

Supporting information

Tough, Rapid-Recovery Composite Hydrogel Fabricated Using Synergic Core–Shell Microgel Covalent Bonding and Fe³⁺

Coordination Cross-Linking

Xuechen Liang,^{ab} Yukun Deng,^{ab} Xiaopeng Pei,^{ac} Kankan Zhai,^{ac} Kun Xu,^{a*} Ying Tan,^{a*} Xinyuan Gong,^a
and Pixin Wang^a

^a Key Laboratory of Polymer Ecomaterials, Changchun Institute of Applied Chemistry, Chinese Academy of Sciences, Changchun 130022,
Jilin, P. R. China.

^b University of Chinese Academy of Sciences, Beijing 100049, P. R. China.

^c University of Science and Technology of China, Hefei 230026, P. R. China.

E-mail: xukun@ciac.ac.cn; tanying@ciac.ac.cn.

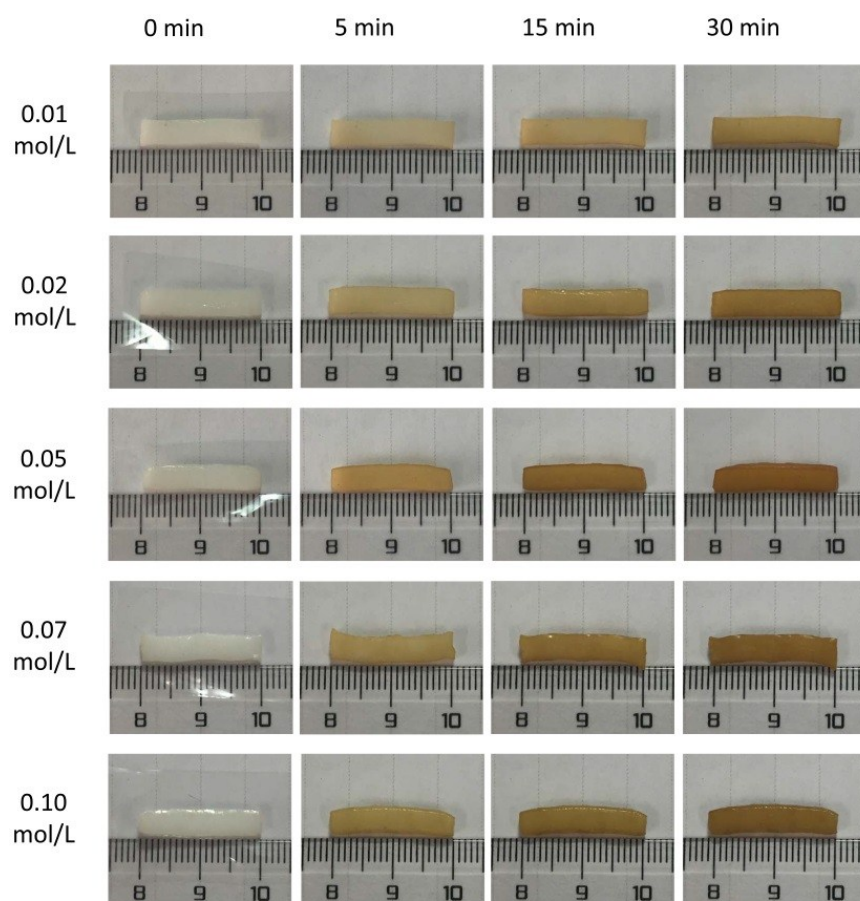


Fig. S1 The photographs of hydrogels immersed in various concentrations of FeCl_3 within 30 min. The size of each photograph is $2.5 \text{ cm} \times 3.0 \text{ cm}$.

Table S1 The polymer volume fractions of hydrogels immersed into 0.05 mol/L FeCl₃ solution.

<i>Soaking time</i> $C_{(\text{Fe}^{3+})}$	0 min	5 min	15 min	30 min	1 h	2 h	3 h	6 h	18 h
	v/v%	v/v%	v/v%	v/v%	v/v%	v/v%	v/v%	v/v%	v/v%
0.05 mol/L	25.32	25.08	24.67	23.83	21.08	20.33	19.57	19.51	19.45

The polymer volume fractions were calculated by V_{dry}/V , where V_{dry} was the volume of dry hydrogel, and V was the volume of hydrogel before dying. The volumes were measured by displacement method, and the solvent was n-hexane.

Table S2 The polymer volume fractions of hydrogels immersed into various concentrations of FeCl₃ solution within 30 min.

$C_{(\text{Fe}^{3+})}$	polymer volume fraction, v/v%
0.01 mol/L	24.07
0.02 mol/L	23.67
0.05 mol/L	23.83
0.07 mol/L	23.90
0.10 mol/L	23.81

Table S3 The polymer volume fractions of hydrogels with various AAm/AAC molar ratios and monomer contents. (0.05 mol/L FeCl₃, 30 min)

Name	polymer volume fraction, v/v%	Name	polymer volume fraction, v/v%
H-15-(93/7)	25.32	Fe ³⁺ -H-15-(93/7)	23.83
H-15-(86/14)	26.08	Fe ³⁺ -H-15-(86/14)	24.15
H-15-(79/21)	25.75	Fe ³⁺ -H-15-(79/21)	23.48
H-10-(93/7)	24.02	Fe ³⁺ -H-10-(93/7)	21.71
H-20-(93/7)	27.37	Fe ³⁺ -H-20-(93/7)	25.67
MBA (0.1 mol%)	25.03	Fe ³⁺ -MBA (0.1 mol%)	22.05

Table S4 The polymer volume fractions of hydrogels immersed into EDTA-2Na/urea solution.

Solvent	polymer volume fraction, v/v%
H ₂ O	16.05
0.01 M EDTA-2Na	16.64
0.02 M EDTA-2Na	16.29
0.05 M EDTA-2Na	15.84
0.5 M CO(NH ₂) ₂	16.88

The swelling ratios of hydrogel samples were fixed at 1.5 times.

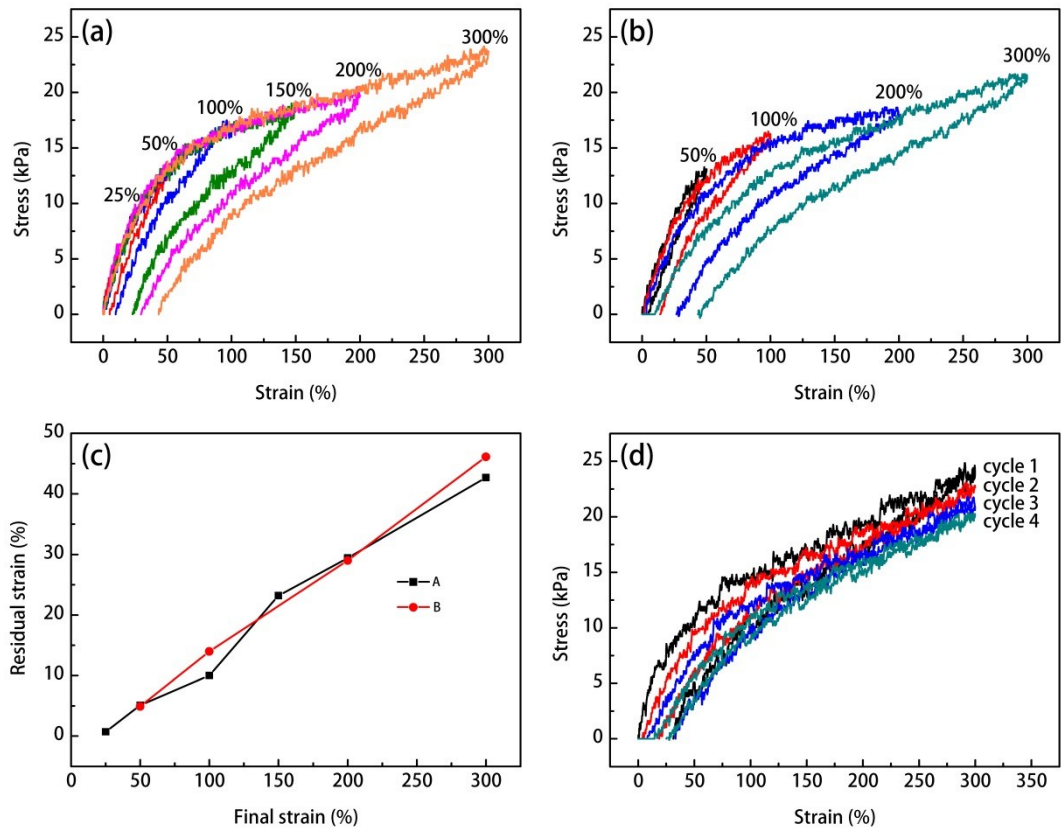


Fig. S2 Cyclic loading–unloading of H-15-(93/7) hydrogel under various conditions: (a) tensile loading–unloading under different strains (25%, 50%, 100%, 150%, 200%, 300%), (b) continuous cyclic tensile tests under incremental strain (50%, 100%, 200%, 300%), (c) the residual strain vs. final strain of Fig. S2a (A) and of Fig. S2b (B), (d) four successive cyclic tensile tests under a strain of 300%.