Supporting Information

Multi-responsive, self-healing and adhesive PVA based hydrogels induced by the ultrafast complexation of Fe^{3+} ions

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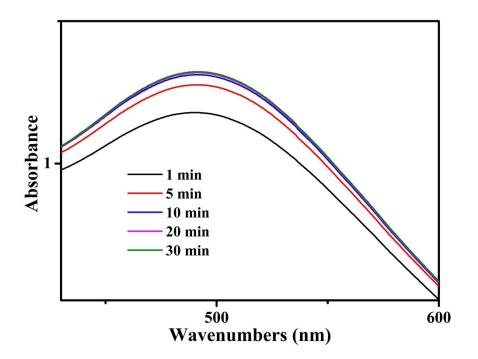


Fig. S1 The time-dependent UV-Vis study of the PVAA-Fe hydrogel. The spectra of PVAA-Fe recorded 1 min after mixing with Fe^{3+} were identical to that after 30 min, which can be ascribed to the extremely fast complexation between the Fe^{3+} ions and the PVAA polymer chain.

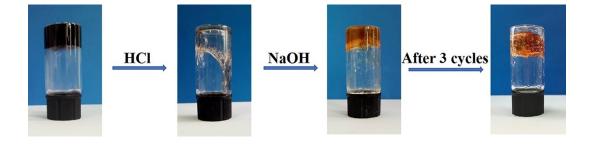


Fig. S2 Photographs of the reversible gel-sol and sol-gel transitions triggered by HCl and NaOH solution. The hydrogel was exposed to 30 μ L of a 4 M aqueous HCl solution, followed by treatment with 30 μ L of a 4 M aqueous NaOH solution.

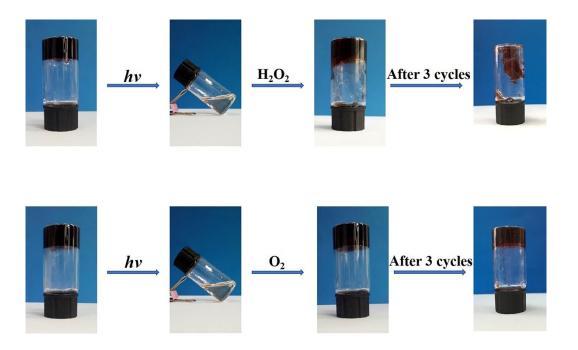


Fig. S3 The reversible gel-sol transitions triggered by UV light. After ultraviolet light irradiation, the gel was converted into colorless sol. Then, this PVAA solution can be recovered to the homogeneous brown gel again after being exposed to oxygen for three days or being added excessive hydrogen peroxide.

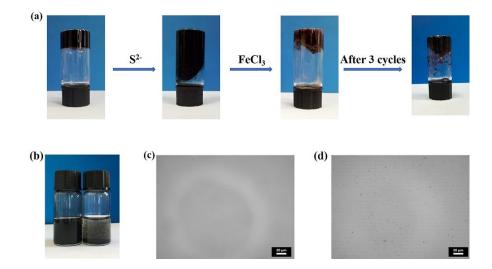


Fig. S4 (a)The reversible gel-sol and sol-gel transition induced by Na₂S. 60 μL of Na₂S solution (100 mg/mL) was added into 3 g of PVAA-Fe hydrogels, and a dark brown solution was obtained. Gel state could be restored by adding excessive Fe³⁺ ions. (b) The photo of PVAA-Fe-S²⁻ sol (left) and Fe-S²⁻ solution (right). (c) The image of optical microscope of PVAA-Fe hydrogel. (d) The

image of optical microscope of PVAA-Fe-S²⁻ sol.

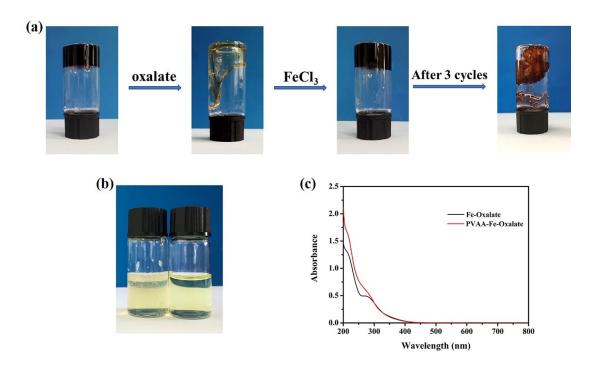


Fig. S5 (a)The reversible gel-sol transition induced by oxalate. 90 μ L of oxalate solution (100 mg/mL) was added into 3g of PVAA-Fe hydrogels, and a light-yellow solution was obtained. Gel state could be restored by adding excessive Fe³⁺ ions. (b) The photo of PVAA-Fe-Oxalate sol (left) and Fe-Oxalate solution (right). (c) The UV/Vis absorption spectra of the PVAA-Fe-Oxalate sol and Fe-Oxalate solution.

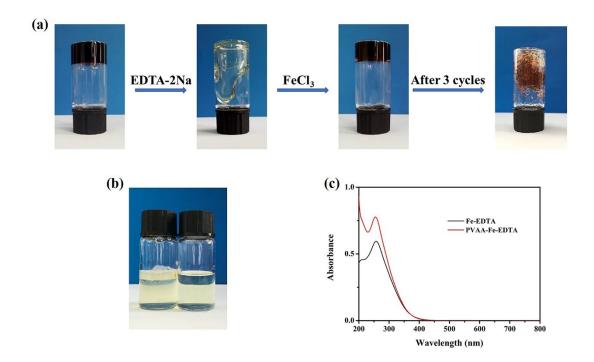


Fig. S6 (a) The reversible gel-sol transition induced by EDTA. $30\mu L$ of EDTA-2Na solution (100 mg/mL) was added into 3 g of PVAA-Fe hydrogels, and a light-yellow solution was obtained. Gel state could be restored by adding excessive Fe³⁺ ions. (b) The photo of PVAA-Fe-EDTA sol (left) and Fe-EDTA solution (right). (c) The UV/Vis absorption spectra of the PVAA-Fe-EDTA sol and Fe-EDTA solution.

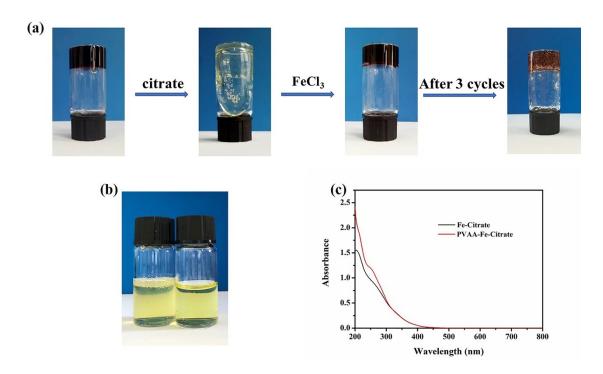


Fig. S7 (a) The reversible gel-sol transition induced by. 30 μ L of citric acid radical solution (100 mg/mL) was added into 3 g of PVAA-Fe hydrogels, and a light-yellow solution was obtained. Gel state could be restored by adding excessive Fe³⁺ ions. (b) The photo of PVAA-Fe-Citrate sol (left) and Fe-Citrate solution (right). (c) The UV/Vis absorption spectra of the PVAA-Fe-Citrate sol and Fe-Citrate solution.

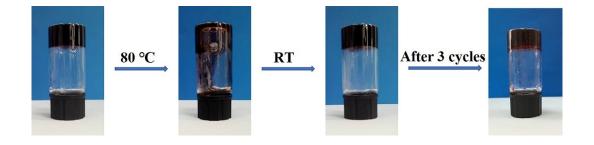


Fig. S8 The reversible gel-sol transitions triggered by raising the temperature to 80 °C. Then, the PVAA solution can be recovered to the homogeneous brown gel again after being placed at room temperature.

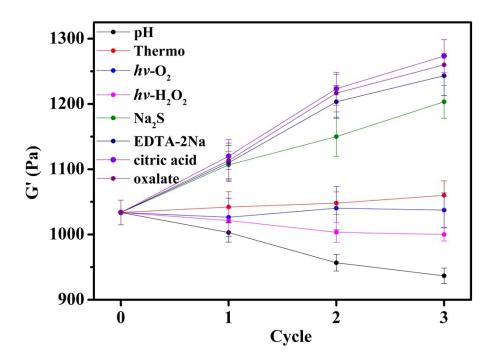


Fig. S9 Rheology analyses of the storage modulus G' on time sweep (25 $^{\circ}$ C, strain = 1%, frequency = 1 Hz) for different responsive process.