Supporting Information

Super Strong and Tough Hydrogel through Physical Crosslinking and Molecular Alignment

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Figure S1. Images of a) PVA-TA30%, and (b) PVA-TA0% hydrogels before (left) and after immersed in saturated NaCl solution (right).



Figure S2. a) Swelling ratios of the PVA-TA0% and PVA-TA30% hydrogels before and after immersed in NaCl solutions. b) Images of the PVA-TA30% (left) and PVA-TA30%-S-NaCl (right) hydrogels in the swelling process. c) Images of the PVA-TA0% (left) and PVA-TA0%-S-NaCl (right) in the swelling process.



Figure S3. Compressive stress-strain curves of the PVA-TA0%, PVA-TA30%, PVA-TA0%-S-NaCl, and PVA-TA30%-S-NaCl hydrogels.



Figure S4. a-b) Tensile stress-strain curves of PVA hydrogels with different TA concentrations before and after immersed in saturated NaCl solutions for 12h. c-d) Elastic moduli and toughnesses of PVA-TA hydrogels before and after immersed in saturated NaCl solutions.



Figure S5. a-b) Tensile stress-strain curves, elastic moduli and toughnesses of PVA-TA30%-S-NaCl hydrgoels with different cycles of freezing/thawing treatment. Tensile stress-strain curves, elastic moduli and toughnesses of PVA-TA30%-y-NaCl hdyrgoels c-d) with different immersing times, and e-f) immersed in different concentrations of NaCl solutions.



Figure S6. Wet-drawing PVA-TA mixtures in a) saline b) water and c) ethanol.



Figure S7. H&E stained images of wounding closing site (a mouse's subcutaneous tissue) with PVA-TA suture (arrows show suturing sites, scale bar = 10 mm) after 6 days.



Figure S8. H&E stained connective tissue around the implanted PVA/TA suture (arrows show the implantation sites, scale bar = 10 mm).

Supporting Movies

- **M1.** The PVA-TA30% hydrogel ball was used to play ping pong. It is bounced and hit like a ping-pong ball.
- M2. The PVA-TA30% hydrogel tube was repeated browned into a balloon via an air pump.
- M3. Video of wet-drawing PVA-TA30% mixtures in different solvents.