In-situ polymerization induced supramolecular hydrogels of chitosan

and poly(acrylic acid-acrylamide) with high toughness

Jin Li, Zhilong Su, Xiaodong Ma, Hongjie Xu, Zixing Shi, Jie Yin, Xuesong Jiang* School of Chemistry & Chemical Engineering, State Key Laboratory for Metal Matrix Composite Materials, Shanghai Jiao Tong University Shanghai 200240, People's Republic of China.

Tel.: +86-21-54743268; Fax: +86-21-54747445. E-mail: ponygle@sjtu.edu.cn

Results and discussion



Figure S1. The cross-section surface morphology of the obtained hydrogels. Representative SEM images of cross-section surface morphology of (a) hydrogel-1, (b) hydrogel-2, (c) hydrogel-3, (d) hydrogel-4, (e) hydrogel-6.



Figure S2. Control experiments. (a) Chemical structure of component used in control tests; (b) Digital photographs of controlled gel-formation tests, (1) 2% CS-10% HAc - 5% AM, (2) 2% CS - 10% AA, (3) 2% MBA - 10% AA - 5% AM, (4) 2% PAAM - 10% AA - 5% AM (PAAM/P(AA*r*-AM)), (5) 2% CS - 10% AA - 5% AM (hydrogel-**5**); (c) Compressive and tensile experiments of chemical P(AA-*r*-AM) hydrogel using MBA as cross-linker; (d) Digital photograph of 2% PAAM - 10%AA - 5%AM (PAAM /P (AA -*r*-AM)) hydrogel withstand a certain weight; (e) Digital photograph of 2% CS - 10 % AA - 5% AM (hydrogel-**5**) hydrogel withstand a certain weight.



Figure S3. Loading-unloading cycling tests of hydrogel-4. (a) Six successive loading-unloading cycles of the hydrogel-4; (b) the recovery of elastic modulus and hysteresis loop area calculated from (a).



Figure S4. Fatigue resistance of hydrogel-4.Cycle compression test of hydrogel-4 undergoing 10 cycles of compression to 80% strain (a); (b) the caculated total toughness and the dissipated toughness of hydrogel-4 during loading-unloading tests based on (a).



Figure S5. Self-healing property. (a) Cycle compression tests of hydrogel-**5** of compression to 80 % strain before self-healing and undergoing 3 cycles after healed for 24 h. (b) the calculated total toughness and the dissipated toughness of hydrogel-**5** before self-healing and after self-healing based on (a).



Figure S6. Self-healing properity. (a) Cycle compression tests of hydrogel-4 of compression to 80%

strain before self-healing and undergoing 3 cycles after healed for 24h. (b) the caculated total toughness and the dissipated toughness of hydrogel-4 before self-healing and after self-healing based on (a).



Figure S7. Stability of the hydrogel-**5**. Digital photographs of hydrogel-**5** before and after swelling in acidic and alkaline conditions.



Figure S8. (a) Saturated adsorption capacities of hydrogel-5 for seven metal ions, (b) the compression tests of hydrogel-5 soaked in different metal ions' solution(c=0.001mol/L).