Electronic Supplementary Information

Dually cross-linked single network poly(acrylic acid) hydrogels with superior mechanical properties

Ming Zhong[a], Yi-Tao Liu[a], Xiao-Ying Liu[a], Fu-Kuan Shi[a], Li-Qin Zhang[a], Meifang Zhu[b] and Xu-Ming Xie[a]*

[a]Key Laboratory of Advanced Materials (MOE), Department of Chemical Engineering, Tsinghua University, Beijing 100084, China.

[b]State Key Laboratory for Modification of Chemical Fibers and Polymer Materials, College of Materials Science and Engineering, Donghua University, Shanghai 201620, China.

*E-mail: xxm-dce@mail.tsinghua.edu.cn

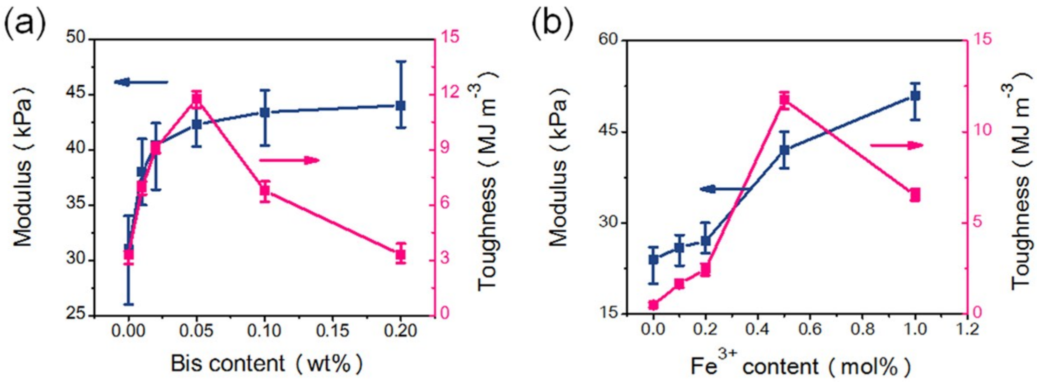

Fig. S1 Influence of (a) Bis, and (b) Fe^{3+} contents on the initial modulus and fracture toughness of the *d*-PAA gels.

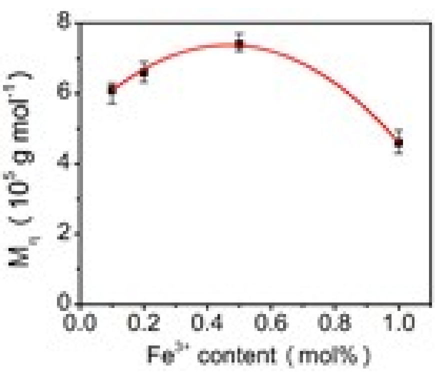

Fig. S2 Influence of Fe^{3+} content on the macromolecular weight of the *i*-PAA gels.

First, the *i*-PAA gels (free of Bis) were added to a certain amount of chelating agent solution, ethylene diamine tetraacetic acid (EDTA), and stirred at room temperature till the *i*-PAA gels were dissolved completely. After that the solution was dialyzed for 1 week in order to remove the Fe^{3+} ions and chelating agent. Linear PAA was separated from the solution by neutralization with NaOH and then by precipitation with methanol. The viscosity measurements were performed by a standard Ubbelhode viscometer, in 0.1 mol L^{-1} NaBr solution, at 25 ^{o}C and pH = 7. The viscous average molecular weight (M_η) of PAA from the *i*-PAA gels was measured and calculated, according to the Mark-Houwink-Sakurada equation:

$$[\eta] = kM_\eta^{\alpha}$$

We used Francois relation[1]: $K = 3.12 \times 10^{-5} L\, g^{-1}$, $\alpha = 0.755$.

$$[\eta] = 3.12 \times 10^{-5}\, M_\eta^{0.755}\ (g\ mL^{-1}).$$

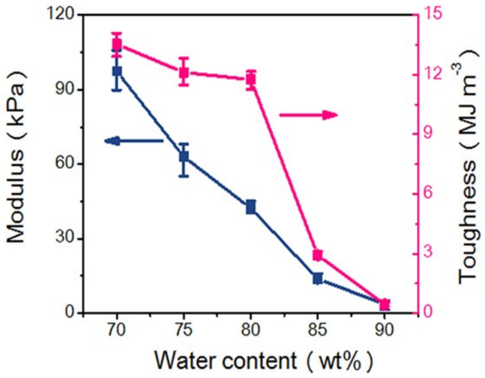

Fig. S3 Influence of water content on the modulus and toughness of the *d*-PAA gels.

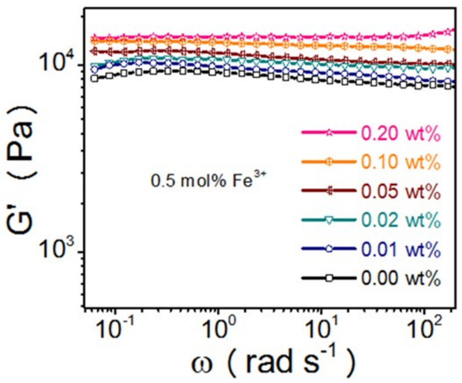

Fig. S4 Influence of Bis content on the storage modulus of the *d*-PAA gels

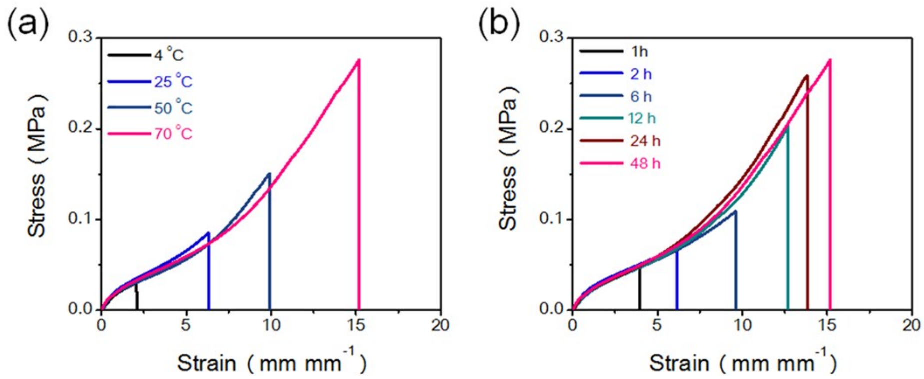

Fig. S5 Typical stress–strain curves of the self-healed *d*-PAA gels (Fe^{3+} = 0.5 mol%, Bis = 0.05 wt%, H_2O = 80 wt%) with (a) temperature and (b) time as a function.

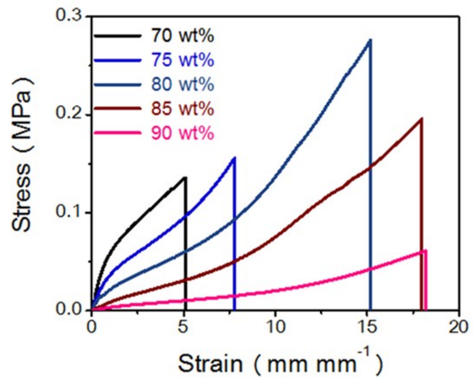

Fig. S6 Typical stress–strain curves of the self-healed *d*-PAA gels (Fe^{3+} = 0.5 mol%, Bis = 0.05 wt%) with different water contents.

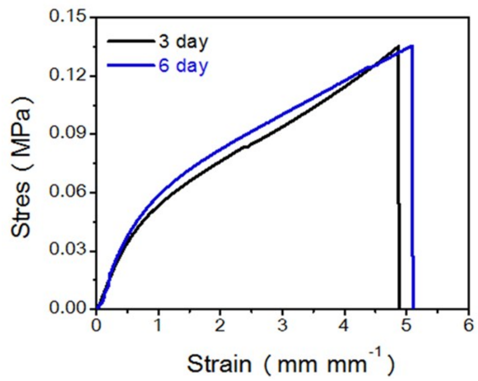

Fig. S7 Typical stress–strain curves of the self-healed *d*-PAA gels (Fe^{3+} = 0.5 mol%, Bis = 0.05 wt%, H_2O = 70 wt%) with time as a function.

Notes and references

S1. J. Brandrup, E. H. Immergut, E. A. Grulke, *Polymer Handbook*. John Wiley & Sons, Inc.: New York, 1999.