## Supporting Information

## Double-crosslinked network design for self-healing, highly stretchable and resilient polymer hydrogels

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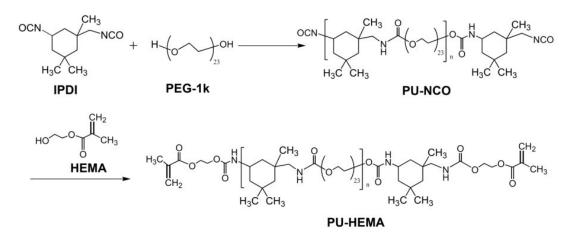
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Scheme S1 The route to preparing PU-HEMA.

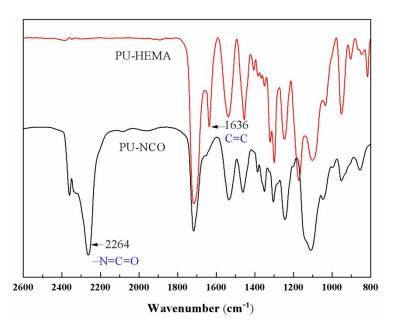


Fig. S1 FTIR spectra of PU-NCO and PU-HEMA.

The FTIR spectra of PU-NCO and PU-HEMA samples are shown in Fig. S1. From the FTIR spectrum of PU-NCO, the characteristic peak at 2264 cm<sup>-1</sup> assigned to the -N=C=O asymmetric stretching vibration can be clearly observed, while in the FTIR spectrum of PU-HEMA, this peak disappears and a new peak appears at 1636 cm<sup>-1</sup>, which is attributed to the stretching vibration of C=C in HEMA unit. It means that no residual PU-NCO exists in the synthesized PU-HEMA, thus confirming the formation of PU-HEMA and proving the completeness of the reaction between PU-NCO and HEMA.



Fig. S2 Photograph of the hydrogel samples.

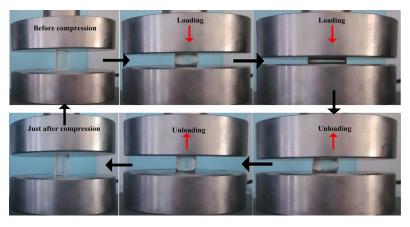
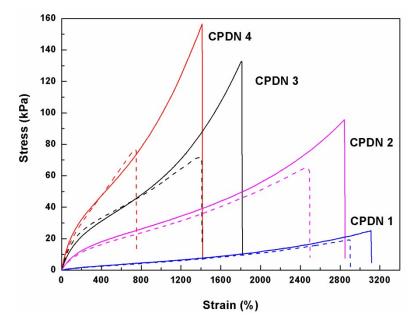


Fig. S3 Photos of the CPDN hydrogel sample during cyclic uniaxial compression tests.



**Fig. S4** Tesile stress–strain curves of the original CPDN hydrogel samples (solid line) and the corresponding self-healed CPDN hydrogel samples at 30-min healing time (dotted line).