
Tannic Acid-Based Multifunctional Hydrogels with Facile Adjustable Adhesion and Cohesion Contributed by Polyphenol Supramolecular Chemistry

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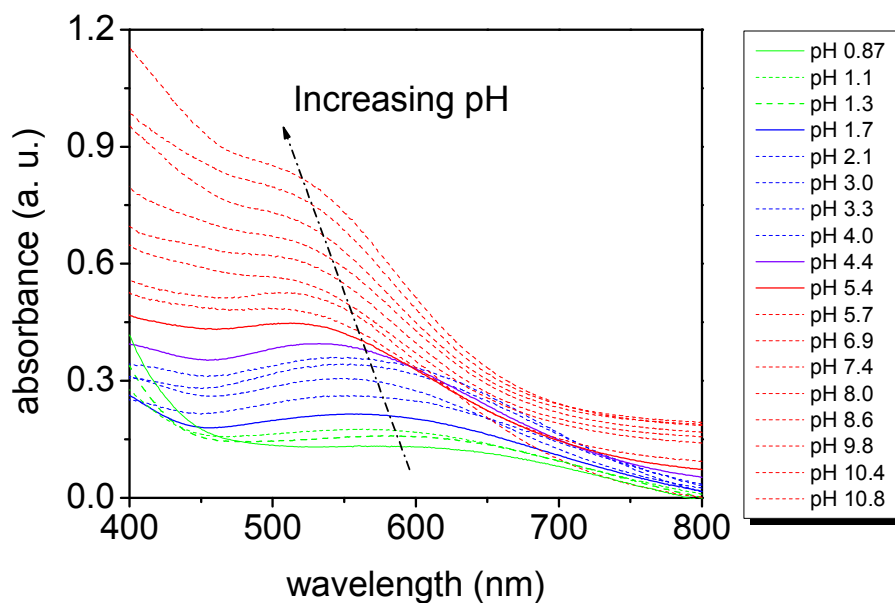


Figure S1. UV-vis absorption PDDA/TA/Fe^{III} complex at pH values from 0.5 to 11.

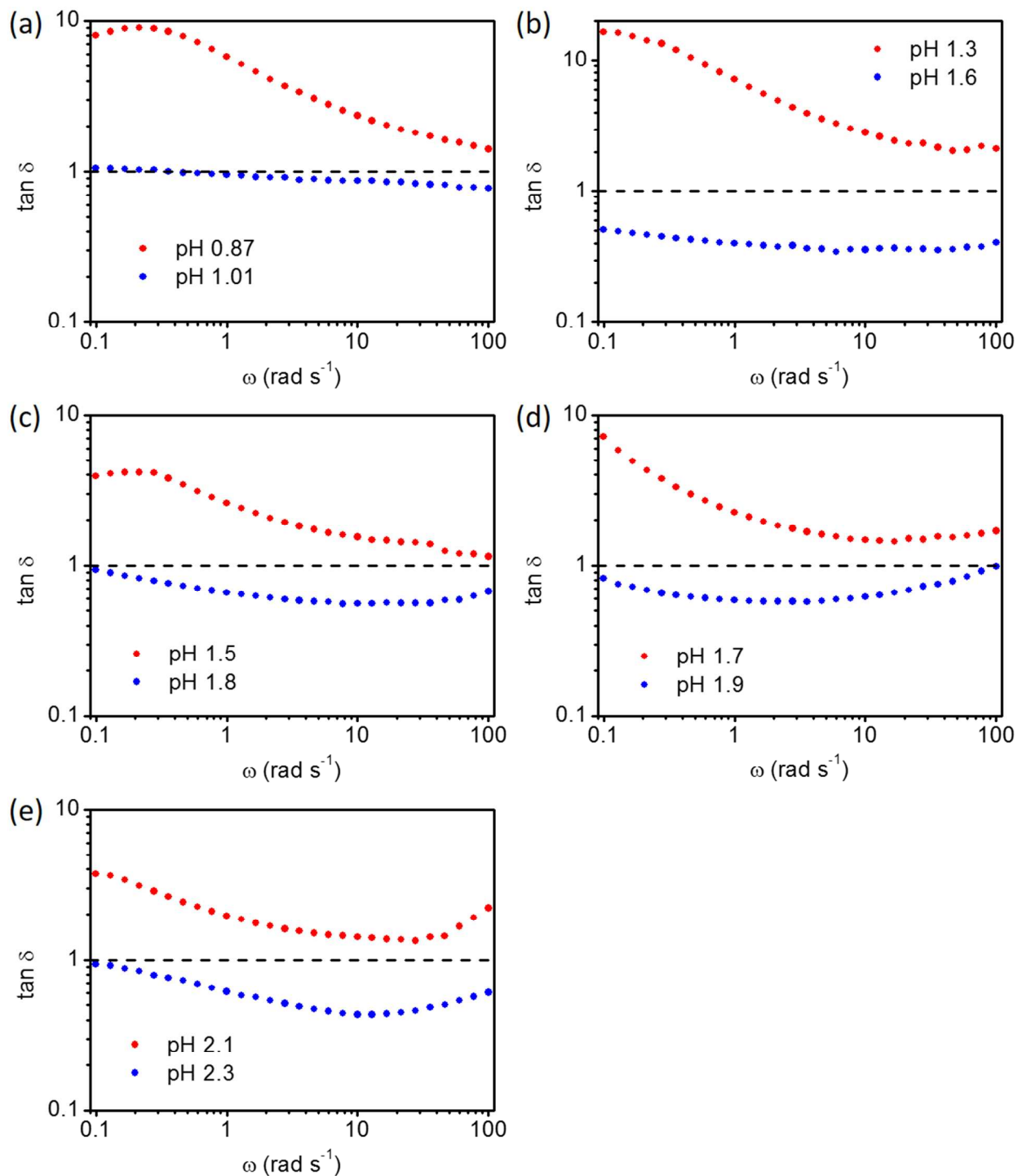


Figure S2. Loss tangent ($\tan \delta$) plotted as a function of the angular frequency of initial PTFe solution and that of hydrogels at critical gelation pH values. (a) PTFe-1, (b) PTFe-2, (c) PTFe-3, (d) PTFe-4, and (e) PTFe-5.

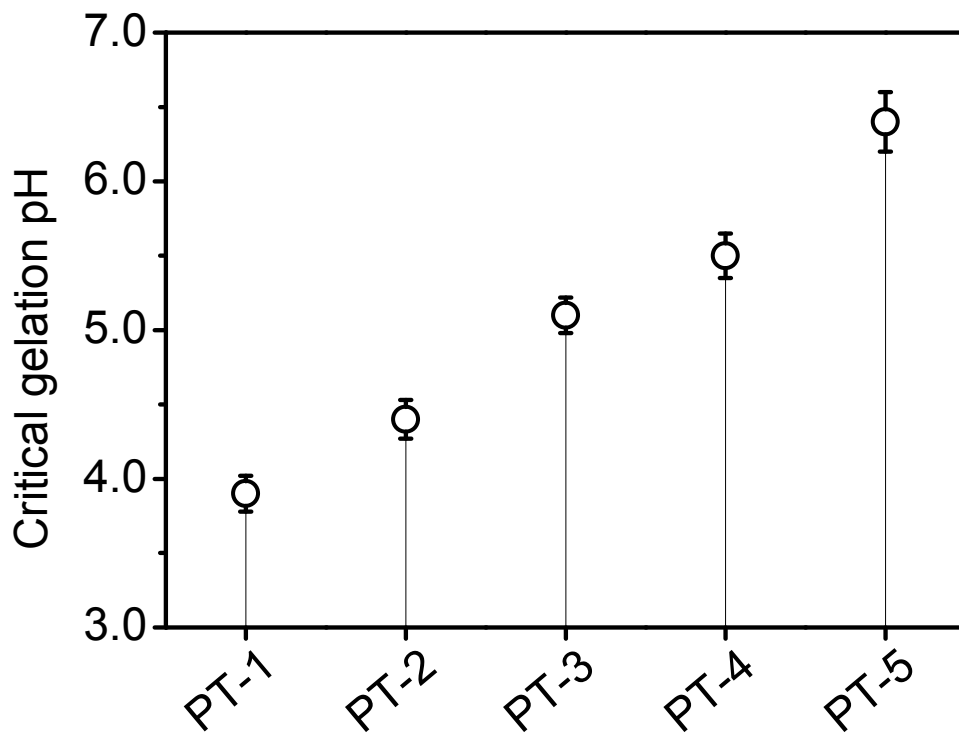


Figure S3. The critical gelation pH of PT hydrogels. The weight ratio of PDDA to TA of PT-1 to PT-5 was 2:1, 4:1, 10:1, 15:1, and 20:1, respectively.

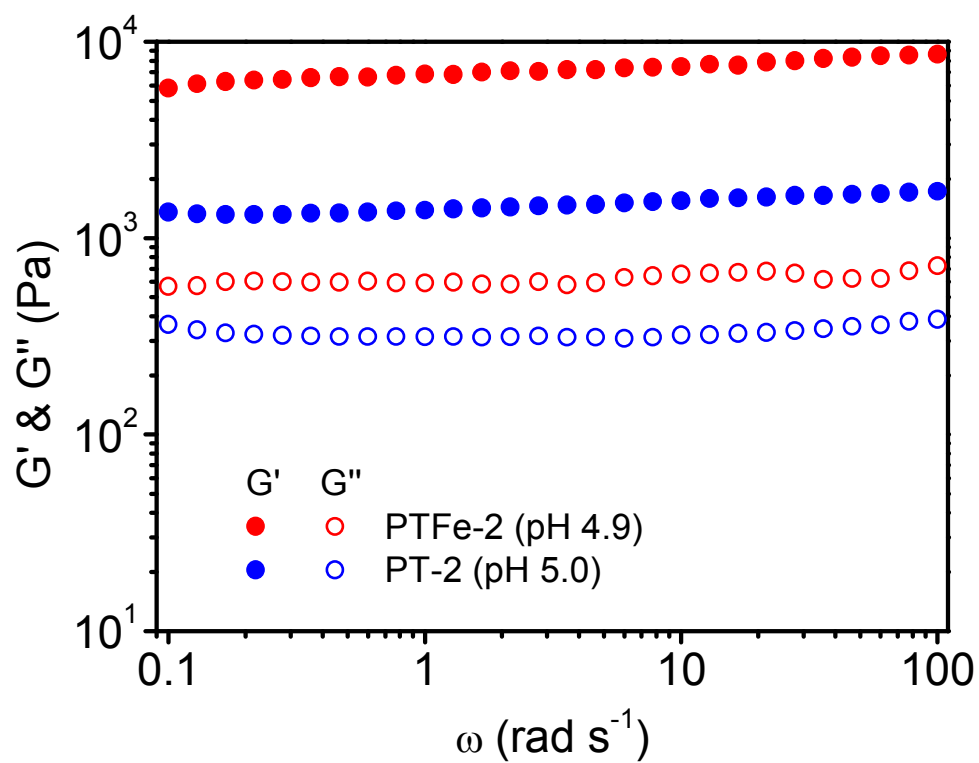


Figure S4. The storage G' and loss G'' moduli of PTFe-2 and PT-2 hydrogels with angular frequency swept between 0.1 and 100 rad s^{-1} . The weight ratio of PDDA to TA of PTFe-2 and PT-2 were 2:1.

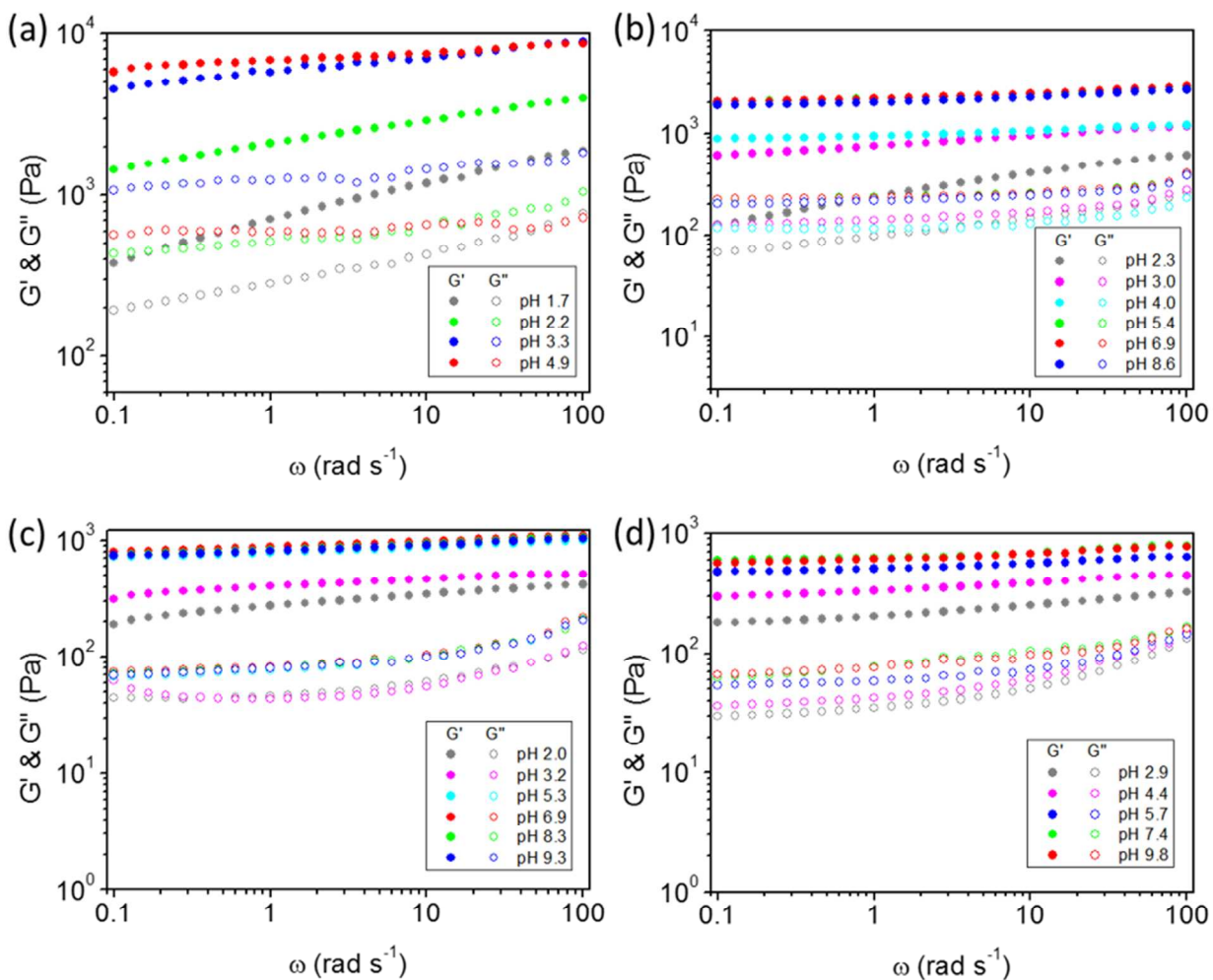


Figure S5. The storage G' and loss G'' moduli of PTFe hydrogels with angular frequency swept between 0.1 and 100 rad s^{-1} . (a) PTFe-2, (b) PTFe-3, (c) PTFe-4, and (d) PTFe-5.

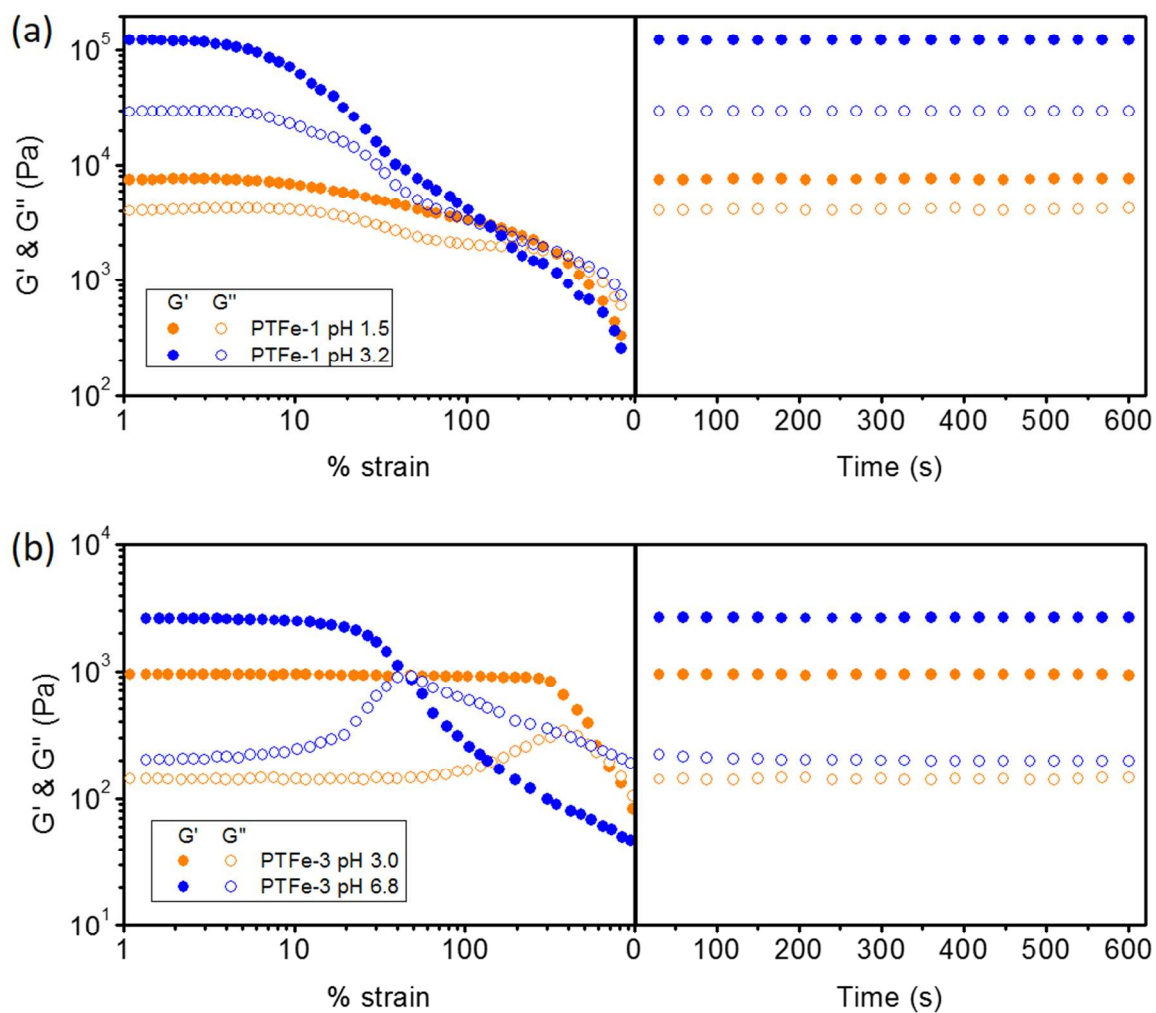


Figure S6. Quantitative tests of the self-healing properties of PTFe hydrogels. Strain was increased from 1% to 1000% at 1 Hz, and the recovery was monitored at 1% strain with 1 Hz. (a) PTFe-1, and (b) PTFe-3.

Table S1. The weight ratios of various components and the initial pH values in the fabrication of PDDA-TA bicomponent hydrogels

| sample name | weight ratio of PDDA:TA | initial solution pH |
|-------------|-------------------------|---------------------|
| PT-1 | 2 : 1 | 1.4 |
| PT-2 | 4 : 1 | 1.8 |
| PT-3 | 10 : 1 | 2.2 |
| PT-4 | 15 : 1 | 2.5 |
| PT-5 | 20 : 1 | 2.7 |

Table S2. The ionic conductivities of hydrogels

| Sample | pH | conductivity ($S\ m^{-1}$) | |
|--------------------|------------------|------------------------------|------------------|
| PTFe-2 | Initial solution | 1.3 | 4.56 ± 0.002 |
| | hydrogel | 2.0 | 4.30 ± 0.09 |
| | | 2.7 | 4.03 ± 0.14 |
| | | 3.2 | 4.32 ± 0.06 |
| | | 4.9 | 4.42 ± 0.02 |
| PTFe-4 | Initial solution | 1.9 | 4.25 ± 0.004 |
| | hydrogel | 3.6 | 4.19 ± 0.04 |
| | | 4.0 | 4.25 ± 0.02 |
| | | 6.4 | 4.28 ± 0.02 |
| | | 8.3 | 4.34 ± 0.02 |
| PDDA-TA- Fe_3O_4 | Initial solution | 2.9 | 3.55 ± 0.001 |
| | hydrogel | 4.7 | 3.68 ± 0.01 |
| | | 5.8 | 3.77 ± 0.03 |
| | | 7.4 | 3.90 ± 0.02 |