Supporting Information for

A Self-Healing and Highly Stretchable Polyelectrolyte Hydrogel via Cooperative Hydrogen-Bonding as Superabsorbent Polymer

Esra Su, Mine YURTSEVER, and Oguz Okay[*]

Department of Chemistry, Istanbul Technical University, Maslak, 34469 Istanbul, Turkey

Table of Contents

Figure S1. The weight swelling ratio m_{rel} and water content of PAMPS hydrogels as a function of AMPS content.

Figure S2. Frequency ω dependences of the storage modulus G' (filled symbols), loss modulus G'' (open symbols), and loss factor $tan \, \delta$ (lines) of T-50 and U-50 gels at 25 °C. $\gamma_0 = 0.01$.
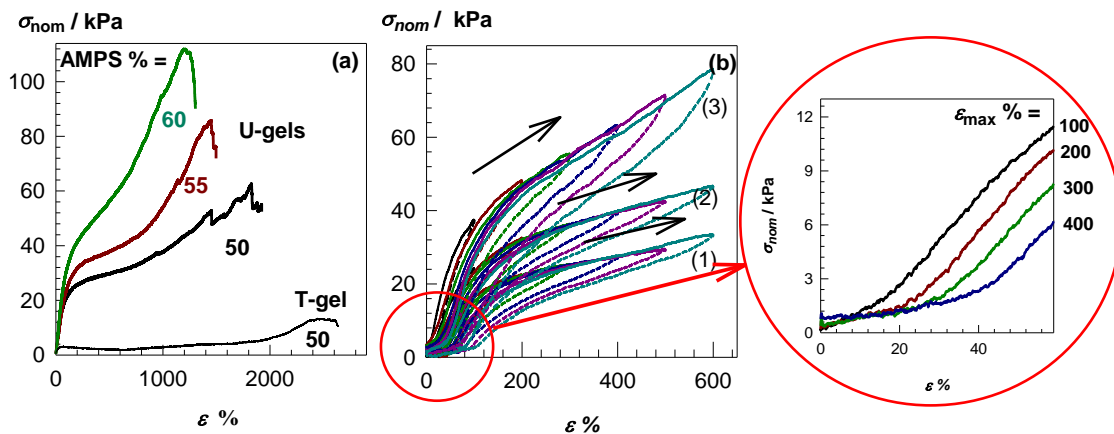
Figure S3. (a): Tensile stress-strain curves of U- and T- hydrogels. AMPS contents as indicated. **(b):** Cyclic tensile tests conducted on U-gels up to a maximum strain ε_{max} of 600% with 100% steps.

Loading curves are indicated by arrows. AMPS = 50 (1), 55 (2), and 60 wt % (3). The inset shows the initial portion of the loading curves.
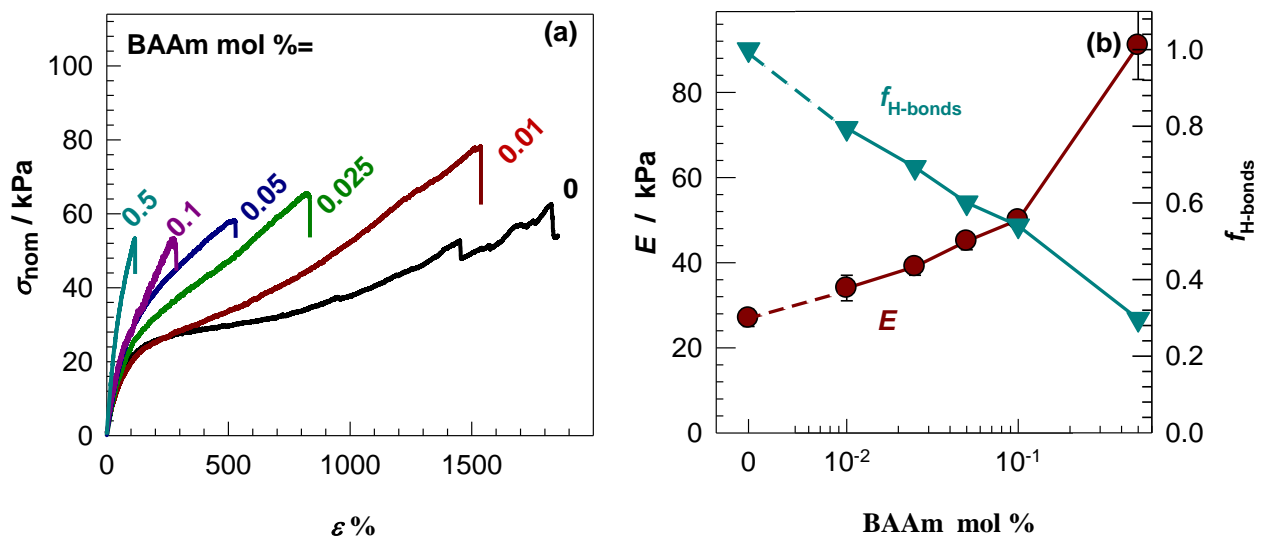
Figure S4. (a): Stress-strain curves of PAMPS hydrogels prepared in the presence of various amounts of BAAm cross-linker. AMPS = 50 wt %. **(b):** Young' modulus E of PAMPS hydrogels and the fraction $f_{H\text{-bonds}}$ of cross-links contributed from the H-bonds as a function of BAAm content. AMPS = 50 wt.%. For hydrid cross-linked hydrogels containing both physical and chemical cross-links, one may calculate the fraction $f_{H\text{-bonds}}$ of cross-links contributed from the H-bonds by comparing the moduli of hydrogels prepared with and without BAAm. Because the cross-link density at a given polymer concentration is directly proportional to the modulus E, $f_{H\text{-bonds}}$ was estimated using the equation $f_{H\text{-bonds}} = E_0/E$ where E_0 and E are Young's moduli of PAMPS hydrogels without and with BAAm, respectively.
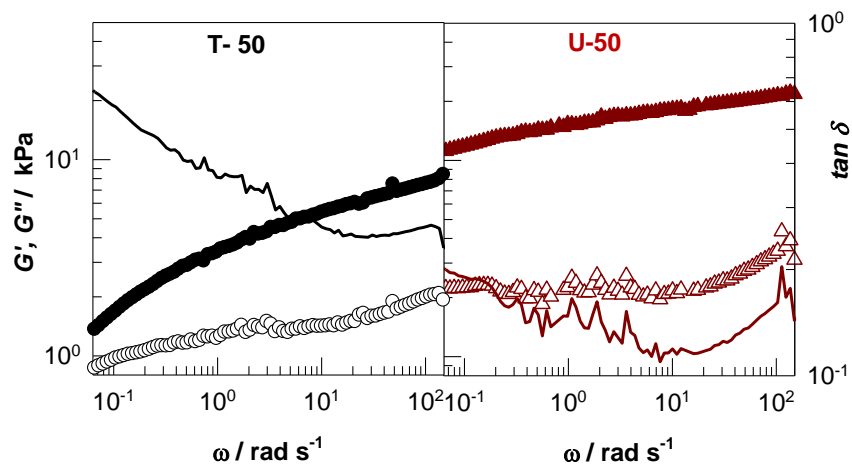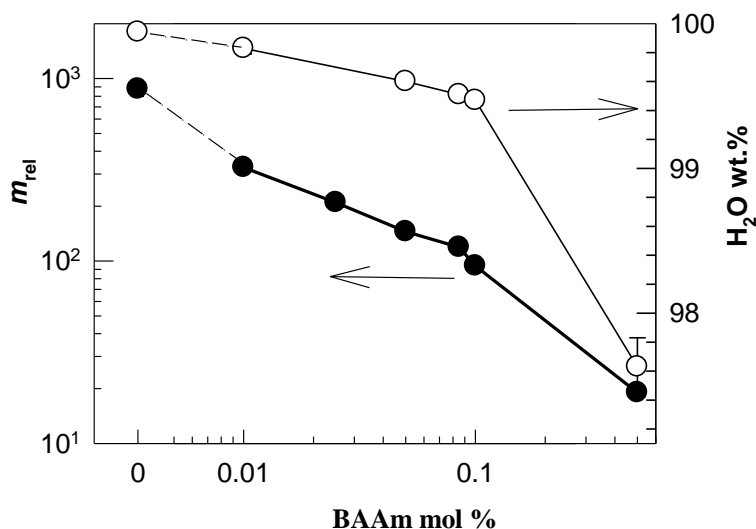

Figure S5. The weight swelling m_{rel} and water contents of PAMPS hydrogels as a function of BAAm content. AMPS = 50 wt %.

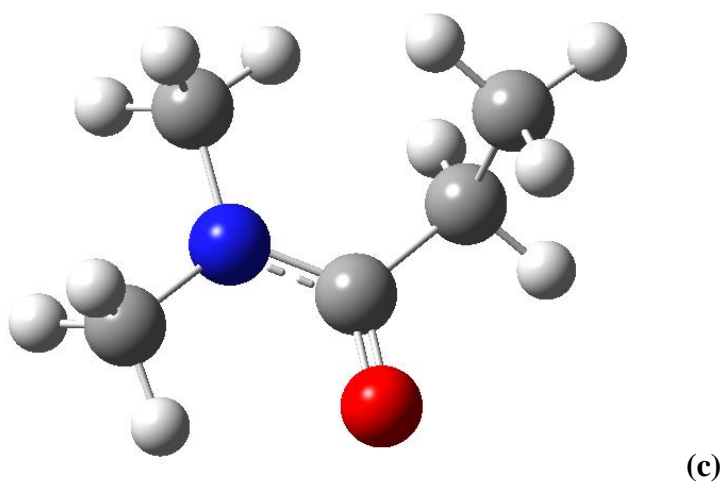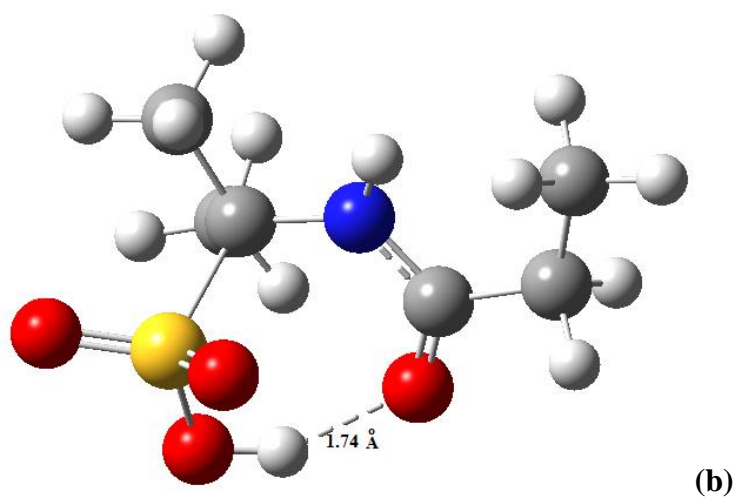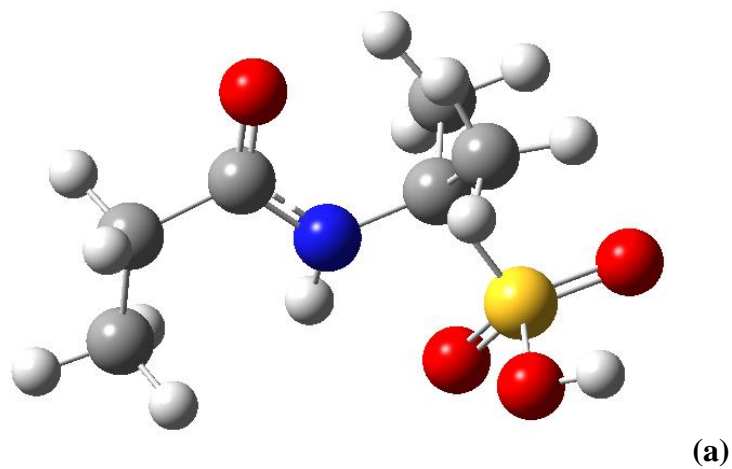**(a)**

(b)

(c)

Figure S6. The optimized geometries of AMPS in which C=O and N-H bonds are trans (a) and cis (b) to each other, and DMAA (c).
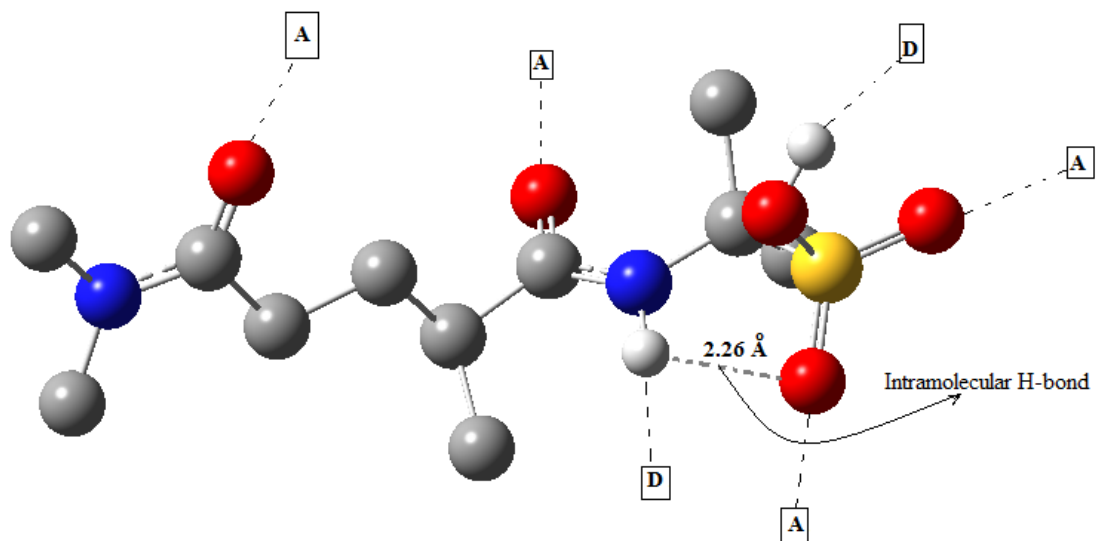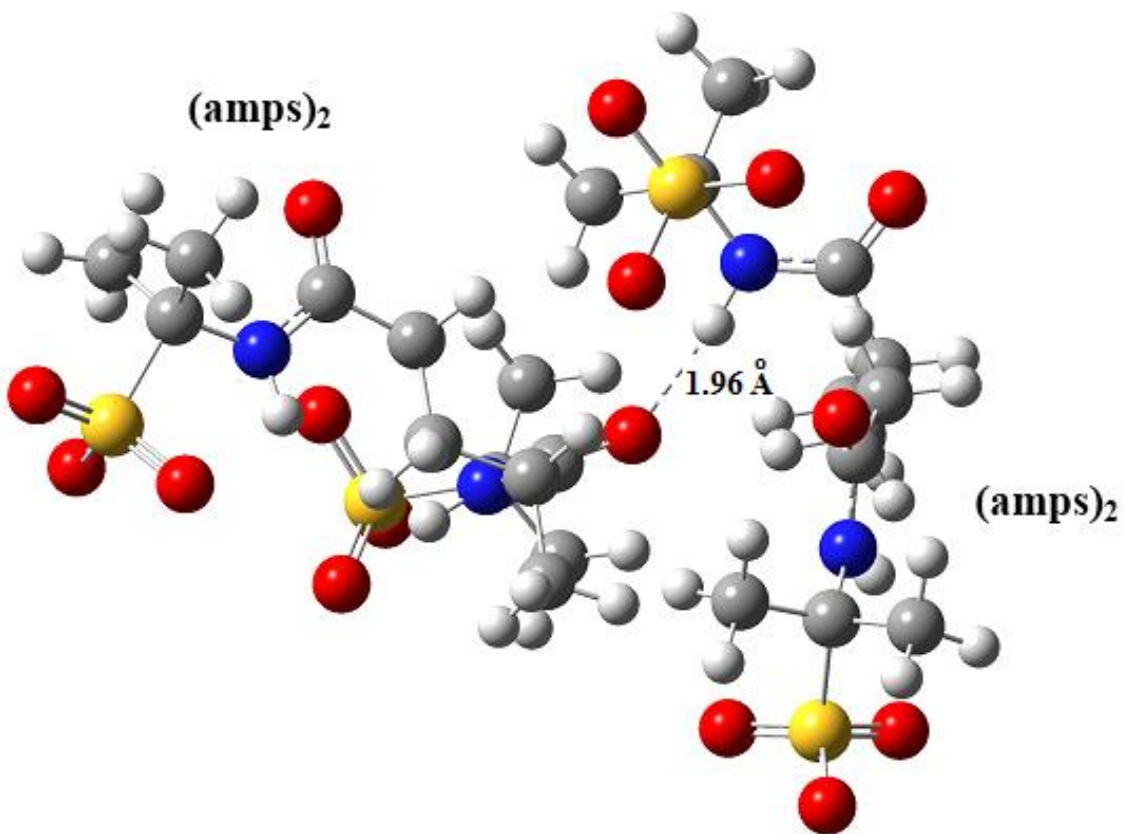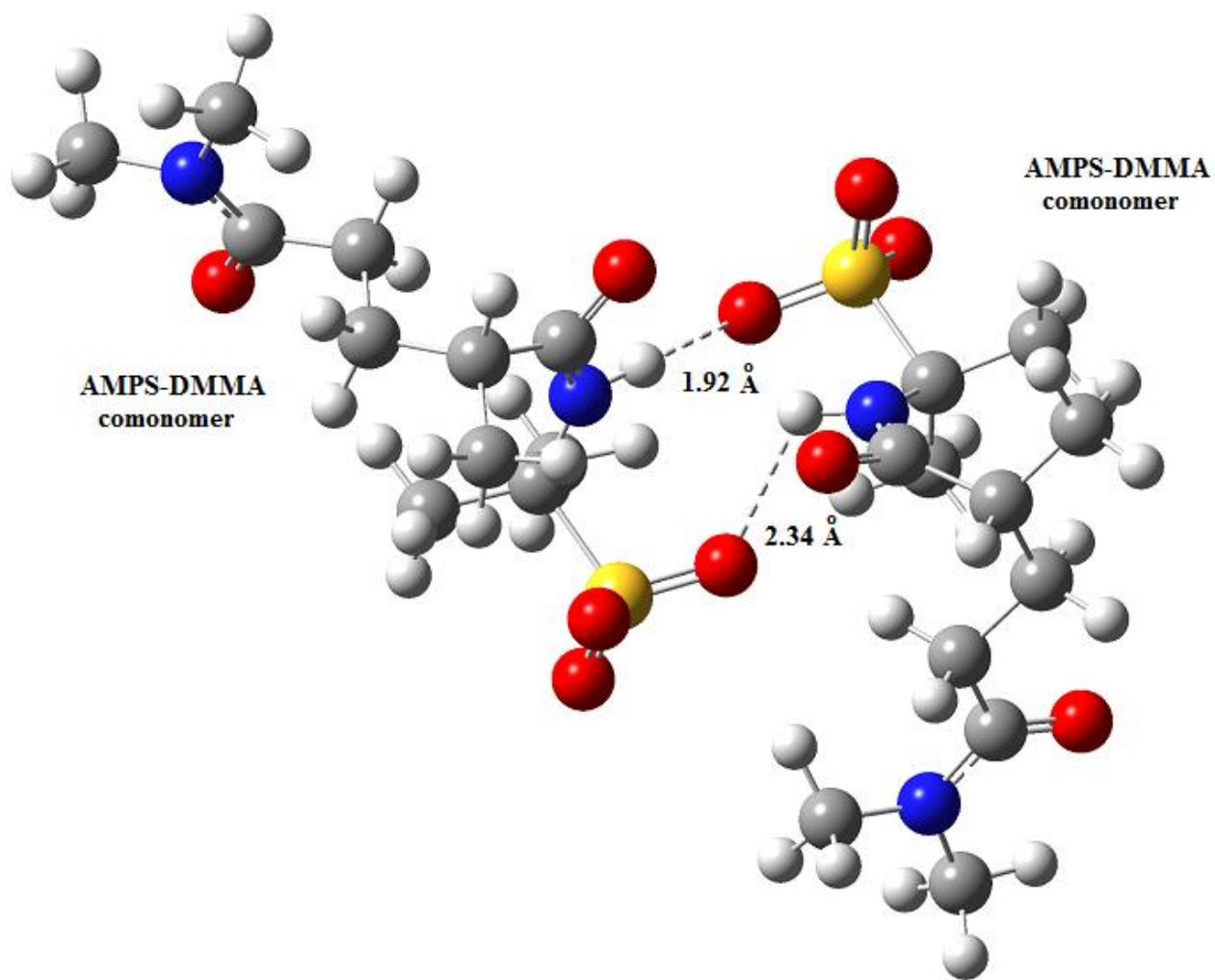
Figure S7. Optimized geometry of AMPS-DMAA dimer. D and A represent H-bond donor and acceptor sites on the molecule, respectively.
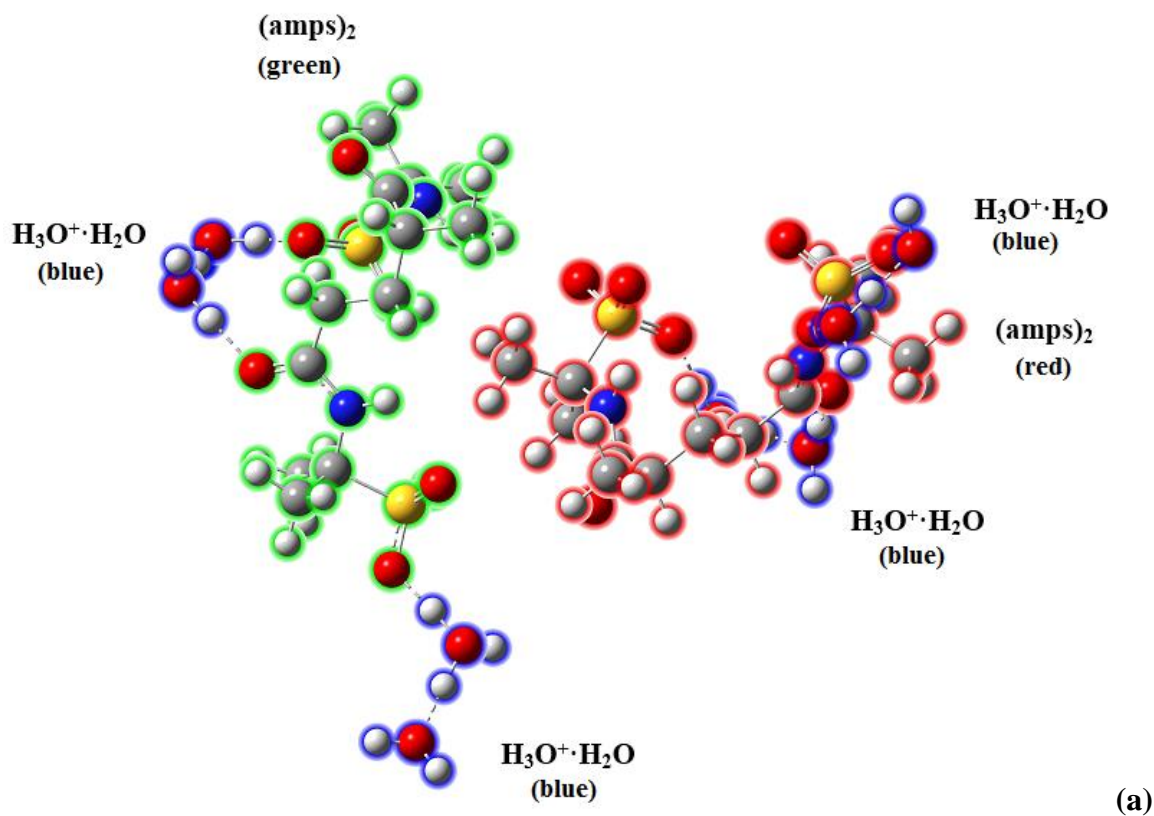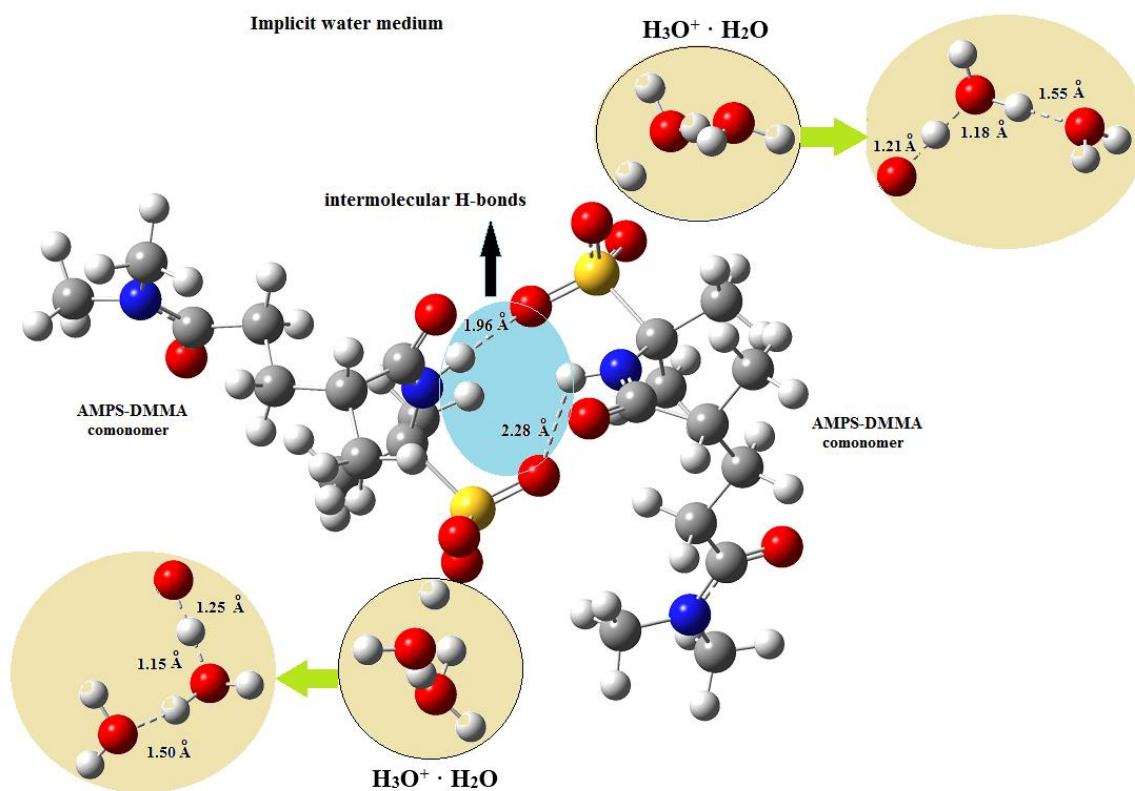

(a)

(b)

Figure S8. Optimized geometries of $(AMPS)_2 - (AMPS)_2$ (a) and AMPS/DMAA - AMPS/DMAA dimers (b) in -4 and −2 anionic states, respectively, in implicit water medium.

Figure S9. Optimized geometries of (AMPS)$_2$ - (AMPS)$_2$ (a) and AMPS/DMAA - AMPS/DMAA dimers (b) in neutral state in explicit water medium in the presence of H$_3$O$^+$·H$_2$O per SO$_3^-$ group.
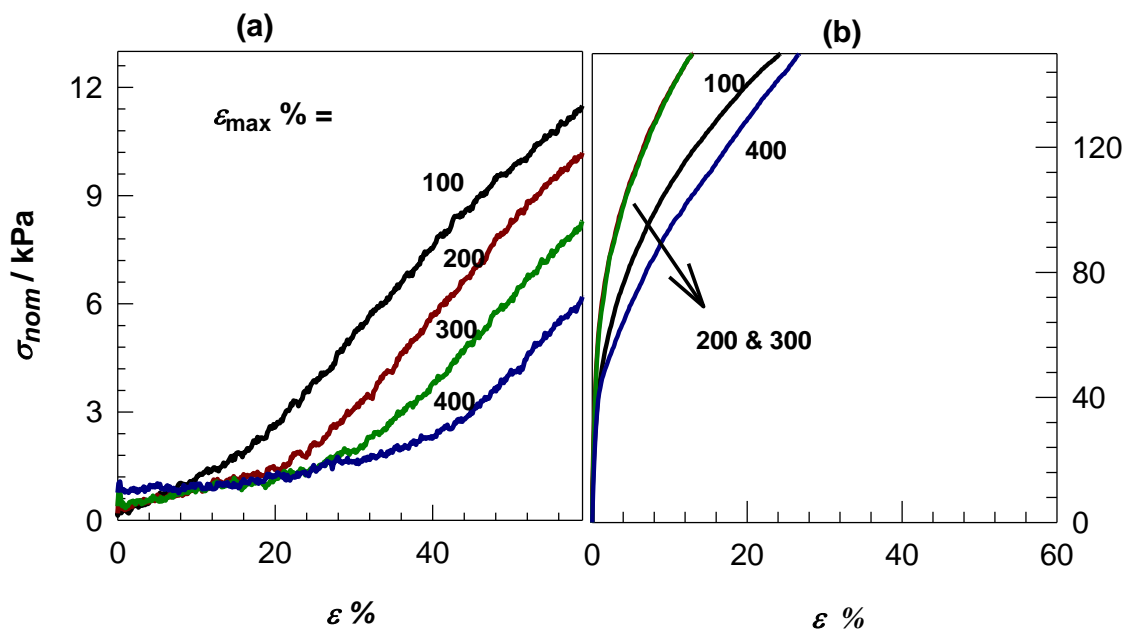
Figure S10. Initial portion of the loading curves of successive cyclic tests conducted on U-50 (a) and U-75/0.6 hydrogels (b).
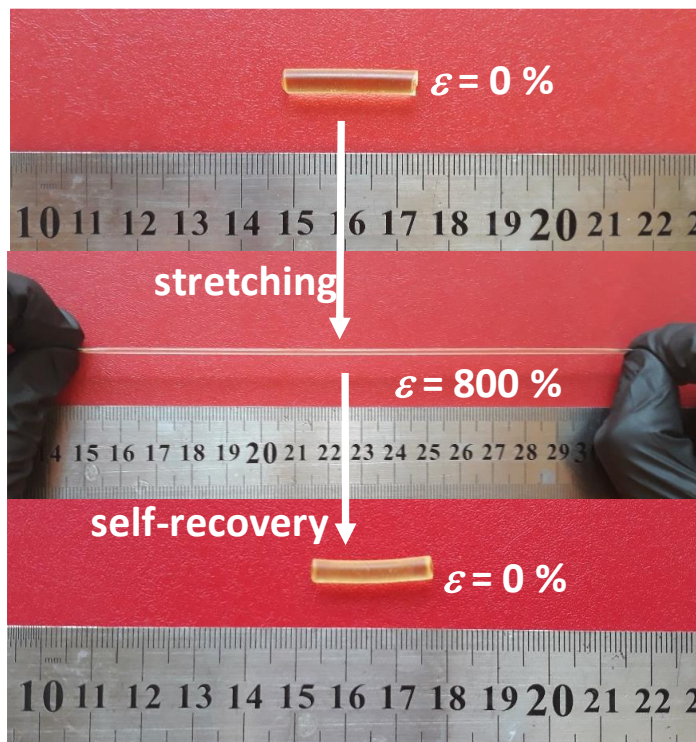

Figure S11. Self-recovery behavior of a rod-shaped U-75/0.6 gel specimen after stretching to 800 % elongation ratio.