

Supplementary Materials

Coupling active polymer networks with solubility-driven gradients for augmented hydrogel thermocells

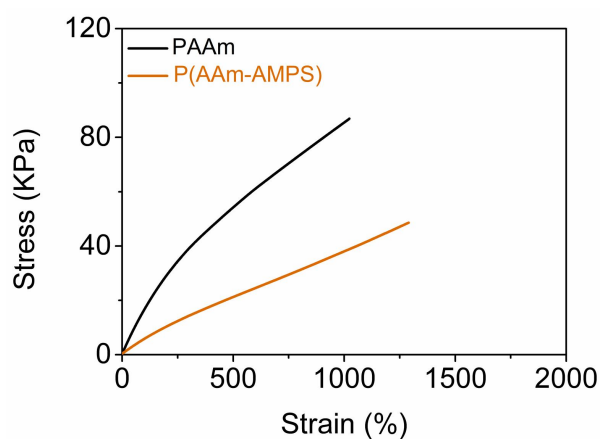
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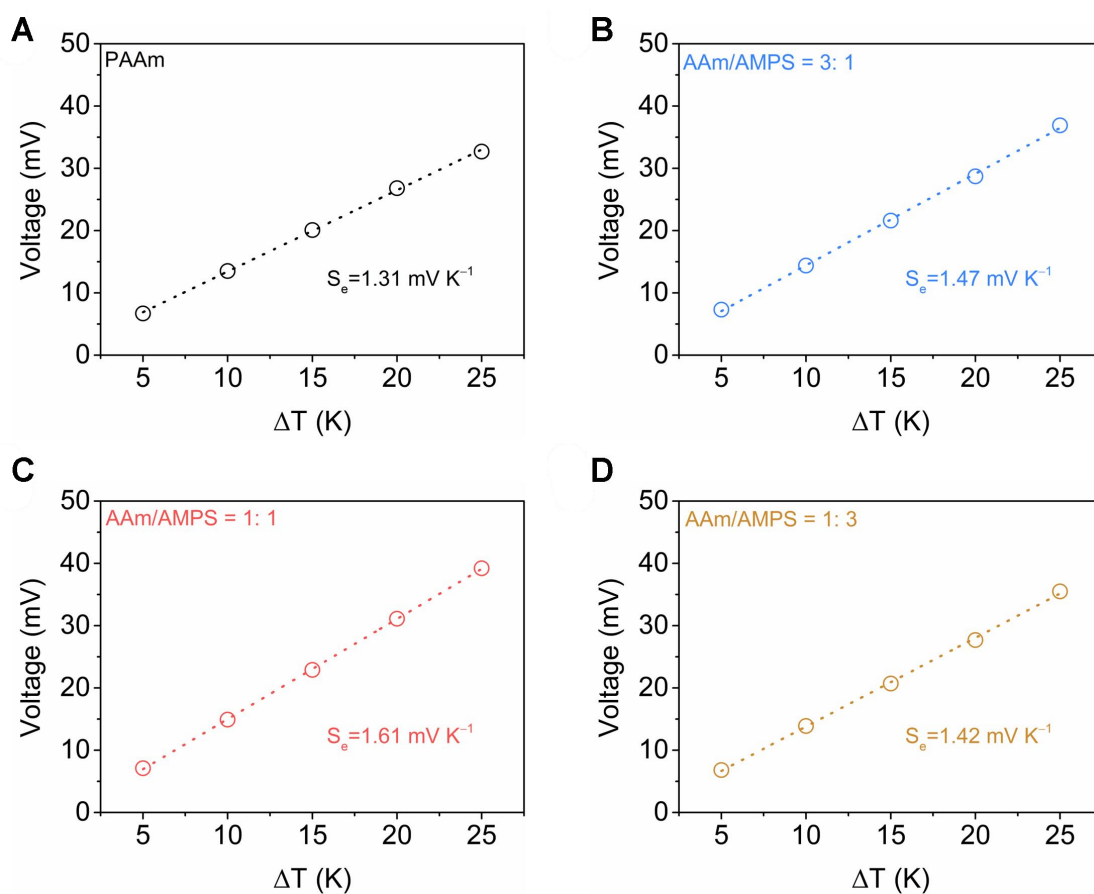
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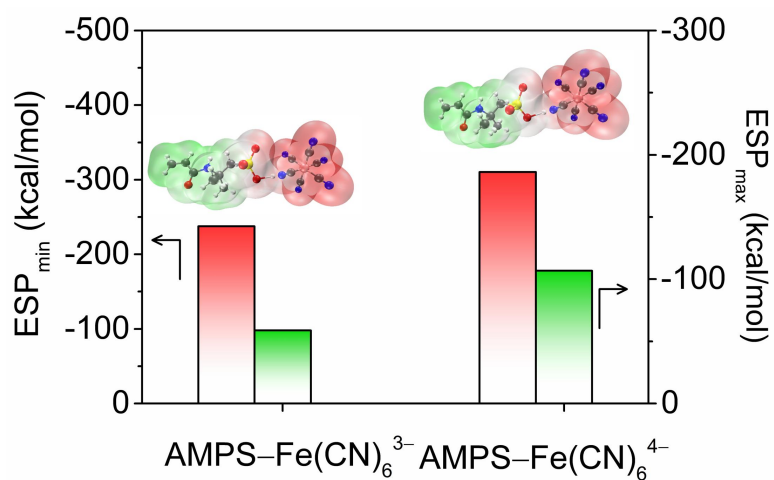
Supplementary Figures



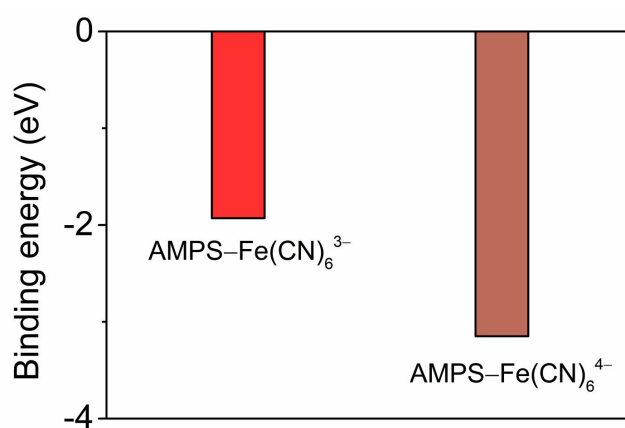
Supplementary Figure 1. Tensile stress-strain curves of PAAm and P(AAm-AMPS) (1:1 mass ratio) hydrogel electrolytes at room temperature.



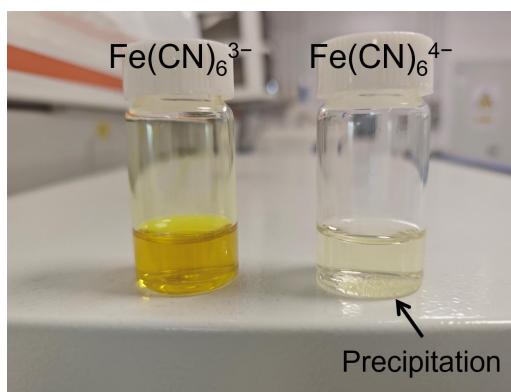
Supplementary Figure 2. Fitted steady-state voltage– ΔT plots for (A) PAAm electrolyte, (B) P(AAm-AMPS) electrolyte with a 3:1 mass ratio, (C) P(AAm-AMPS) electrolyte with a 1:1 mass ratio, and (D) P(AAm-AMPS) electrolyte with a 1:3 mass ratio.



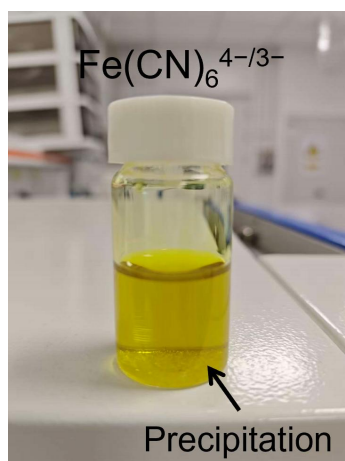
Supplementary Figure 3. Electrostatic potential maps of AMPS-Fe(CN)₆³⁻ and AMPS-Fe(CN)₆⁴⁻, with the corresponding ESP minimum and maximum values marked.



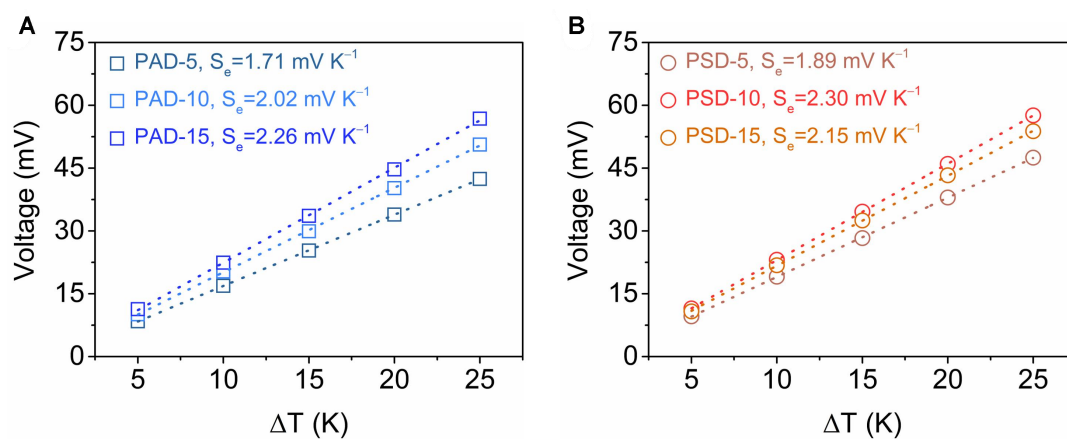
Supplementary Figure 4. Binding energies of AMPS-Fe(CN)₆³⁻ and AMPS-Fe(CN)₆⁴⁻.



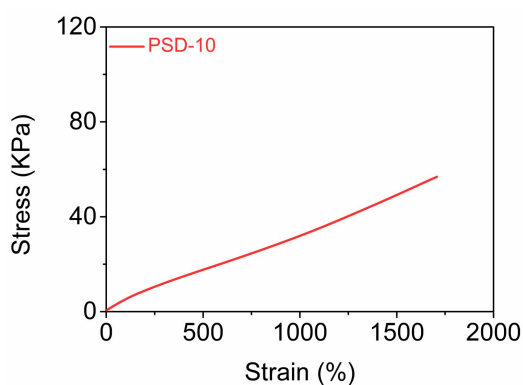
Supplementary Figure 5. Photographs of the 0.4 M Fe(CN)₆³⁻ and 0.4 M Fe(CN)₆⁴⁻ aqueous solutions incorporated with DMSO.



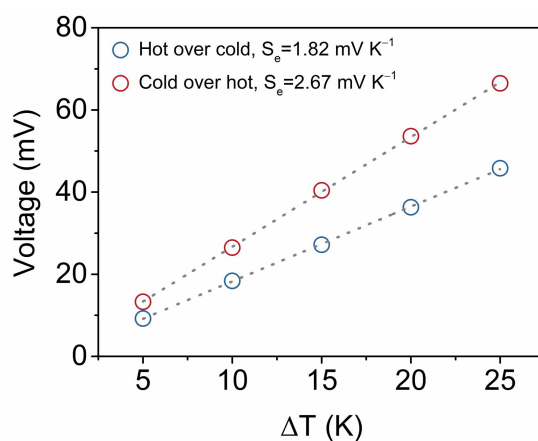
Supplementary Figure 6. Photographs of the 0.4 M $\text{Fe(CN)}_6^{4-/3-}$ aqueous solutions incorporated with DMSO.



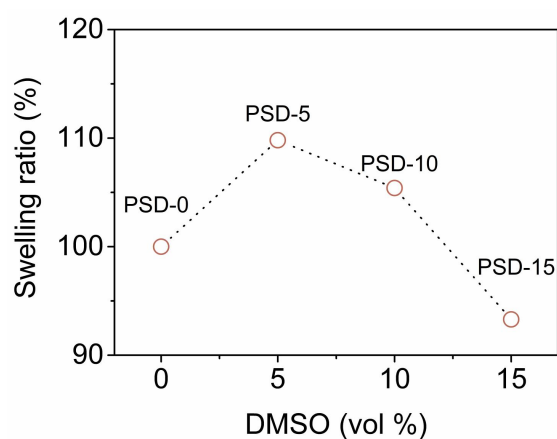
Supplementary Figure 7. Fitted steady-state voltage– ΔT plots for (A) PAD-X series (PAAm-based) and (B) PSD-X series (P(AAm-AMPS)-based), where X denotes the volume percentage of DMSO.



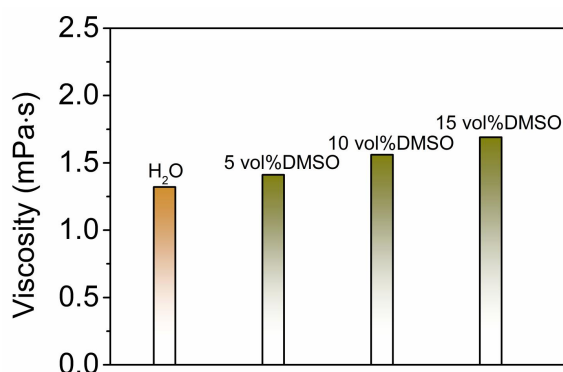
Supplementary Figure 8. Tensile stress-strain curves of PSD-10 hydrogel electrolyte at room temperature.



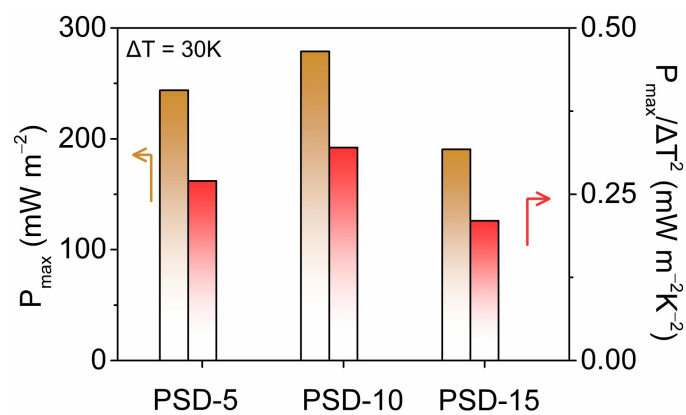
Supplementary Figure 9. Fitted steady-state voltage– ΔT plots for PSD-10 electrolyte at different heat orientations.



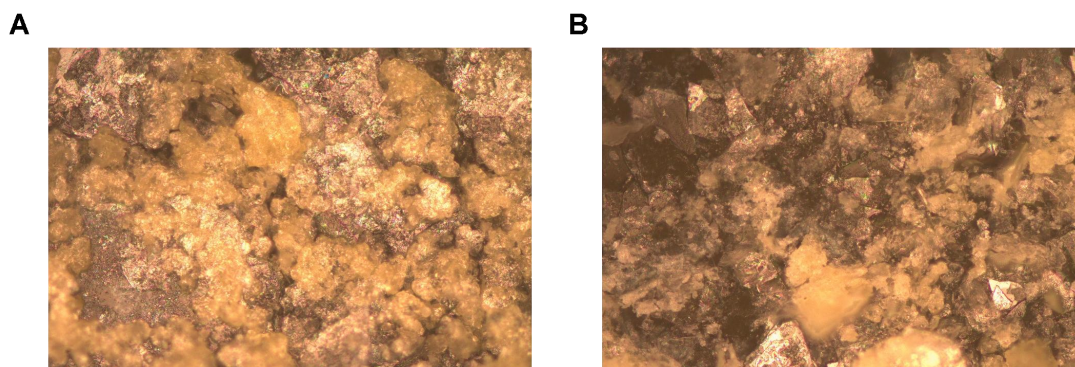
Supplementary Figure 10. Swelling ratio of PSD-X electrolyte as a function of DMSO volume percentage. The swelling ratio at 0 vol% DMSO is normalized to 100%.



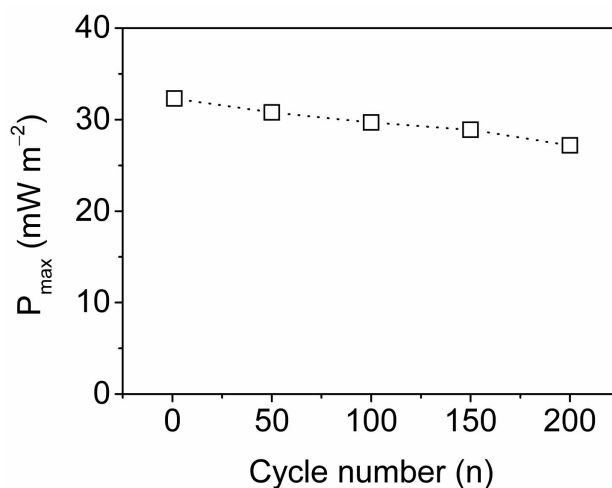
Supplementary Figure 11. Viscosity test results of H₂O/Fe(CN)₆^{4-/3-} and H₂O/DMSO (5, 10, 15 vol%)/Fe(CN)₆^{4-/3-} solutions at room temperature.



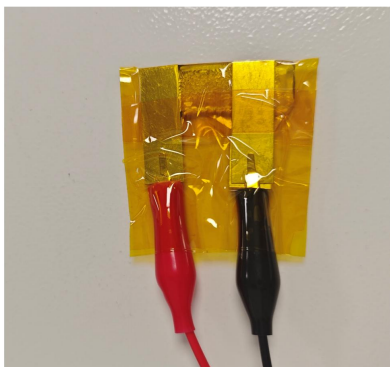
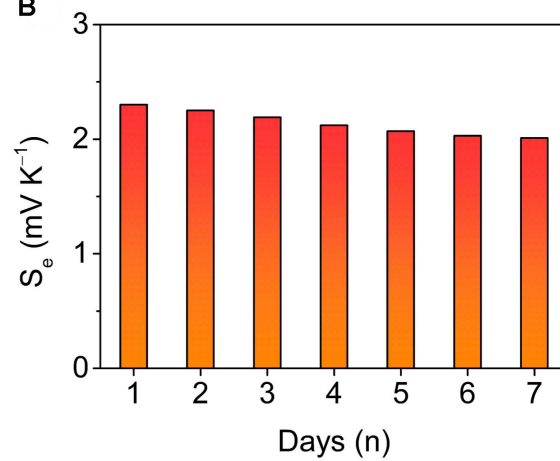
Supplementary Figure 12. Maximum power density ($P_{\max}$) and the normalized power density ($P_{\max}/\Delta T^2$) of PSD-5, PSD-10, and PSD-15 under a $\Delta T=30$ K.



Supplementary Figure 13. Optical microscopy images of PSD-10 (A) at room temperature and (B) after heating at 60 °C for 10 min.



Supplementary Figure 14. Variation of $P_{\max}$ during the quasi-continuous discharge process over 200 cycles.

A**B**

Supplementary Figure 15. (A) Optical image of the encapsulated QTEC (B) Stability of S_e for PSD-10 over 7 days.

Supplementary Table 1. Comparison of the S_e and $P_{\max}/\Delta T^2$ for the PSD-10 electrolyte developed in this work with those hydrogel electrolytes reported in recent literature

Electrolyte	S_e (mV K ⁻¹)	$P_{\max}/\Delta T^2$ (mW m ⁻² K ⁻²)	Ref.
Gelatin/KI/I ₂	0.75	0.0055	[1]
PVA/FeCl ₂ /3	0.78	0.171	[2]
3FT-PVA/K _{3/4} Fe(CN) ₆	1.50	0.22	[3]
PAAm/Fe(ClO ₄) _{2/3}	1.65	0.43	[4]
PVA/EG/choline chloride/ K _{3/4} Fe(CN) ₆	1.68	0.13	[5]
p(SBMA-MMA)/Fe(ClO ₄) _{2/3}	1.70	0.44	[6]
PNAGA-F68/K _{3/4} Fe(CN) ₆	1.76	0.23	[7]
PVA/starch/PDA-HNTs/LiCl/ K _{3/4} Fe(CN) ₆	1.82	0.1825	[8]
PAAm/EG/MXene/K _{3/4} Fe(CN) ₆	2.02	0.16	[9]
P(SBMA)/PEG/K _{3/4} Fe(CN) ₆	2.04	0.196	[10]
Bacterial cellulose/propylene glycol/K _{3/4} Fe(CN) ₆	2.30	0.07	[11]
PSD-10	2.30	0.32	This work

PVA: Polyvinyl alcohol.

3-FT PVA: PVA after three cycles of unidirectional freezing training.

EG: Ethylene glycol.

p(SBMA-MMA): Sulfobetaine methacrylate/methyl methacrylate after polymerization. PNAGA-F68: Poly (N-acryloyl glycinamide)/diacrylate capped Pluronic F68.

HNTs: Halloysite nanotubes.

PEG: Polyethylene glycol.

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