Supporting Information

Hydrogel thermocells with enhanced thermopower induced by thermosensitivity

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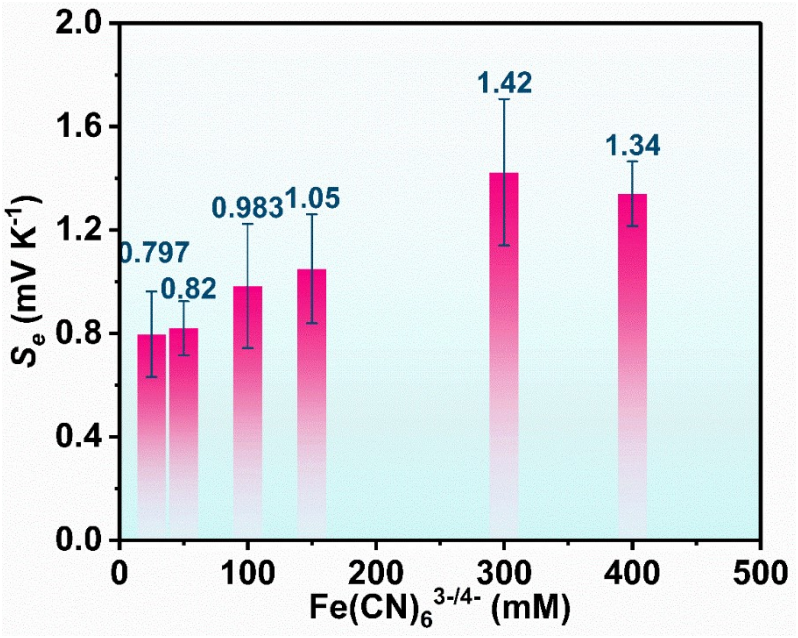

Fig. S1 The S_e value of $N_0A_{10}F_{0.3}$ doped with various concentrations of $Fe(CN)_6^{3-/4-}$

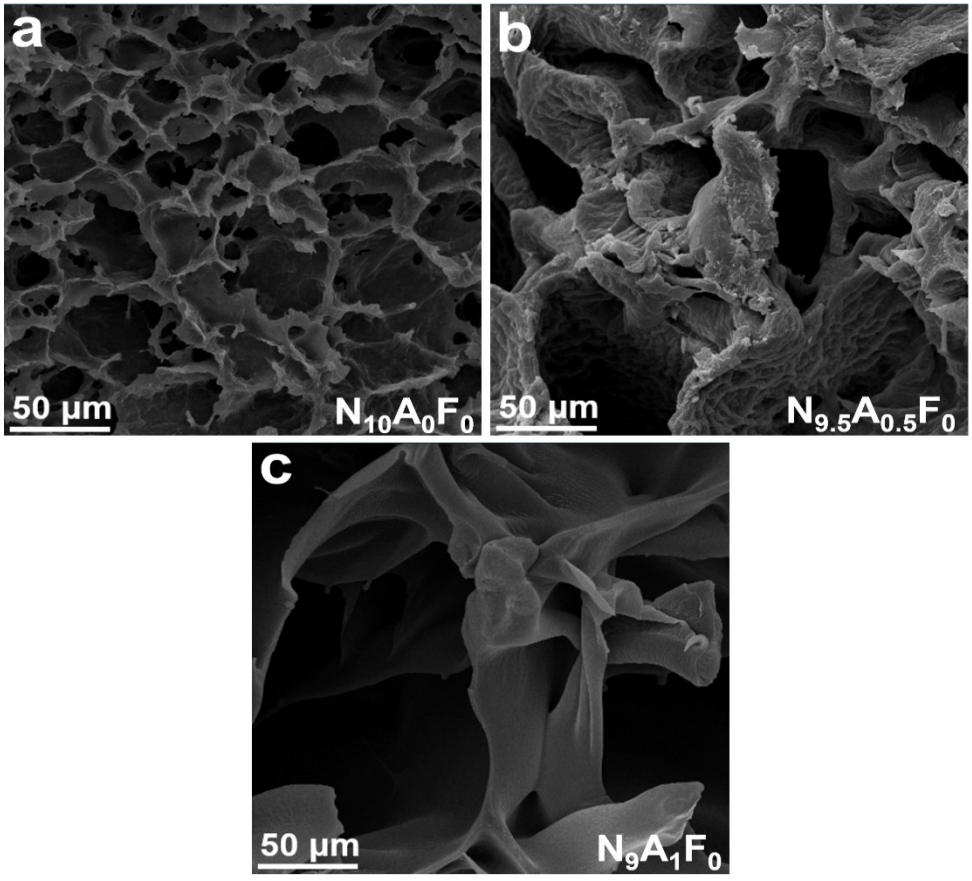

Fig. S2 SEM images of p(NIPAAm-co-Am) samples. (a) $N_{10}A_0F_0$. (b) $N_{9.5}A_{0.5}F_0$. (c) $N_9A_1F_0$

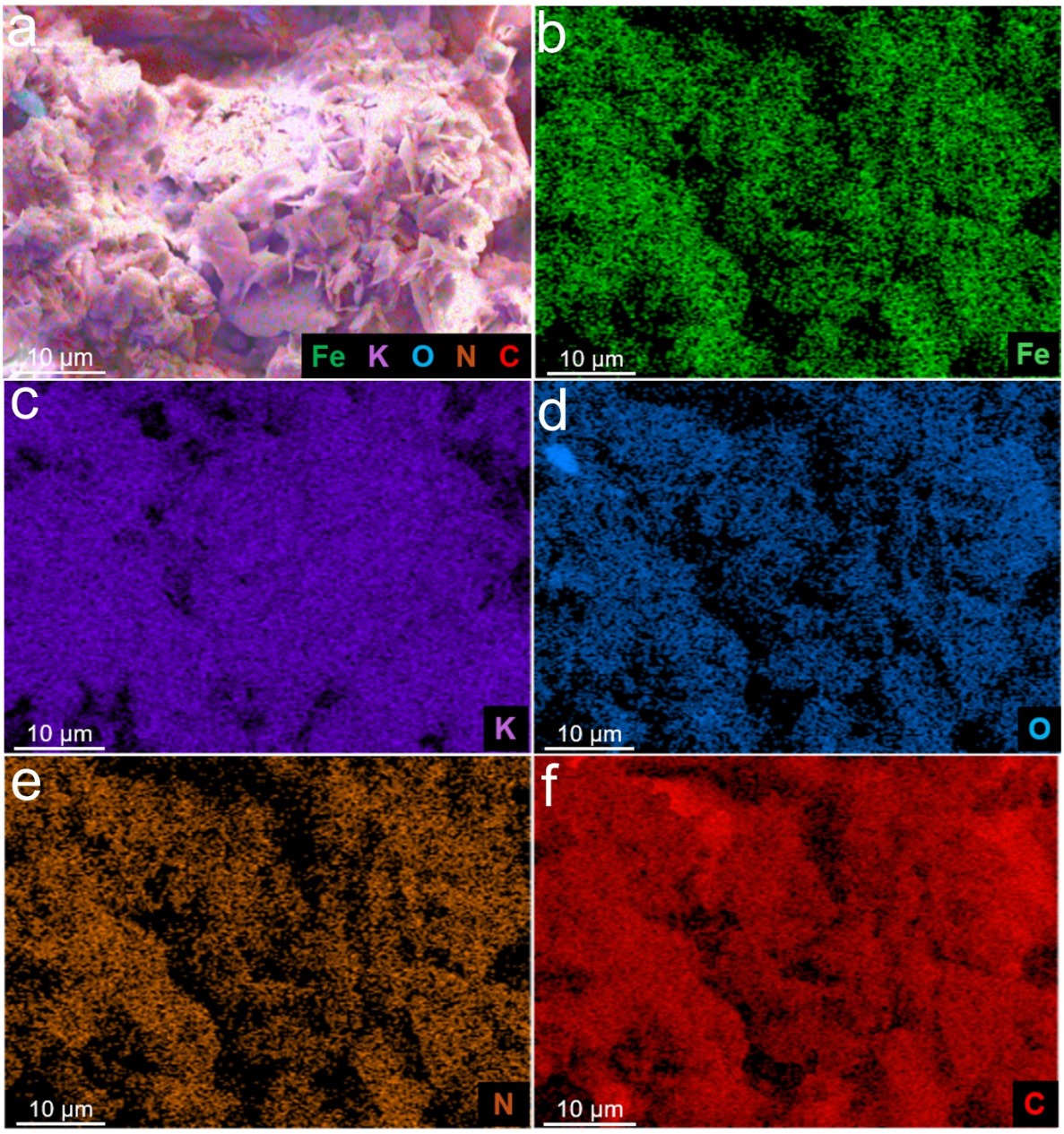

Fig. S3 (a) EDS mapping of p(NIPAAm-co-Am). (b) Fe, (c) K, (d) O, (e) N, and (f) C element distribution

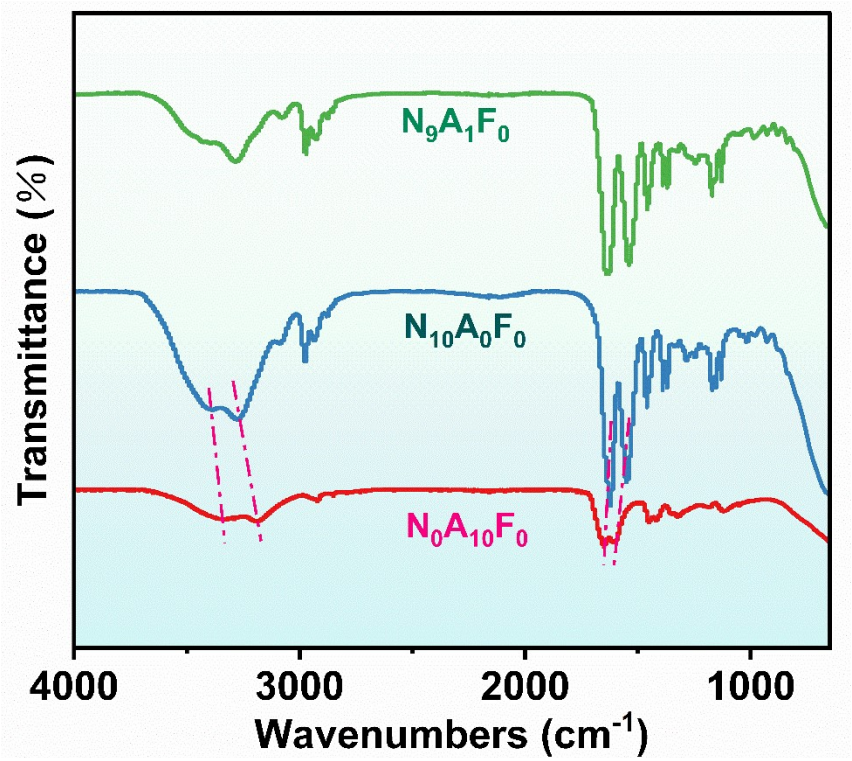

Fig. S4 FTIR spectra of $N_9A_1F_0$, $N_{10}A_0F_0$ and $N_0A_{10}F_0$

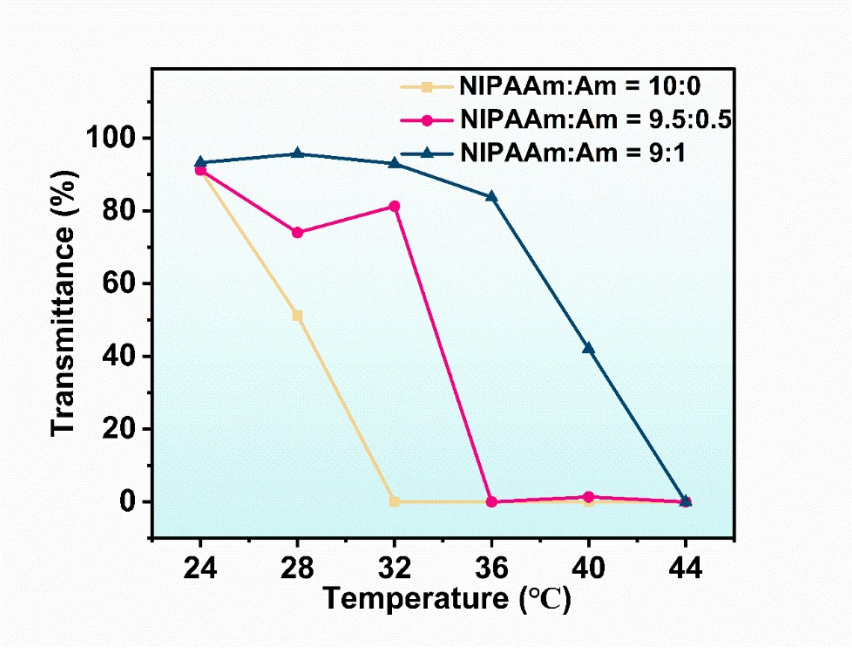

Fig. S5 Temperature- and composition-dependent transmittance of p(NIPAAm-co-Am) hydrogels at 700 nm

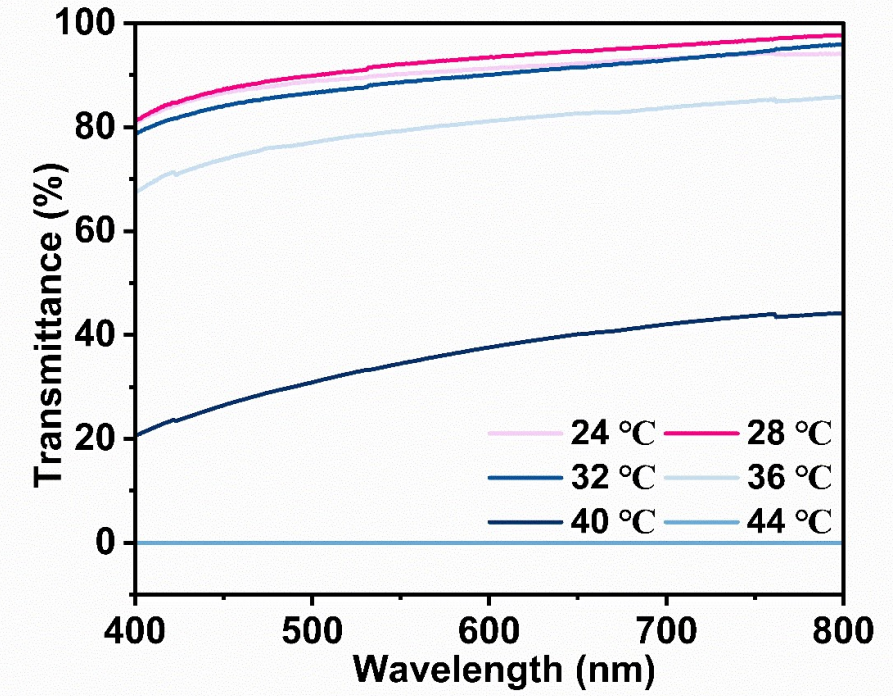

Fig. S6 Temperature dependent transmittance of of $N_9A_1F_{0.3}$

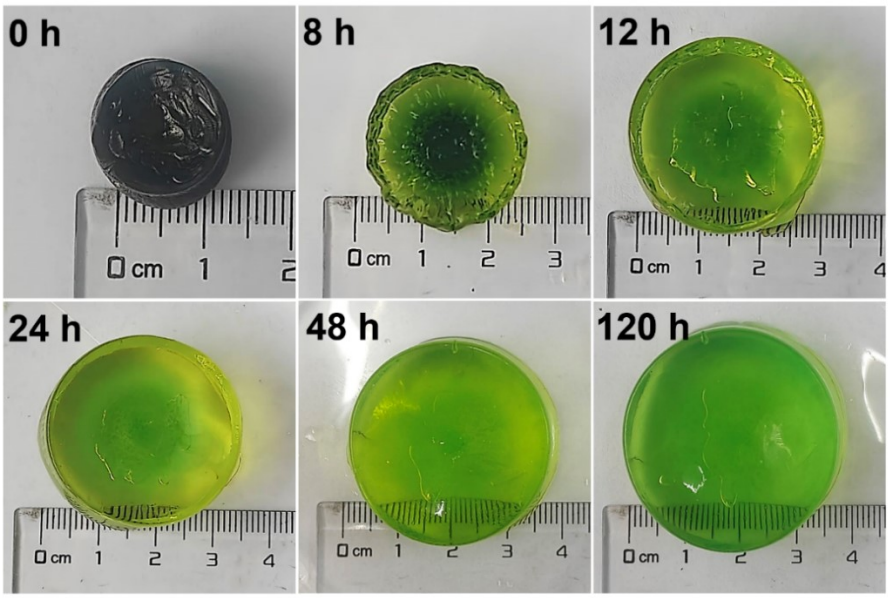

Fig. S7 Optical images of $N_0A_{10}F_{0.3}$ in deionized water at room temperature for different soaking times

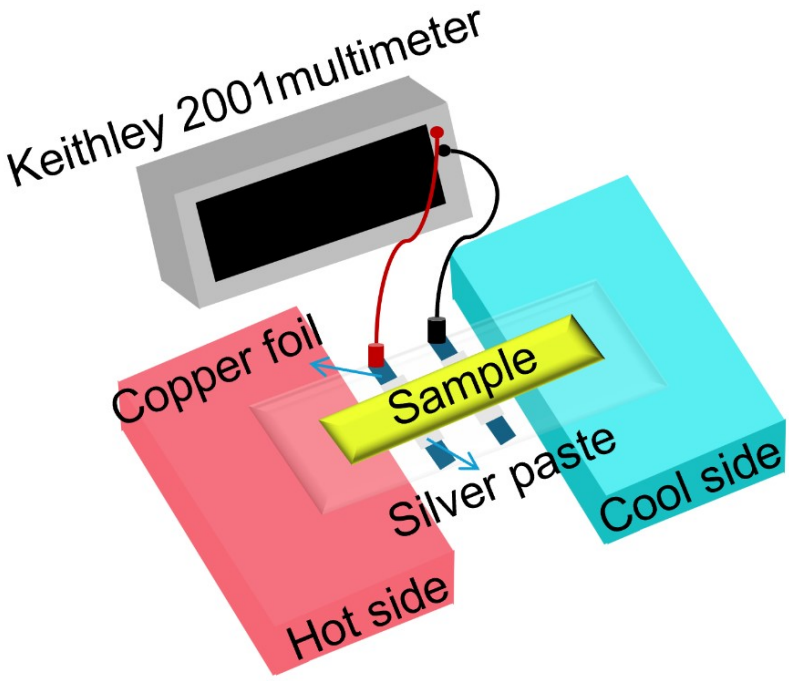

Fig. S8 Self-made ionic Seebeck coefficient measurement setup: The red and blue parts represent the heat source. The temperature and voltage are recorded by K-type thermocouple and voltage meter.

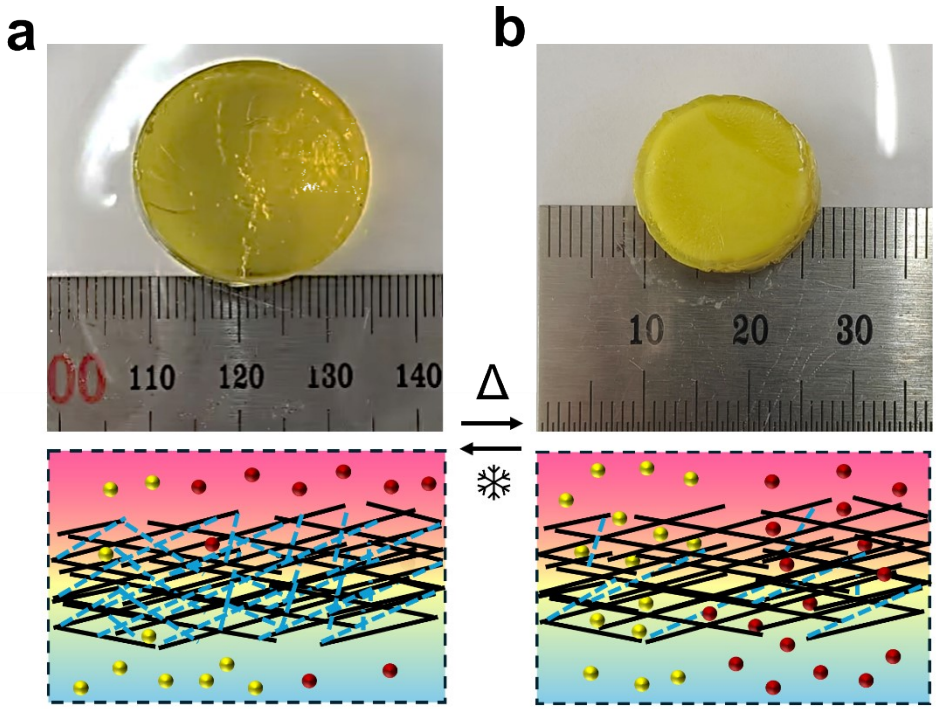

Fig. S9 Optical image and ion diffusion schematic of $N_9A_1F_{0.3}$ when a. T < LCST and b. T > LCST

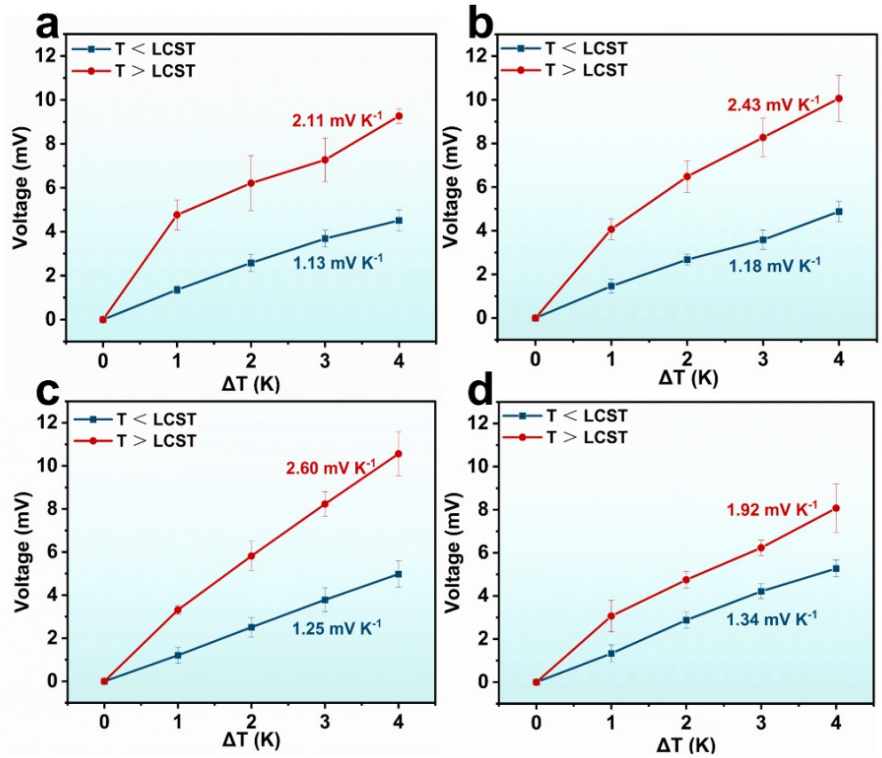

Fig. S10 The plot of ΔV-ΔT curves of p(NIPAAm-co-Am) hydrogel TECs. **(a)** $N_{10}A_0F_{0.3}$. **(b)** $N_{9.5}A_{0.5}F_{0.3}$. **(c)** $N_9A_1F_{0.3}$. **(d)** $N_{8.5}A_{1.5}F_{0.3}$.

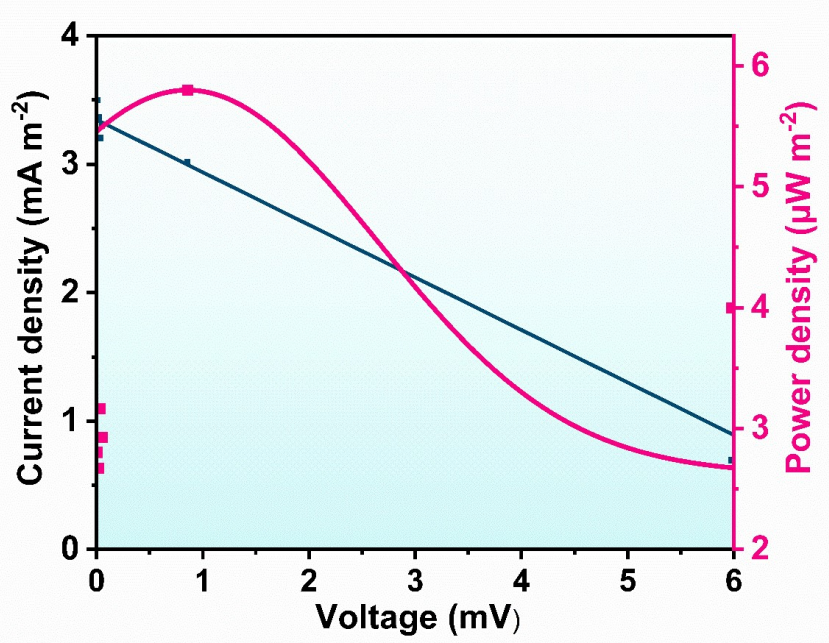

Fig. S11 The outpower density of $N_9A_1F_{0.3}$ hydrogel TEC at ΔT =5 K

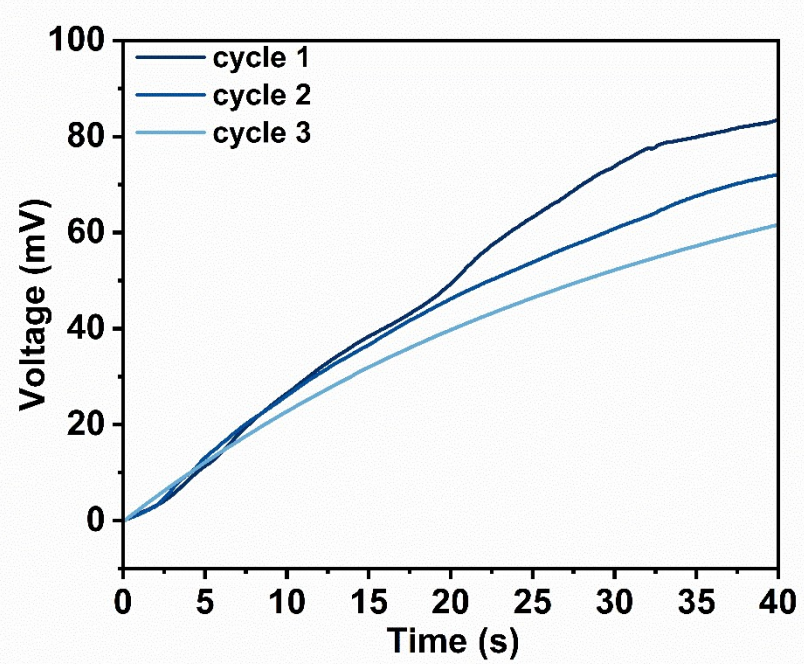

Fig. S12 Heating curve of TEC at a ΔT = 30 K and the cold end temperature of 20 °C

Table S1 The formulations for preparing composite hydrogels.

Rm	NIPAAm (g)	AAm (g)	MBAA (g)	APS (g)	TEMED (μL)	$[Fe(CN)_6]^{3-/4-}$ (M)
$N_{10}A_0F_0$	2.26	0	0.0032	0.016	20	0
$N_{9.5}A_{0.5}F_0$	2.148	0.07	0.0032	0.016	20	0
$N_9A_1F_0$	2.04	0.14	0.0032	0.016	20	0
$N_{8.5}A_{1.5}F_0$	1.92	0.21	0.0032	0.016	20	0
$N_0A_{10}F_0$	0	1.42	0.0032	0.016	20	0
$N_{10}A_0F_{0.3}$	2.26	0	0.0032	0.016	20	0.3
$N_{9.5}A_{0.5}F_{0.3}$	2.148	0.07	0.0032	0.016	20	0.3
$N_9A_1F_{0.3}$	2.04	0.14	0.0032	0.016	20	0.3
$N_{8.5}A_{1.5}F_{0.3}$	1.92	0.21	0.0032	0.016	20	0.3
$N_0A_{10}F_{0.3}$	0	1.42	0.0032	0.016	20	0.3

Table S2 Properties comparison of recent hydrogel TECs

Redox couple	Matrix	Stress (kPa)	Strain (%)	S_e (mV K^{-1})	Power density (μW m^{-2})	Ref.
I^-/I_3^-	PVA/Betaine	/	600	1	3	1
$Fe(CN)_6^{3-/4-}$	Gelatine/CBP	/	/	1.27	145.3	2
$Fe(CN)_6^{3-/4-}$	PVA	120	220	1.05	26.7	3
$Fe(CN)_6^{3-/4-}$	PVA/Gelatin	560	240	1.02	1.2	4
$Fe^{2+/3+}$	PVA/Gelatin	380	320	1.09	2.8	5
$Fe(CN)_6^{3-/4-}$	PA/PEI/PAAM	/	/	1.26	1.47	6
$Fe(CN)_6^{3-/4-}$	PNIPAM/AAM	36	580	2.60	157	This work

References

1 Z. Wang, N. Li, X. Yang, Z. Zhang, H. Zhang and X. Cui, *Microsyst. Nanoeng.*, 2024, **10**, 55.

2 Z. Xu, S. Lin, Y. Yin and X. Gu, *Chem. Eng. J.*, 2024, **493**, 152734.

3 Y. Jiang, H. Ye, S. Zhang, Y. Pan, Z. Huang, H. Li, J. Guo and C. Zhu, M. Yuan, B. Dai, J. Li, W. Yang, L. Gao, Y. Xie, *Nano Energy*, 2024, **131**, 110329.

4 C. Bai, X. Li, X. Cui, X. Yang, X. Zhang, K. Yang, T. Wang and H. Zhang, *Nano Energy*, 2022, **100**, 107449.

5 X. Li, J. Li, T. Wang, S.A. Khan, Z. Yuan, Y. Yin and H. Zhang, *ACS Appl. Mater. Interfaces*, 2022, **14**, 48743–48751.

6 J. Shen, C. Yang, Y. Ma, M. Cao, Z. Gao, S. Wang, J. Li, S. Liu, Z. Chen and S. Li, *EcoMat*, 2023, **6**, e12428.