

Supporting Information

High Strength and Self-Healable Gelatin/Polyacrylamide Double Network Hydrogels

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Table S1. Effect of AAm concentration on the tensile properties of Gelatin/PAAm gels^a.

| AM (% w/v) | E (kPa) | σ_f (MPa) | ε_f (mm/mm) | W (MJ/m ³) |
|------------|---------|------------------|-------------------------|------------------------|
| 15 | 21.4 | 0.0623 | 13.30 | 0.347 |
| 20 | 41.6 | 0.1579 | 24.33 | 2.157 |
| 24 | 50.7 | 0.1789 | 24.85 | 2.540 |
| 30 | 56.2 | 0.2102 | 28.44 | 3.510 |
| 35 | 81.4 | 0.2001 | 30.65 | 4.581 |

a: gelatin= 50 mg/mL, MBA/AAm= 0.05 mol%, APS/AAm=0.1 mol%

Table S2. Effect of gelatin concentration on the tensile properties of Gelatin/PAAm gels^a.

| gelatin | E (kPa) | σ_f (MPa) | ε_f (mm/mm) | W (MJ/m ³) |
|---------|---------|------------------|-------------------------|------------------------|
| 0 | 22.9 | 0.0377 | 4.95 | 0.125 |
| 5 | 28.4 | 0.0395 | 5.19 | 0.133 |
| 10 | 29.1 | 0.0587 | 8.63 | 0.312 |
| 15 | 30.9 | 0.0968 | 14.33 | 0.743 |
| 30 | 32.5 | 0.1554 | 20.56 | 1.655 |
| 50 | 49 | 0.1834 | 21.97 | 2.225 |

a: AAm=24% w/v, MBA/AAm= 0.05 mol%, APS/AAm=0.14 mol%

Table S3. Effect of MBA concentration on the tensile properties of Gelatin/PAAm gels^a.

| MBA/AAm (mol%) | E (kPa) | σ_f (MPa) | ε_f (mm/mm) | W (MJ/m ³) |
|-------------------|---------|------------------|-------------------------|------------------------|
| 0 | 14.2 | 0.0902 | 35.38 | 1.228 |
| 0.01 | 22.4 | 0.1394 | 25.19 | 1.782 |
| 0.03 | 42.5 | 0.1906 | 27.15 | 2.730 |
| 0.05 | 35.2 | 0.1843 | 23.03 | 2.218 |
| 0.07 | 43.4 | 0.1950 | 16.23 | 1.562 |
| 0.1 | 43.1 | 0.0884 | 7.31 | 0.412 |

a: AAm=24% w/v, gelatin= 50 mg/mL, APS/AAm=0.14 mol%

Table S4. Effect of APS concentration on the tensile properties of Gelatin/PAAm gels^a.

| APS/AM (mol%) | E (kPa) | σ_f (MPa) | ε_f (mm/mm) | W (MJ/m ³) |
|---------------|---------|------------------|-------------------------|------------------------|
| 0.01 | 34.8 | 0.0663 | 7.26 | 0.326 |
| 0.03 | 44.6 | 0.1491 | 22.83 | 2.128 |
| 0.05 | 63.4 | 0.1971 | 26.44 | 3.016 |
| 0.10 | 50.7 | 0.1789 | 24.85 | 2.540 |
| 0.14 | 49.0 | 0.1846 | 22.47 | 2.225 |

a: AAm=24% w/v, MBA/AAm= 0.05 mol%, gelatin= 50 mg/mL

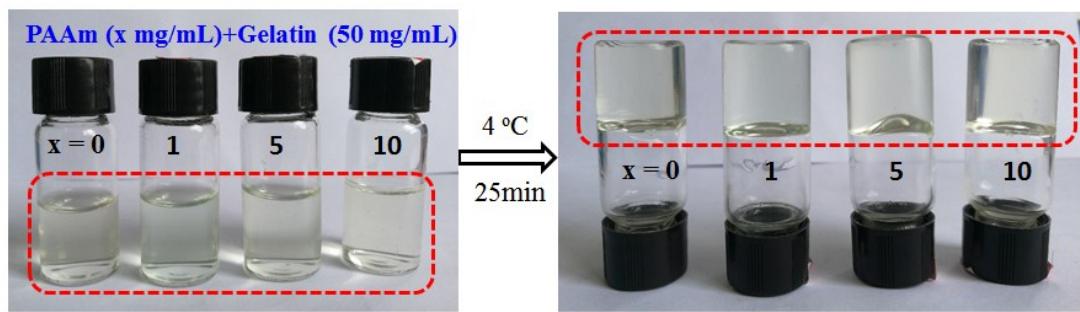


Figure S1. Gelation of gelatin (50 mg/mL) in various PAAm solutions from 60 °C.

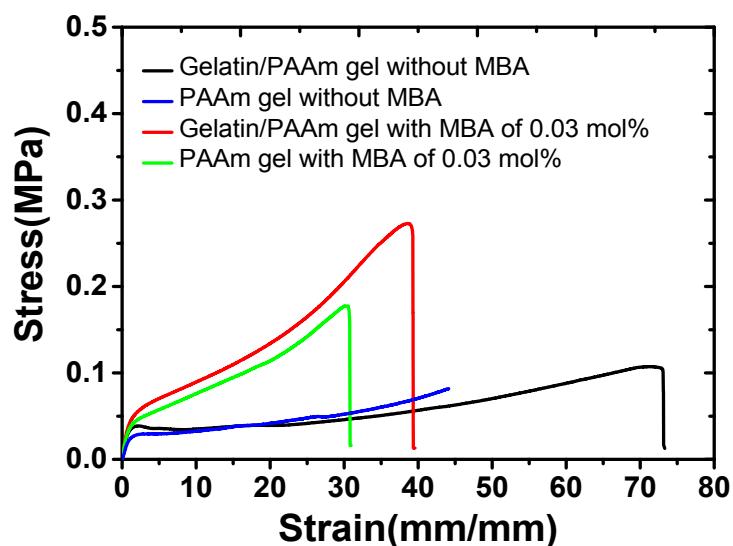


Figure S2. Stress-strain curve of PAAm gel and Gelatin/PAAm gel without MBA, and PAAm gel and Gelatin/PAAm gel with MBA of 0.03 mol%. gelatin of 50 mg/mL, AAm of 35 % (w/v) and APS/AAm of 0.05 mol%.

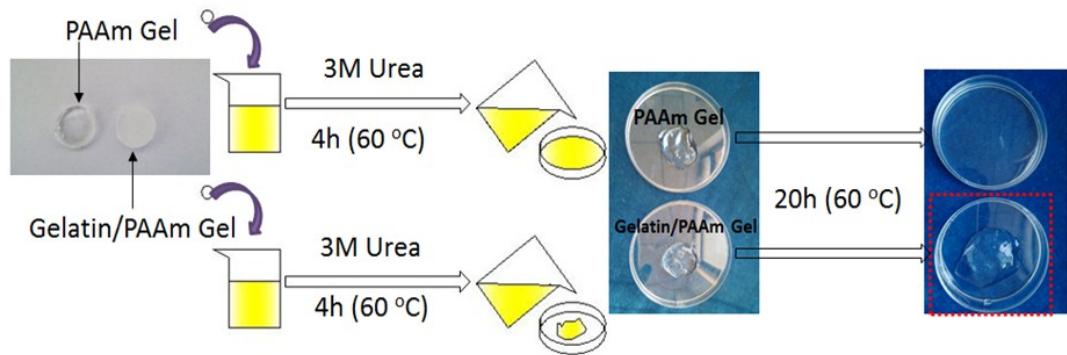


Figure S3. The swelling behavior of PAAm gel and Gelatin/PAAm gel (both of them without chemical crosslinkers) in the 3M urea solution at 60 °C with 4 h and 20 h.

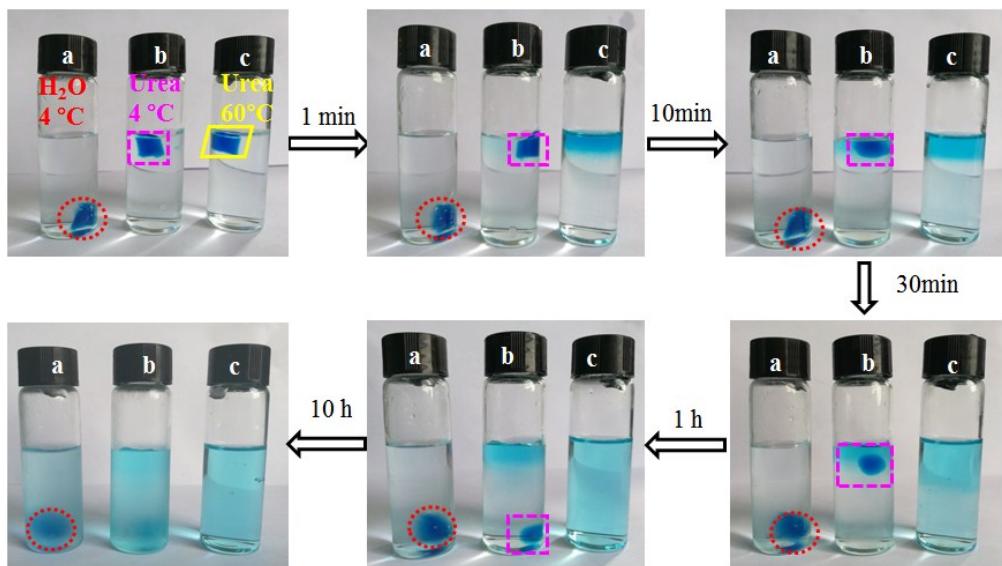


Figure S4. Stability of gelatin gel in various solutions. The concentration of gelatin was 50 mg/mL, and the gelatin gels were dyed by methylene blue. The urea solution was 3 mol/L.

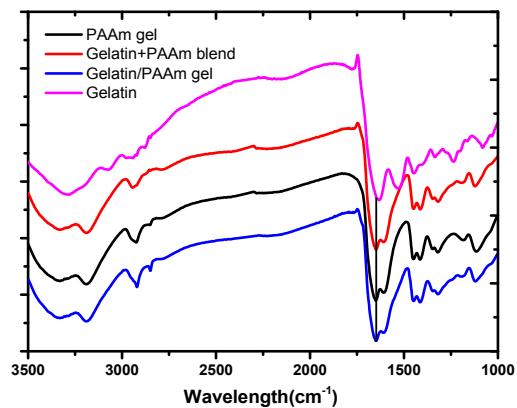


Figure S5. FTIR curves of PAAm gel, Gelatin+PAAm blend, Gelatin/PAAm gel and gelatin

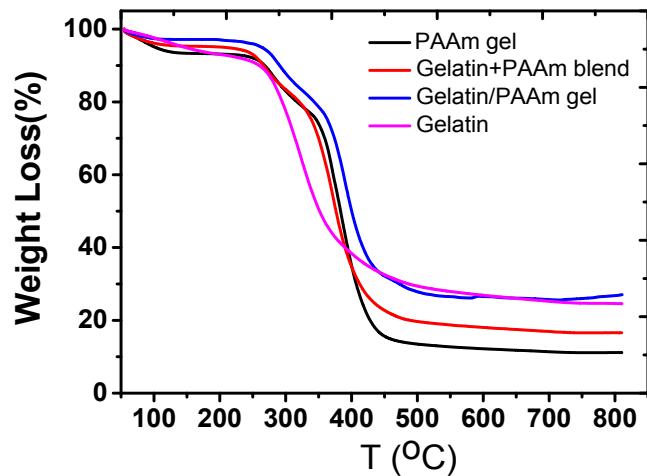


Figure S6. TGA curves of PAAm gel, Gelatin+PAAm blend, Gelatin/PAAm gel and gelatin