

## Electronic Supporting Information

### **Dynamic nanoconfinement strategy towards self-healing soft electronics with superstretchability, ultrahigh strength and reliably high conductivity**

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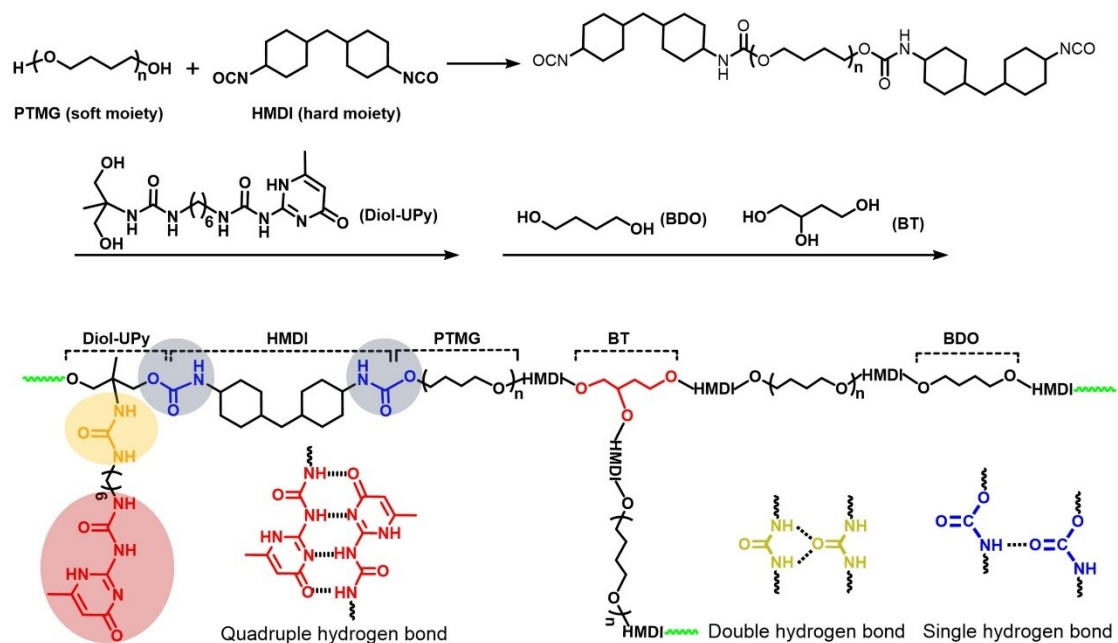
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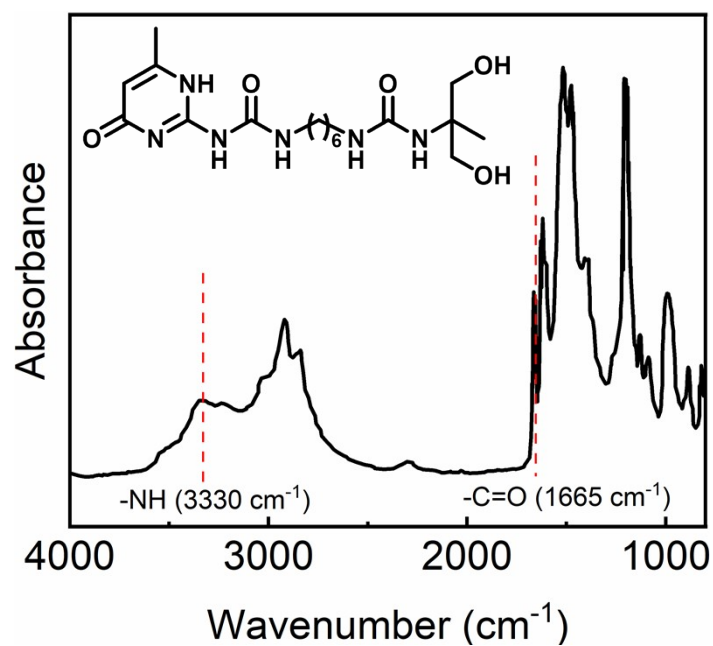
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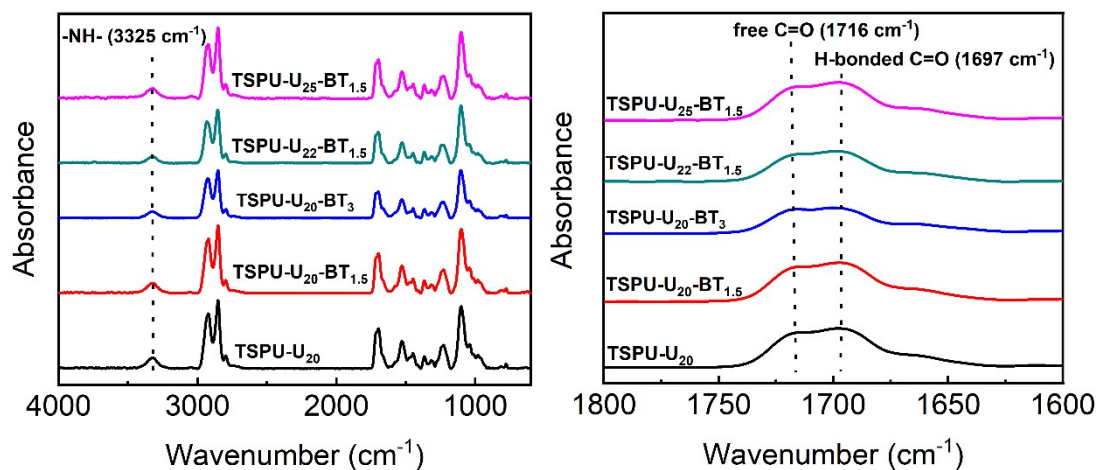
Keywords: bioinspired nanoconfinement, self-healing electronics, superstretchability, ultrahigh strength, reliably high conductivity



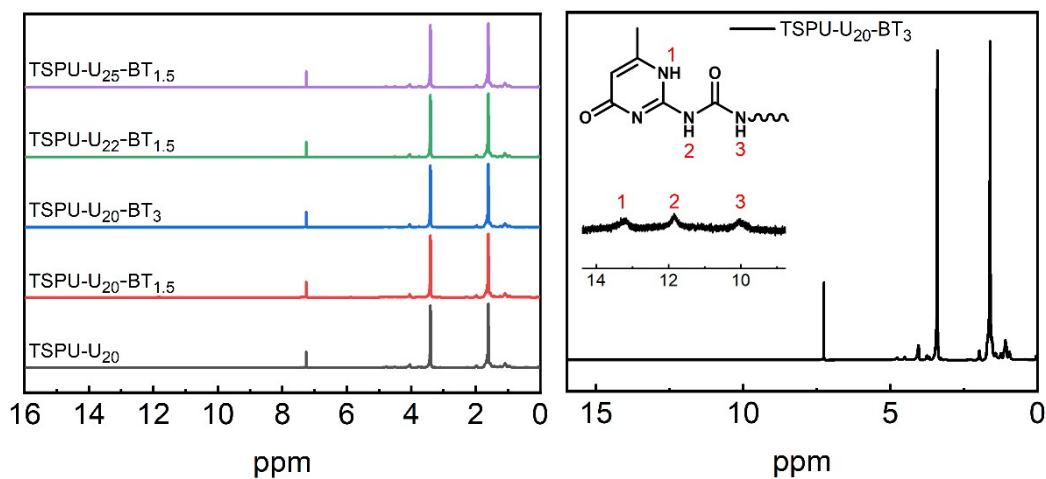
**Figure S1.** The synthesis route of TSPU polymer. In the final self-healing polymer matrix, there existed hierarchical (single, double, quadruple) hydrogen bonding interactions, which induced the aggregation of H-bonds from hard moieties forming nanoconfined phase.



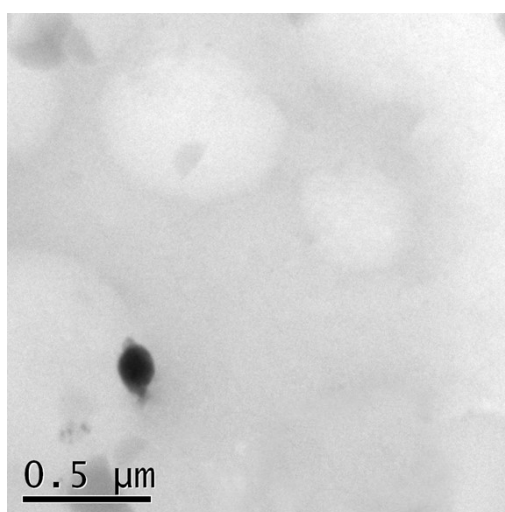
**Figure S2.** The FTIR spectrum of Diol-UPy.



**Figure S3.** FTIR spectra of TSPU elastomers. The absorption peaks at  $3325\text{ cm}^{-1}$ ,  $1716\text{ cm}^{-1}$  and  $1697\text{ cm}^{-1}$  were attributed to the absorbance of  $\text{-NH-}$ , free  $\text{C=O}$  and H-bonded  $\text{C=O}$ , respectively.



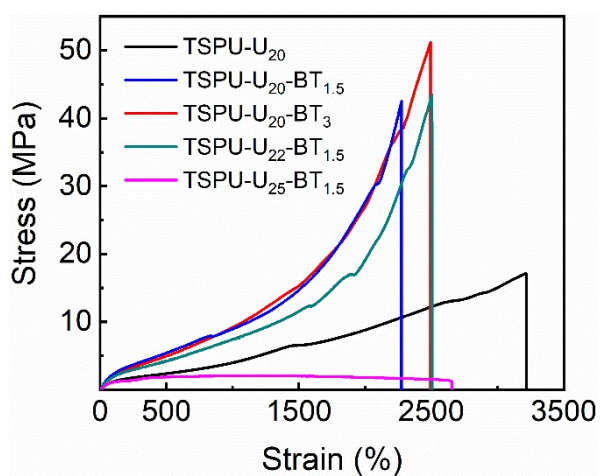
**Figure S4.**  $^1\text{H}$ NMR spectra of TSPU elastomers. The chemical shifts at  $13.21\text{ ppm}$ ,  $11.85\text{ ppm}$ , and  $10.06\text{ ppm}$  demonstrated the existence of UPy groups.



**Figure S5.** Characterization of TSPU- $\text{U}_{25}\text{-BT}_{1.5}$  polymer with TEM.



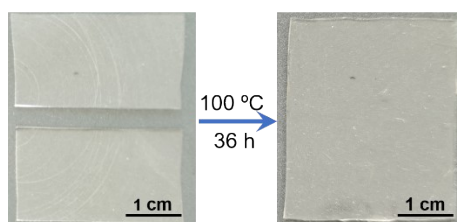
**Figure S6.** Optical photographs of TSPU elastomer at 0% (left) and 2000% (right) strains.



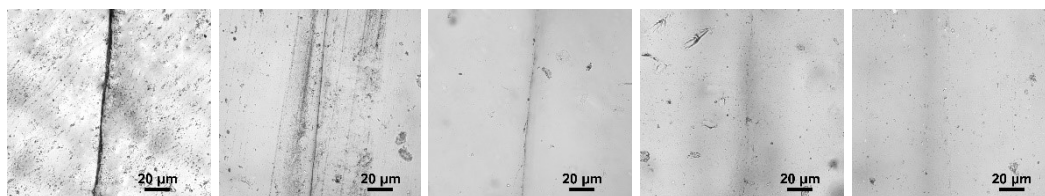
**Figure S7.** The engineering stress-strain curves of as-synthesized TSPU elastomers.



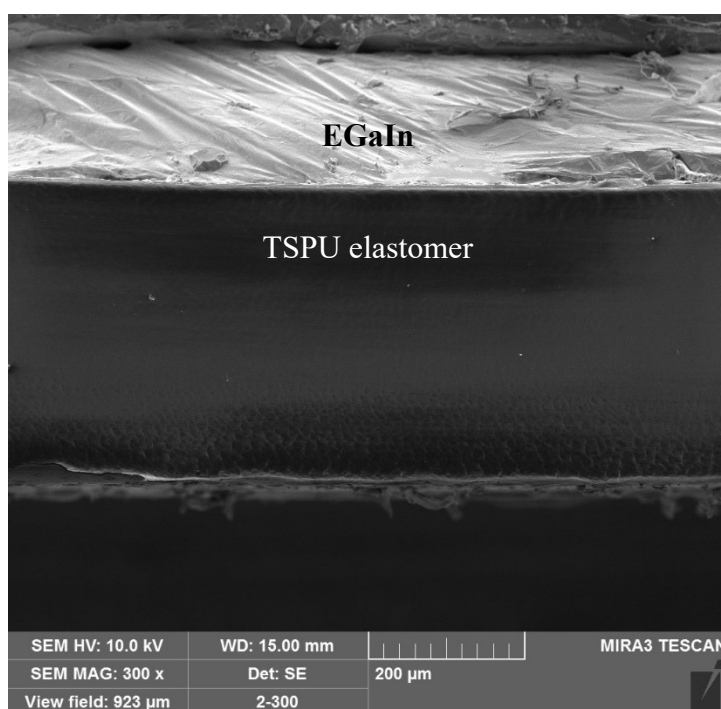
**Figure S8.** The weights of dumbbell and TSPU sample used to lift up dumbbell.



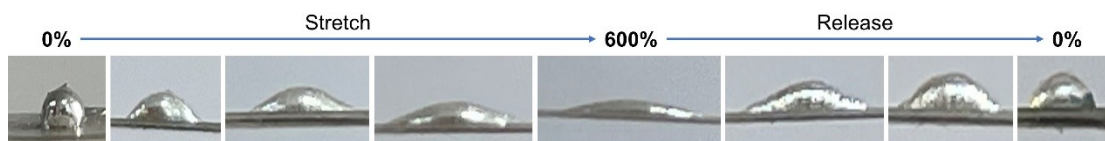
**Figure S9.** The cut-off sample was self-healed after handled at 100 °C for 36 h.



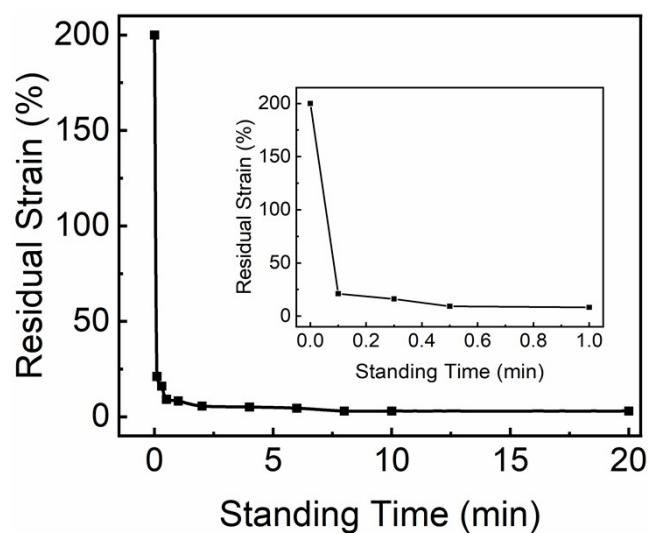
**Figure S10.** The microscopy images of self-healing process for reattached two cut-off pieces at 100 °C for 0 h, 4 h, 12 h, 24 h, 36 h (from left to right).



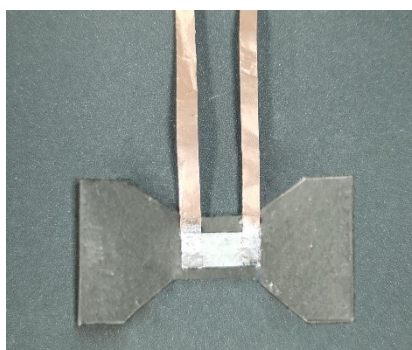
**Figure S11.** Cross-profile image of electrode characterized by SEM. It is shown that EGaIn liquid metal spreads on TSPU elastomer and the thickness of EGaIn is ca. 170 μm.



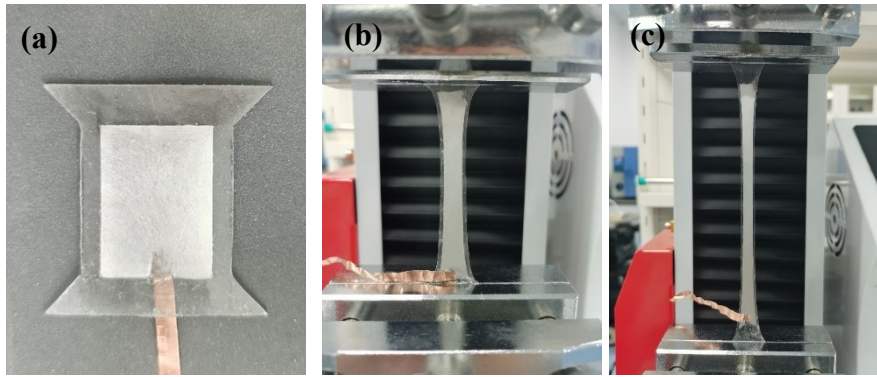
**Figure S12.** (a) Photographs of EGaIn droplet illustrating a well surface reconciliation on a TSPU-U<sub>20</sub>-BT<sub>3</sub> specimen during stretching and releasing process.



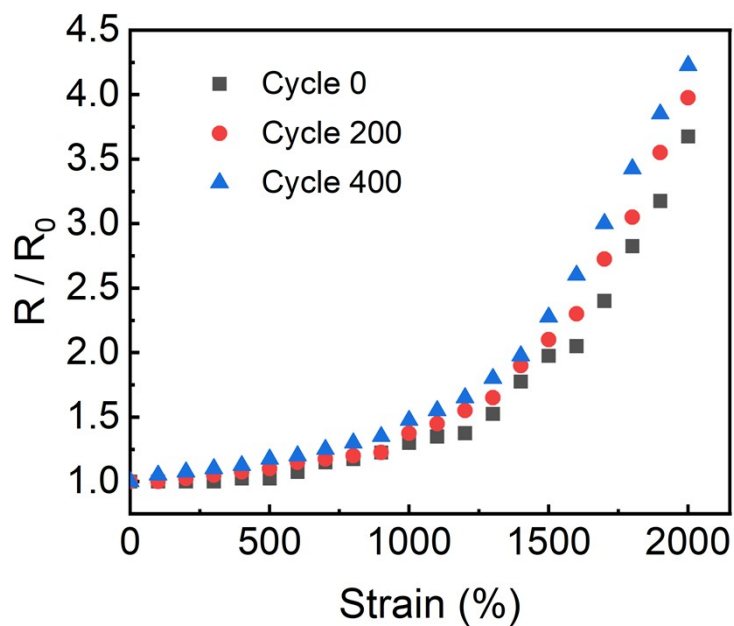
**Figure S13.** The dependence of residual strain on standing time of relaxed TSPU after being stretched to 300% strain.



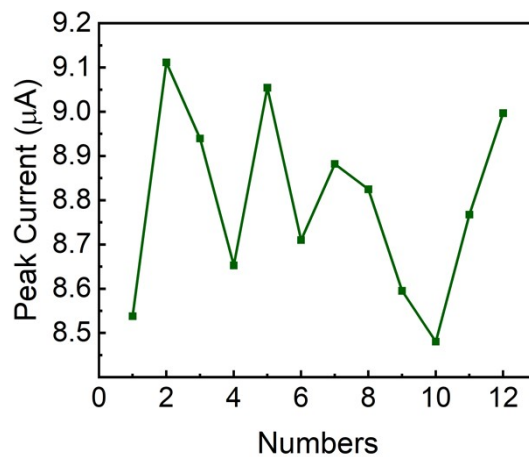
**Figure S14.** Soft electrode used for measuring its conductivity.



**Figure S15.** The photography images of supertough self-healing triboelectric nanogenerator (TS-TENG, working area: 3 cm × 2 cm) (a) and its tensile states at 500% (b) and 900% (c) strains.



**Figure S16.** The dependence of  $R / R_0$  on strain of healed electrode after varied stretch-release cycles. One stretch-release cycle is referred to the electrode stretched to 100% and then released.



**Figure S17.** The variation of peak short circuit current of TS-TENG versus testing numbers. The standard

Entry	Electrodes	Strain (%)	Stress (MPa)	R/R <sub>0</sub>	Healing efficiency (%)	Ref.
A	DMSO/PEDOT:PSS	32	11.5	9.5@40%	No	1
B	P(AAm-co-HEMA)/PANI	90	17.5	1.8@70%	No	2
C	F-PNIPAAm/PANI	187	0.042	5.5@120%	No	3
D	Ionic conductive cellulose hydrogel (CCH)	220	2	2.62@220%	No	4
E	P(AN/AANa)	425	23.17	3.5@425%	No	5
F	PVA <sub>2</sub> PEI <sub>1-75</sub>	500	0.6	14.5@460%	No	6
G	PADL <sub>3</sub> hydrogel	500	0.052	31@500%	93	7
H	PEDOT:PSS-PAAm organogels	525	0.03	17@300%	No	8
I	PVA-1% CNF	625	1.4	6.76@400%	No	9
J	PAA-rGO hydrogel	630	0.4	5.25@500%	75	10
K	SACP film	700	1.2	81@400%	No	11
L	PVA/GE/GL/NaCl gels	715	1.04	6@600%	No	12
M	HPC/PVA <sub>16%</sub> hydrogel	800	1.3	9@400%	No	13
N	PAA/PANI/glycerol/Fe <sup>3+</sup>	991	0.0357	31@275%	90.8	14
O	GN-CNF@PVA hydrogel	1000	0.0037	21@500%	98.9	15
P	MXene/PVA/PAAm	1000	0.035	76@350%	85	16
Q	PAAm-oxCNTs	1044	0.71	21@700%	No	17
R	Cel <sub>14</sub> /PAA-Fe <sub>2.50</sub> <sup>3+</sup> hydrogel	1755	0.1	64@1100%	No	18
S	VP/PP/ZP/Al <sup>3+</sup> hydrogels	1769	0.46	23.5@700%	91.8	19
T	PDMS-MPU <sub>0.4</sub> -IU <sub>0.6</sub>	1842	1.5	6@500%	78	20
U	HPAAN/PDA	1845	0.438	1.3@200%	49	21
V	AFPs <sub>50</sub> .PAM/PVA <sub>0.5</sub>	2358	0.058	97@1000%	No	22
W	MGP	2700	0.3	41@2700%	No	23
X	LPBP hydrogel	2850	0.2	18@500%	98.6	24
Y	DAE-3	5100	1.9	301@100%	86	25
Z	This work	~2500	~50	3.63@2000%	82.9	

deviation is 0.21  $\mu$ A.

**Table S1** Comparison of comprehensive performance of electrode herein to that of previously reported stretchable electrodes.



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