Supporting information

Highly Transparent, Stretchable, and Conductive Supramolecular Ionogels Integrated with Three-Dimensional Printable, Adhesive, Healable, and Recyclable Character

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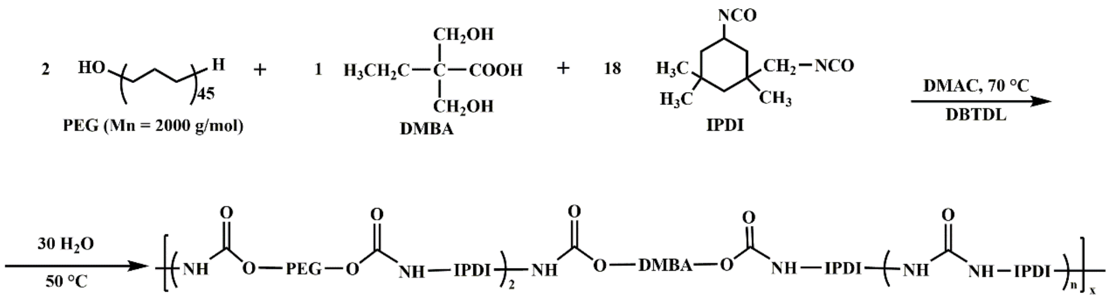

Figure S1. Synthetic procedure of the multiurea linkage segmented PUU copolymer.

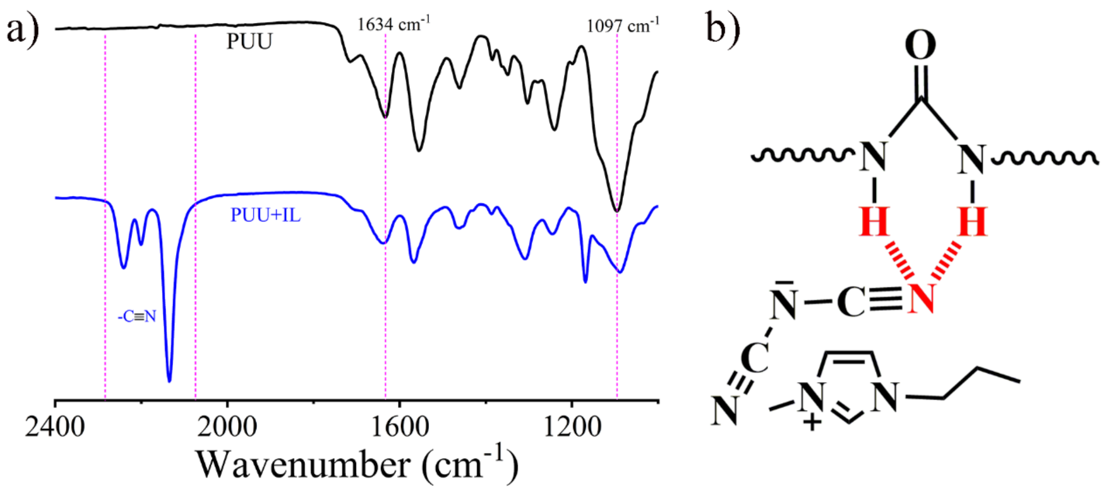

Figure S2. (a) FTIR spectra of the PUU and 60 wt % IL-containing supramolecular ionogel film, and (b) schematic illustration of H-bonding between the urea and cyanide groups.

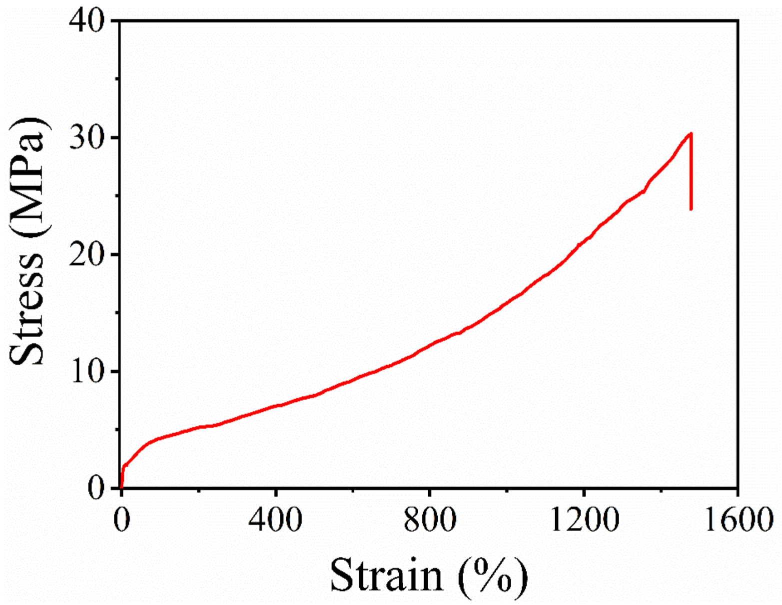

Figure S3. Tensile testing stress-strain curve of the PUU dry film.

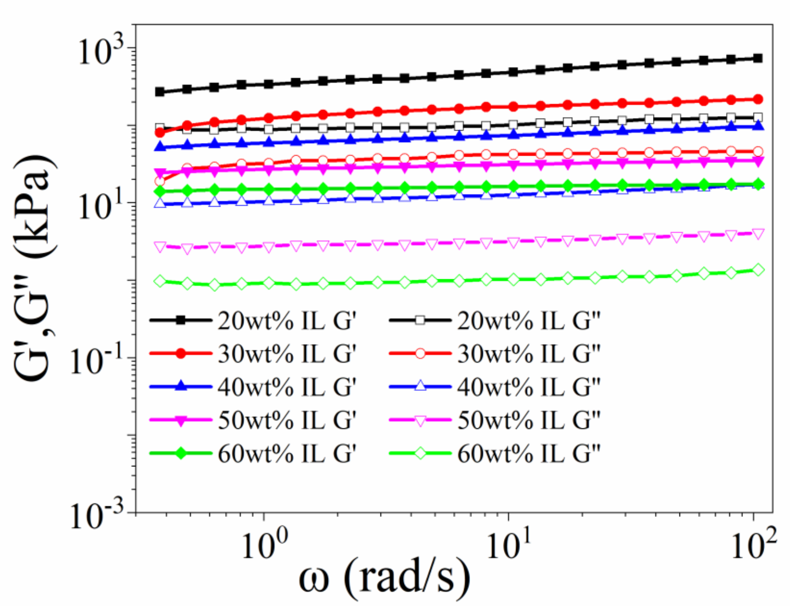

Figure S4. Storage modulus (G′) and loss modulus (G″) of the ionogels with various IL contents.

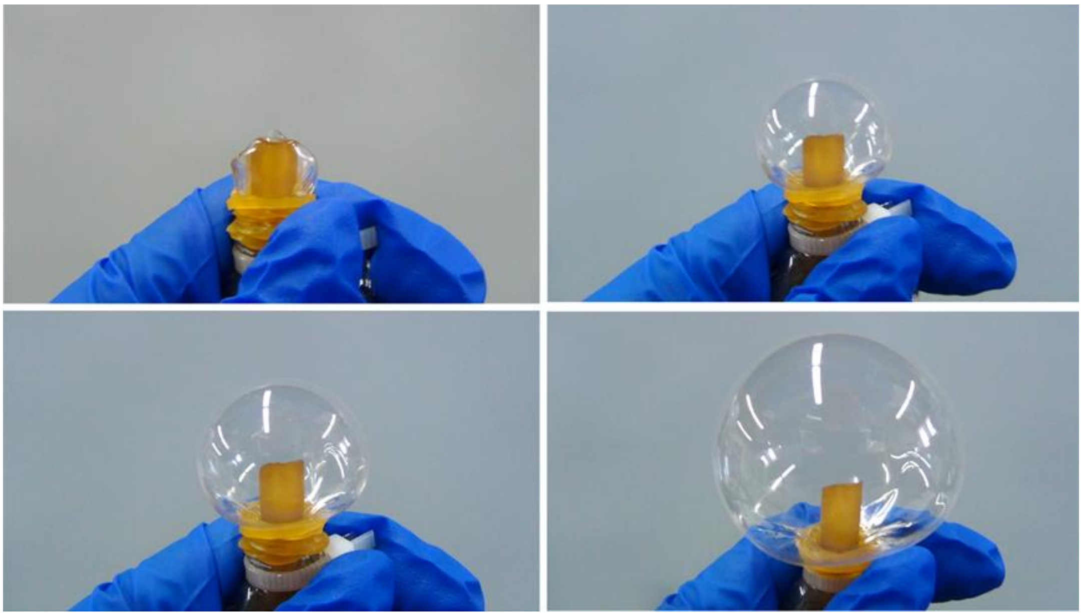

Figure S5. Digital images showing the ultra-tough character of the 60 wt % IL-containing supramolecular ionogel.

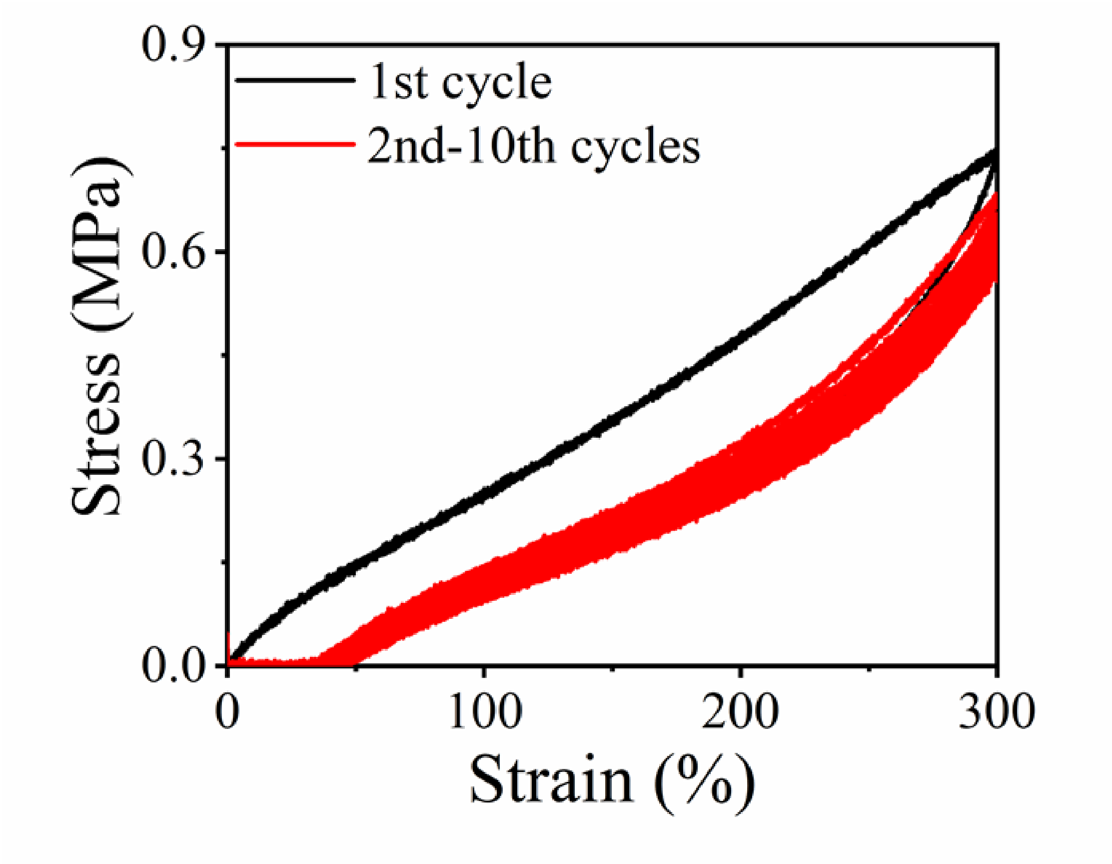

Figure S6. Cyclic tensile tests for the 60 wt % IL-containing supramolecular ionogel with a maximum strain of 300%.

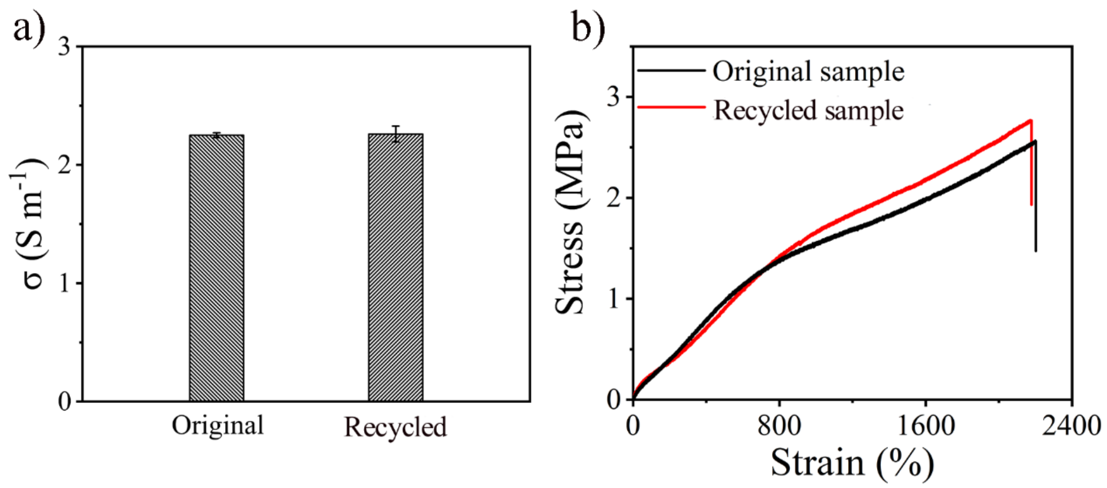

Figure S7. (a) The ionic conductivity of the original and recycled 60 wt % IL-containing supramolecular ionogel films. (b) Tensile testing curves of the original and recycled 60 wt % IL-containing supramolecular ionogel films. Recycled film was prepared as follows: original ionogel film was firstly cut into small pieces and then dissolved in methanol. The methanol solution was poured into Teflon mold, where the solvent evaporated to form solid film, which was further dried under high vacuum at 40 °C to yield dry film.

Table S1. F_{adh} of ionogels with different IL content to various substrates

Substrates / The content of IL	Brown paper	Plastic	Wood	Stainless steel
20 wt %	1956 kPa	161.4 kPa	3800 kPa	888 kPa
30 wt %	1548 kPa	107 kPa	2366 kPa	804 kPa
40 wt %	1231 kPa	64 kPa	1647 kPa	727 kPa
50 wt %	830 kPa	52 kPa	1314 kPa	568 kPa
60 wt %	509 kPa	20 kPa	692 kPa	462 kPa

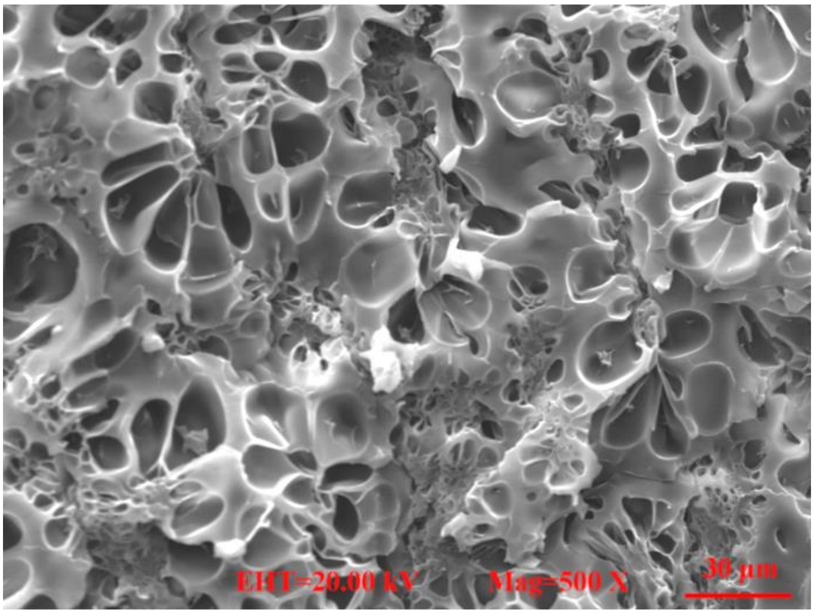

Figure S8. SEM image showing the porous structure of the 60 wt % IL-containing supramolecular ionogel after removal of the IL, indicating the homogenous and continuous dispersion of the IL in the PUU network.

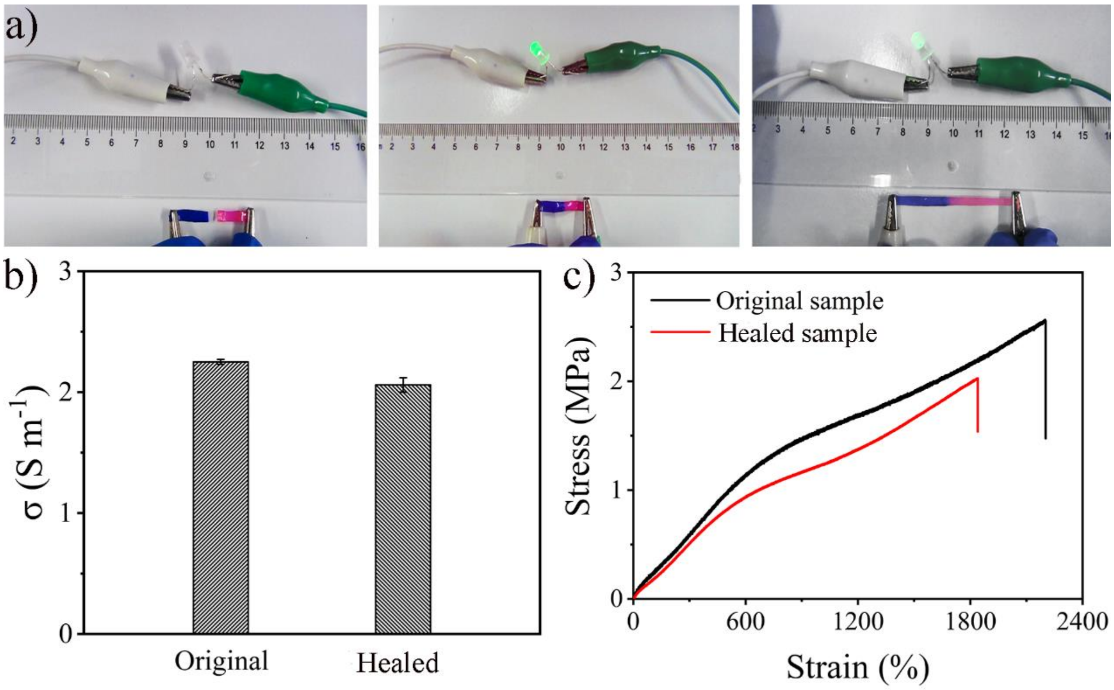

Figure S9. (a) Photo images showing the methanol assisted healable behavior of the 60 wt % IL-containing supramolecular ionogel. (b) The ionic conductivity of the original and healed60 wt % IL-containing supramolecular ionogel films. (c) Tensile testing curves of the original and healed 60 wt % IL-containing supramolecular ionogel films.

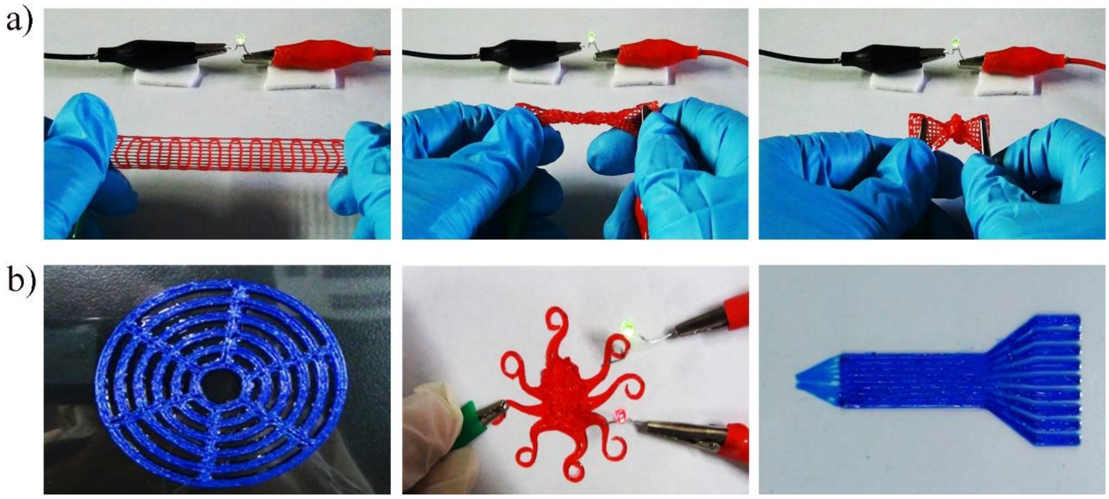

Figure S10. 3D-printed free-standing and stretchable soft ionic conductive structures. (a) 3D-printed mesh film still shows excellent ionic conductivity under stretching, knotting and twisting states. (b) 3D-printed spider web, octopus and electrode. Red or Blue dyes were used for easy observation.

Movie S1. Squeezing test on the 60 wt % IL-containing supramolecular ionogel showing no leakage of IL on the plain paper under the vigorous squeezing, indicating the IL is effectively embedded in the PUU matrix.

Movie S2. 3D printing process of the 60 wt % IL-containing supramolecular ionogel based spider web structure. The ink is colorless transparent, so Reactive Blue 19 was added for easy observation.

Movie S3. The 3D-printed ionogel mesh film with a length of 60 mm, width of 20 mm and thickness of 0.5 mm even could withstand a weight of 1.0 kg loading (~3400 times its own weight, 0.29 g).

Movie S4. A 0.47 g 3D-printed cuboid specimen with a dimension of length × width × height = 15 mm × 15 mm × 5 mm could bear ~ 10000 times (4.7 kgf) its own weight without fracture and immediately recover to its original shape after the load is removed.