figure S3. Particle size distribution of WPU emulsion.

Table S1. Number average pa	article size of WPU emulsions.
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Sample code	Number average diameter (nm)
WPU-C0	40.8
WPU-C1	37.1
WPU-C2	48.3
WPU-C3	182.5

Table S2. Gel contents of WPU films.

Sample code	Gel content* (%)
WPU-C0	0.35 (±0.03)
WPU-C1	48.85 (±0.71)
WPU-C2	61.58 (±0.24)
WPU-C3	66.53 (±4.37)

* Determined from the ratio of sample weight after removal of soluble parts and the weight of pristine WPU



Figure S4. ¹H NMR spectra of the model compounds for the exchange reaction between Urea-1 and Urethane: (**a**) butanol; (**b**) Urethane; (**c**) products of the exchange reactions between Urea-1 and Urethane.



Figure S5. ¹H NMR spectra of the model compounds for the exchange reaction between Urea-1 and Urea-2: (**a**) octyl amine; (**b**) Urea-2; (**c**) products of the exchange reactions between Urea-1 and Urea-2.



Figure S6. The FT-IR spectra of polyurethanes (PU) films before and after heat treatment for 24 h at 110 °C.



Figure S7. FT-IR spectra of WPU films before and after heat treatment for 24 h at 110 °C in the wavenumber of 2000~1000 cm⁻¹.



Figure S8. SAXS data of WPU films before and after heat treatment for 24 h at 110 °C.



Figure S9. SAXS data of WPU films before and after heat treatment for 24 h at 110 °C.

Sample code	d-spacing (nm)	
	Before heat treatment	After heat treatment
WPU-C0	28	28
WPU-C1	29	29
WPU-C2	35	33
WPU-C3	38	35

Table S3. d-spacing of WPU films based on SAXS data before and after heat treatment for 24 h at 110 °C.





WPU-C1



WPU-C2



WPU-C3



Figure S10. SEM images of the WPU films after the second self-healing test by the additional scratch and heat treatments at 110 °C for 24 h.