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Supporting Information

Chitosan-based double cross-linked ionic hydrogels as a strain and

pressure sensor with broad strain-range and high sensitivity

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Cytotoxicity Tests of the P(AAm-co-AA)/CS-Fe³⁺ Hydrogel

NIH3T3 cells (mouse embryonic fibroblasts) (iCell Bioscience Inc) were used for the cytotoxicity assay. The growth media (Dulbecco's modified eagle medium, DMEM, Coring) was supplemented with 10% Fetal Bovine Serum (FBS) (Shanghai yuanye Bio-Technology Co., Ltd), 1% sodium pyruvate (Geno Biomedical Technology Co., Ltd) and 1% glutamine (Geno Biomedical Technology Co., Ltd). NIH3T3 cells were seeded on the 96-well plates (6000 cells/well) with the growth media (200 μ L) in an incubator (5% CO₂, 37 °C). After 24 h, a certain amount of P(AAm-co-AA)/CS-Fe³⁺ hydrogel (0.99 mg) was added. After being incubated with the hydrogel, the cell viability was measured by CCK-8 assay. Typically, 100 μ L of CCK-8 solution (Invigentech) was added to each well of the plates. NIH3T3 cells were incubated for another 2 h. Afterward, the media were transferred into another 96-well plate to measure the absorbance at 450 nm by using a microplate reader.

Antimicrobial Tests of the P(AAm-co-AA)/CS-Fe³⁺ Hydrogel

Escherichia coli (ATCC25922, *E. coli*), *Staphylococcus aureus* (ATCC25923, *S. aureus*) and *Pseudomonas aeruginosa* (CMCC(B)10104, *P. aeruginosa*) were used to test the antibacterial activity of the P(AAm-co-AA)/CS-Fe³⁺ hydrogel *via* an inhibition zone method. The three kinds of bacteria were activated in the Luria–Bertani (LB) broth (37 °C, 180 rpm) for 24 h. Then the concentration of the bacteria suspensions was adjusted to 10⁷ CFU/mL. 90 μ L of each suspension was seeded to the plates of LB culture medium. The hydrogel samples with a regular size (cylinder shape with a diameter of 6 mm) were exposed to the three kinds of bacteria suspension on the plates respectively. All samples were incubated (37 °C, 180 rpm) for 24 h, and then the inhibition zone was observed and measured. In addition, drug sensitive paper and 0.9 %g/L NaCl were used as positive control and negative control respectively.

Sample	Mass of reagents (g)				
	CS	AA	AAm	Irgacure 2959	H ₂ O
P(AAm-co-AA)	0	0.144	1.42	0.025	10
P(AAm-co-AA)/CS	0.2	0.144	1.42	0.025	10
P(AAm-co-AA)/CS-Fe ³⁺ -1	0.2	0.086	1.42	0.025	10
P(AAm-co-AA)/CS-Fe ³⁺ -2	0.2	0.115	1.42	0.025	10
P(AAm-co-AA)/CS-Fe ³⁺ -3	0.2	0.144	1.42	0.025	10
P(AAm-co-AA)/CS-Fe ³⁺ -4	0.2	0.172	1.42	0.025	10
P(AAm-co-AA)/CS-Fe ³⁺ -5	0.1	0.144	1.42	0.025	10
P(AAm-co-AA)/CS-Fe ³⁺ -6 ^a	0.2	0.144	1.42	0.025	10
P(AAm-co-AA)/CS-Fe ³⁺ -7	0.3	0.144	1.42	0.025	10

Table S1. The dosage of each reagent used in the fabrication process of hydrogels.

^a The sample for extensive mechanical and sensing performance study.

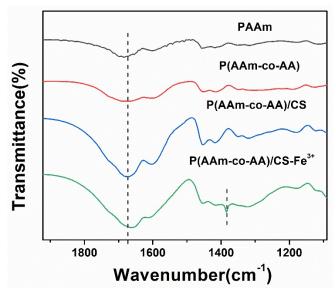


Figure S1. FTIR spectra of PAAm, P(AAm-co-AA), P(AAm-co-AA)/CS, and P(AAm-co-AA)/CS-Fe³⁺.

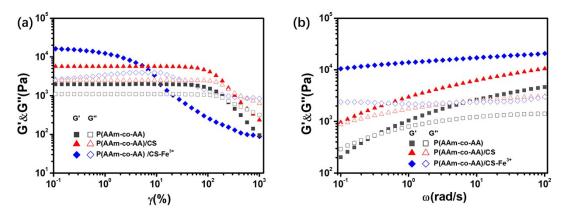


Figure S2. Storage modulus (G') and loss modulus (G'') of P(AAm-co-AA), P(AAm-co-AA)/CS and P(AAm-co-AA)/CS-Fe³⁺ hydrogels in amlitude sweep (a) and frequency sweep (b).

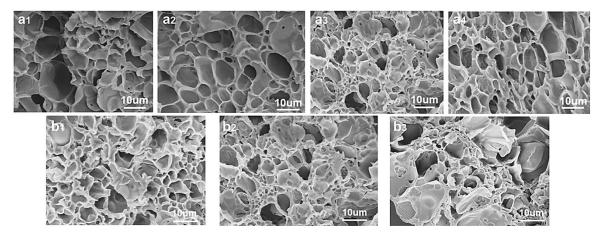


Figure S3. SEM images of the cross section of the P(AAm-co-AA)/CS-Fe³⁺ hydrogel with different molar ratios of AA/AAm (a1: 6 mol%, a2: 8 mol%, a3: 10 mol%, and a4: 12 mol%) and different CS contents (b1: 1 wt%, b2: 2 wt%, and b3: 3 wt%).

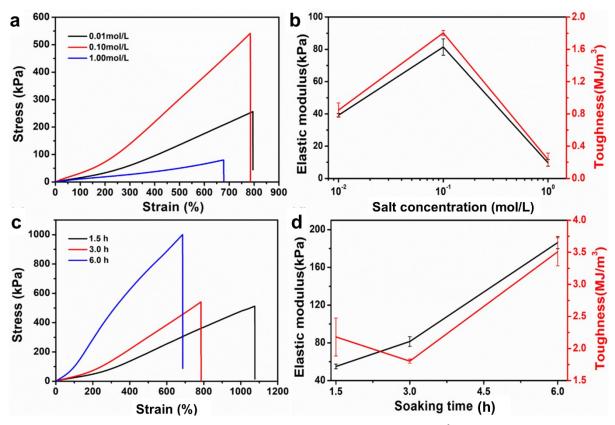


Figure S4. The stress-strain curves of the $P(AAm-co-AA)/CS-Fe^{3+}$ hydrogel prepared by soaking in $Fe(NO_3)_3$ solution with different concentrations (a) and soaking times (c); elastic modulus and toughness of the hydrogels as a function of $Fe(NO_3)_3$ concentration (b) and soaking time (d)

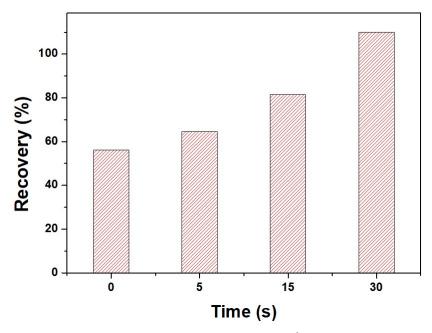


Figure S5. The recovery rate of the P(AAm-co-AA)/CS-Fe³⁺ hydrogel at different rest times in cyclic tensile tests.

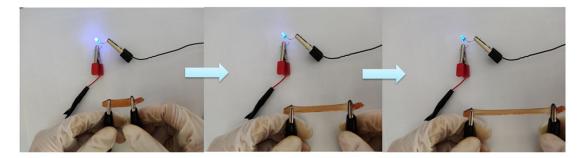


Figure S6. The LED brightness varies with different deformation.

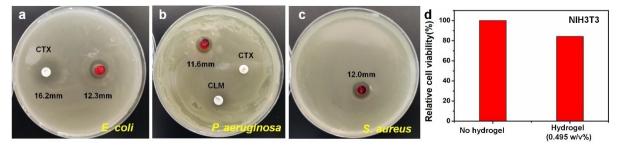


Figure S7. Antibacterial activity of the P(AAm-co-AA)/CS-Fe³⁺ hydrogel on E. coli (a), P. aeruginosa (b) and S. aureus (c) evaluated by an inhibition zone method and relative viability of NIH3T3 cells incubated with the P(AAm-co-AA)/CS-Fe³⁺ hydrogel by using a CCK-8 method. The red sample in a-c is the P(AAm-co-AA)/CS-Fe³⁺ hydrogel and the white disc is cefotaxime (CTX) or chloramphenicol (CLM) used as positive control.