Supporting information for

## Mechanically Strong and Stretchable Polyurethane-Urea Supramolecular Hydrogel Using Water as Additional In-situ Chain Extender

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Fig. S1 Normalized FTIR spectra of -NCO group at different reaction time.

Completion of the prepolymer formation was determined by the amount of the isocyanate group using the di-n-butylamine-acetone method associated with normalized FTIR spectroscopy following the intensity of the strong -NCO groups centered around 2270 cm<sup>-1</sup>.<sup>1</sup>



**Fig. S2** Compression measurements of the hydrogel with [NCO]/[OH] = 22/14. The sample was compressed to strain = 90%. The diameter of the sample was 24 mm with thickness of 21.5 mm. Here the compression force at a strain of 90% is about 16.5 kN, which is very close to the up limit of the machine (20 kN). To protect the machine, we did not get the strain and stress or force at break.



Fig. S3 Loss modulus of the PUUS hydrogels with various [NCO]/[OH] ratios.



Fig. S4 Photo image of the resultant viscous acetone solution.



Fig. S5 TG curve of the dry PUU material with [NCO]/[OH] = 22/14.



**Fig. S6** The temperature dependence of the dynamic storage modulus (G') and loss modulus (G") for the different PUU dry films with [NCO]/[OH] ratios of 24/14 and 26/14.

1. Xiong J., S., F., Du, H. Chinese Journal of Analysis Laboratory, 2007, 26, 73.