Supporting Information

High strength, Self-adhesive and Strain-sensitive Chitosan/Poly(acrylic acid) Double-network Nanocomposite Hydrogels Fabricated by Salt Soaking Strategy for Flexible Sensors

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Figure S1. FTIR spectrum of CNC, TA and TA@CNC.



Figure S2. Digital images of CNC and TA@CNC in CS acid solution (pH = 3) at concentration of 0.5 wt%.



Figure S3. SEM images of interconnected filamentous porous structures formed in the CS/PAA-TA@CNC-60.



Figure S4. FTIR spectrum of CS, PAA and CS/PAA-TA@CNC-60.



Figure S5. Frequency dependence of the loss tangent (tan δ) (a), storage modulus (G') and loss modulus (G'') (b) of the CS/PAA-TA@CNC DN nanocomposite hydrogels with different soaking times (10, 30 and 60 min).

The shear frequency dependent viscoelastic properties of CS/PAA-TA@CNC DN nanocomposite hydrogels can be explained by the classic rubber elastic theory, where the network modulus *G* of the hydrogels can be calculated by $G = G_c + G_p$, where G_c and G_p are network modulus contributed by covalent crosslinks and noncovalent cross-links, respectively. In this respect, the contribution to network modulus from entangled CS chains is related to the immersion time in NaCl solution, which is frequency-dependent caused by the stress-relaxation of the entanglement. That is, the high density of entanglement is, the longer time is required for the entangled chains to pass through each other and become unraveled. Specifically, at low frequency, the entangled chains can gradually unravel and hardly contribute to the network modulus, and the *G*' is dominated by the covalent cross-links G_c . However, with the increase of frequency, the chain-entanglement of CS in the network has no enough time to relax, and both G_c and G_p contribute to the network modulus thereby.

Rheological Measurements.

The storage modulus (G') and loss modulus (G") of CS/PAA-TA@CNC DN nanocomposite hydrogel disks (15 mm in diameter) with different soaking times were tested by a rheometer (TA AR2000) fitted with parallel plates (15 mm in diameter). Under a fixed strain level (10%), the angular frequency was swept from 0.1 to 1000 rad/s. The loss tangent (tan δ) was calculated from G''/G'.



Figure S6. Frequency dependence of the viscosity of the CS/PAA-TA@CNC DN nanocomposite hydrogels with different soaking times (10, 30 and 60 min).



Figure S7. Dissipated energy with different resting time.



Figure S8. Fifty cyclic loading-unloading test of CS/PAA-TA@CNC-60 min at 100% strain.



Figure S9. Antibacterial annulus of CS/PAA-TA@CNC-60 to (a) Colibacillus and (b) Staphylococcus aureus.

Anti-bacterial Tests.

For anti-bacterial test, solid medium was initially prepared. In brief, 2 g peptone, 1 g yeast and 2 g NaCl were dissolved in 200 mL deionized water with pH adjusted to 7.2 by 1 M NaOH solution, then 3 g agar powder was dissolved into the mixture to obtain precursor solution, which was subsequently sterilized in high-pressure steam sterilization pot under 0.142 MPa and 126 °C for 30 min to thoroughly kill bacterial. The bioclean precursor solution was then poured into disposable petri dish to form solid medium. The hydrogel samples were submerged into ultrapure water for 48 h before soaked into saturated NaCl solution to totally remove residual toxic monomer, followed by ultraviolet sterilization for 20 min. Finally, 0.5 mL colibacillus and staphylococcus aureus nutrient solution was spin coated uniformly onto solid medium surface respectively with a round piece of hydrogel sample (1 cm in diameter) attached on the coated surface afterward. The prepared samples were put into constant temperature oscillator under 40 °C for 12 h to promote the growth of bacterial.