Supporting Information

Mussel-inspired ultra-stretchable, universally sticky, and highly-conductive

nanocomposite hydrogels

Qin Chen¹, Lan Feng¹, Huitong Cheng¹, Yilin Wang¹, Hao Wu², Tao Xu¹, Weifeng

Zhao ^{1,*}, Changsheng Zhao ^{1,3,4}

¹ College of Polymer Science and Engineering, State Key Laboratory of Polymer

Materials Engineering, Sichuan University, Chengdu, 610065, China.

² Department of Orthopedics, West China Hospital, Sichuan University, Chengdu,
610041, China.

³ College of Biomedical Engineering, Sichuan University, Chengdu, 610064, China

⁴College of Chemical Engineering, Sichuan University, Chengdu, 610065, China

* Corresponding author

E-mail: <u>zhaoscukth@163.com</u>; <u>weifeng@scu.edu.cn</u> (Weifeng Zhao)

Sample name	Py (µL)	DA (g)	$FeCl_3 \cdot 6H_2O(mg)$	Tris(g)	Molar ratio (Py:DA)
1	1.6	0.045	20.4	1.5	0.1
2	16	0.045	20.4	1.5	1
3	80	0.045	20.4	1.5	5
4	160	0.045	20.4	1.5	10

Table S1 The formula for preparing PPy-PDA nanoparticles.

Cytocompatibility Experiment

We conducted *in vitro* biocompatibility tests using the extract medium of hydrogels for cell culture. To prepare the extract medium, we incubated different contents (5, 10, 20 and 30 mg) of the dry PPy-PDA/PAA hydrogels in 1mL Dulbecco's modified Eagle medium (DMEM) at 37 °C for 24 h. Pure PBS was served as a blank group. Pristine DMEM was used as a control group. We analyzed 6 independent samples for each group. L929 fibroblasts were cultured in DMEM added with 10% fetal bovine serum (FBS, Hyclone, USA), and 1% Penicillin Streptomycin in a controlled incubator in 5% CO₂ at 37 °C. L929 fibroblasts were seeded in 96-well plates (5×10^2 cells/well) for culturing 12 h after treating with trypsin. The cells were then treated with the extract medium and incubated in 5% CO₂ at 37 °C for 24, 72 and 120 h, respectively. Cell viability was assessed by adding 10 µL Cell Counting Kit-8 (CCK-8) reagent into the culture medium and incubated at 37 °C for 1 h, and then optical density (OD) value at 450 nm was determined by a microplate reader.

L929 fibroblasts with a density of 1×10^4 cells/well were seeded into 24-well

plates with treated hydrogels and then incubated at 37 °C for 24, 72 and 120 h. Next, the medium was carefully removed and washed three times with PBS. After that, cells were fixed using 4% paraformaldehyde at 4 °C for 15 min, and incubated in 2.5% Triton X-100 at 37 °C for 10 min. The cells were then thoroughly washed with PBS and incubated with a phalloidin (1:1000) diluted in PBS at 37 °C for 1 h in the dark. Finally, the cells were stained with DAPI to visualize the nuclei. Fluorescence images were captured by using an inverted fluorescence microscope (Leica, DMi8).

As shown in Figure S1, the peaks displayed at 3408, 2922, 1546, 1466 and 1292 cm⁻¹ would be attributed to -OH/N-H, -C-H, C=C, C=C (benzene ring) and C-C/C-N stretching, respectively. The peaks located at 3408, 1546 and 1292 cm⁻¹ were related to PPy stretching vibrations. The peaks displayed at 3408, 2922 and 1466 cm⁻¹ would be attributed to -OH, -C-H and C=C stretching on the benzene ring, respectively, which were corresponding to the presence of -OH of phenol and C=C functional groups of PDA. The results demonstrated that PPy-PDA nanoparticle had been successfully prepared.



Figure S1. Fourier Transform Infrared Spectroscopy (FTIR) characterization of PPy-PDA nanoparticles.



Figure S2. Schematic diagram of the mechanism of Fe 3+ enhancing the mechanicalpropertiesofPAAhydrogel.



Figure S3. Electrical conductivity of the PPy-PDA/PAA hydrogels with different PPy-PDA nanoparticles (a)Py/DA=1/1 and (b) Py/DA=5/1.