Electronic Supplementary Information

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

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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 °C and pH = 7. The viscous average molecular weight ($M_n$) of PAA from the $i$-PAA gels was measured and calculated, according to the Mark-Houwink-Sakurada equation:

$$[\eta] = k M_n^\alpha$$

We used Francois relation$^1$: $K = 3.12 \times 10^{-5} L g^{-1}$, $\alpha = 0.755$, $[\eta] = 3.12 \times 10^{-5} M_n^{0.755}$ (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 ($\text{Fe}^{3+} = 0.5$ mol%, Bis = 0.05 wt%, $\text{H}_2\text{O} = 80$ 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$_2$O = 70 wt%) with time as a function.

**Notes and references**