Supplementary Information

Thermo- and redox-responsive dendronized polymer hydrogels

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Scheme S1 Synthesis of **MTpy**. Reagent and conditions: Reagents and conditions: a) KOH, dry DMSO, 60 °C, 12 h, 88%; b) DMAP, TEA, dry DCM, 0 to 25 °C, 12 h, 90%.





Fig. S1 ¹H NMR spectra of copolymers: A) $P(G1_{0.86}Tpy_{0.14})$, B) $P(G1_{0.92}Tpy_{0.08})$, C) $P(G1_{0.96}Tpy_{0.04})$ in CDCl₃, D) $P(G2_{0.75}Tpy_{0.25})$ in DMSO- d_6 and E) $P(G0_{0.97}Tpy_{0.03})$ in

 $CDCI_3$.



Fig. S2 UV-vis spectra of $P(G1_{0.92}Tpy_{0.08})$ and $P(G1_{0.92}Tpy_{0.08})$ by addition of Fe²⁺ with [Tpy]/[Fe²⁺] = 2: 1 in aqueous solutions, after addition of H₂O₂, further addition of excess sodium ascorbate, and $P(G1_{0.92}Tpy_{0.08})$ in the presence of Fe³⁺ with [Tpy]/[Fe³⁺] = 2: 1 in aqueous solutions.



Fig. S3 Photographs of (a) $P(G1_{0.92}Tpy_{0.08})$ with addition of Fe^{2+} in different concentration aqueous solutions, and (b) $PG2_{0.75}Tpy_{0.25}$ with addition of Fe^{2+} .



Fig. S4 UV-vis spectra of **2** and **2** in the presence of Fe^{2+} with [**2**]/[Fe²⁺] = 2: 1 in aqueous solutions, addition of H₂O₂, further addition of excess amount of sodium ascorbate, and **2** in the presence of Fe³⁺ with [**2**]/[Fe³⁺] = 2: 1 in aqueous solution.



Fig. S5 UV-vis spectra of **2** with Fe²⁺ and Fe³⁺ for Job's plots. The MLCT bands from Fe³⁺-**2** complex are slight more complicated. There are two MLCT band around 493 nm and 555 nm, respectively. Only the intensities of MLCT band at 493 nm changes the order, therefore, Job's plots were based on the intensities of this MLCT band.



Fig. S6 Job's plots from complexation of 2 with Fe^{2+} (a) and Fe^{3+} (b).



Fig. S7 Change of lengths and widths of hydrogels with temperature from P(G1_{0.92}Tpy_{0.08}).



Fig. S8 Normalized refractive index traces from gel permeation chromatography of copolymers.





Fig. S10 ¹H NMR spectrum of compound MTpy in CDCI_{3.}