Supplementary Information (SI) for Journal of Materials Chemistry A.

Electronic supplementary information

A Transparent, Tough, Highly Stretchable and Self-Adhesive Zwitterionic Dual-Network Eutectogel for Wearable Flexible Sensors

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Synthesis of pAA eutectogels

Different mass fractions (30 wt%, 40 wt%, 50 wt%, and 60 wt%) of AA monomer were added to 2.56 mL [ChCl][EG] with 0.0087 g MBAA and 0.0875 g I2959 and stirred to form a homogeneous solution. Subsequently, the above solutions were transferred into a custom-made glass mold and placed under 365 nm UV light (365 nm, 40 W) for 30 min, resulting in pAA eutectogels with different AA contents, which were named AA_3, AA_4, AA_5 and AA_6, respectively.

Synthesis of p(AA-co-HEMAA) eutectogels

Different mass ratios (10:0, 8:2, 6:4, 4:6, 2:8, and 0:10 for AA:HEMAA, respectively) of AA monomer and HEMAA monomer were added to 2.56 mL [ChCl][EG] with 0.0087 g MBAA and 0.0875 g I2959, respectively, and stirred to make a homogeneous solution. Subsequently, it was transferred into a customized glass mold and placed under 365 nm UV light (365 nm, 40 W) for 30 min, resulting in p(AA-co-HEMAA) eutectogels with different AA to HEMAA mass ratios, which were designated as AA, A_8H_2, A_6H_4, A_4H_6, A_2H_8, and HEMAA, respectively.

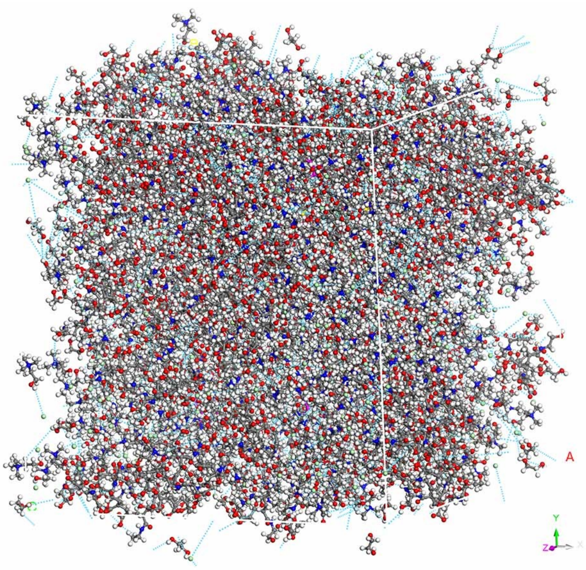

Fig. S1. Molecular dynamics calculations of AHD eutectogel. (DMAPS:AA-HEMAA:DES = 1:480:280, 3 DMAPS molecules; 1440 AA-HEMAA; 840 DES molecules)

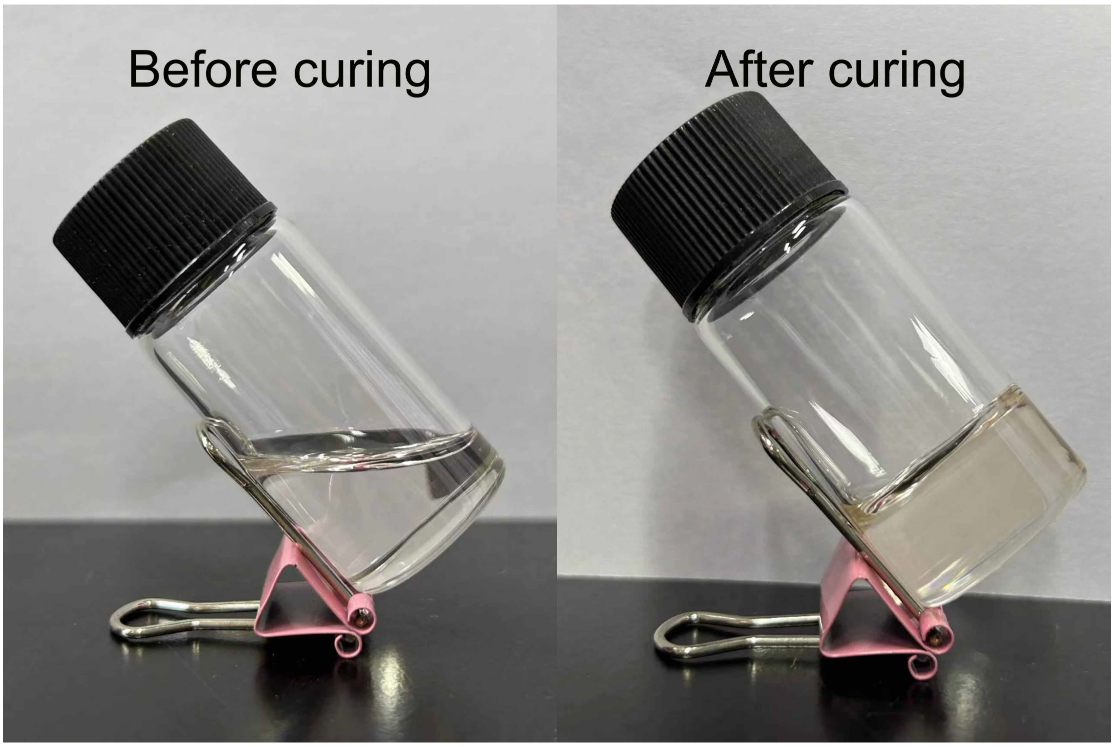

Fig. S2. Macroscopic picture of AHD eutectogel prepared.

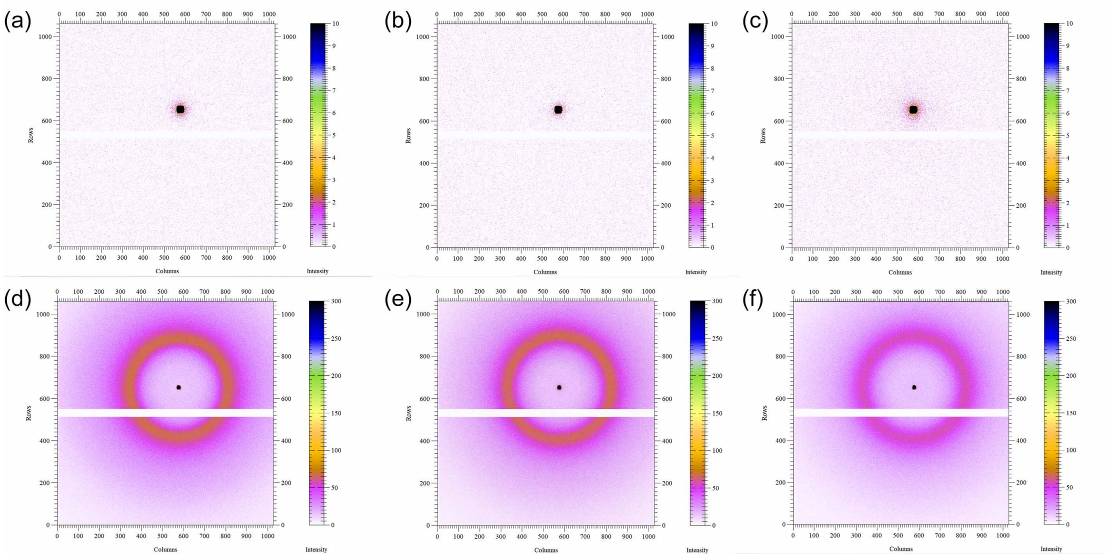

Fig. S3. (a-c) 2D SAXS and (d-f) 2D WAXS patterns of AA eutectogel, AH eutectogel and AHD eutectogel.

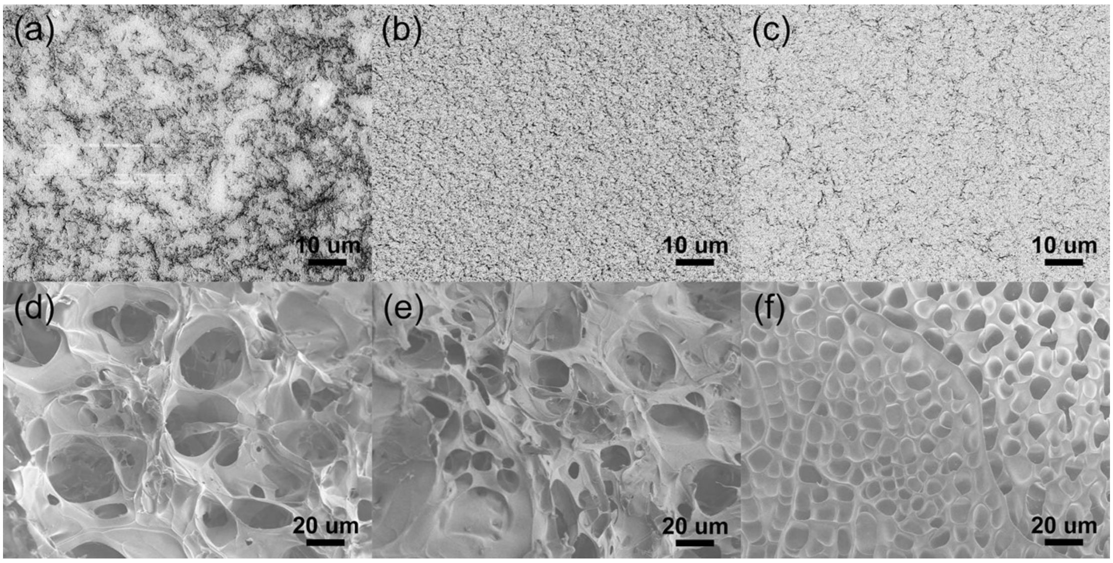

Fig. S4. SEM images of (a) AA eutectogel, (b) AH eutectogel and (c) AHD eutectogel. Post-freeze-drying SEM images: (d) AA eutectogel, (e) AH eutectogel and (f) AHD eutectogel.

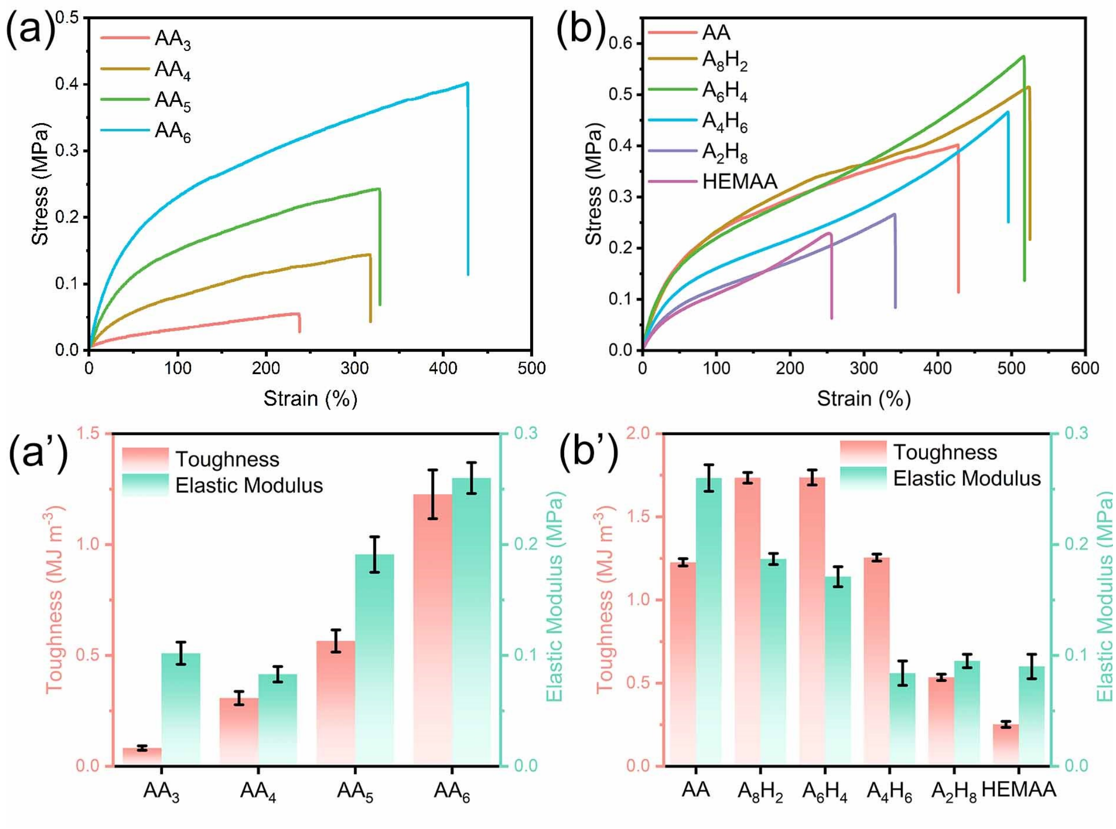

Fig. S5. Stress-strain curves and toughness-elastic modulus statistics for (a, a') AA eutectogels, (b, b') AH eutectogels.

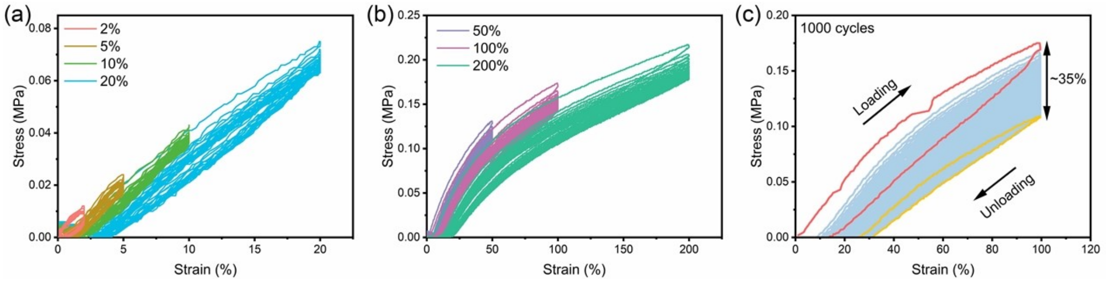

Fig. S6. (a, b) The continuous loading-unloading cyclic tests at fixed strains (2%-200%, 20 cycles per strain) of $A_6H_4D_5$ eutectogel. (c) The rapid continuous loading-unloading cyclic tests at 100% strain with 1000 cycles per strain for the $A_6H_4D_5$ eutectogel (tensile speed: 500 mm min^{-1}).

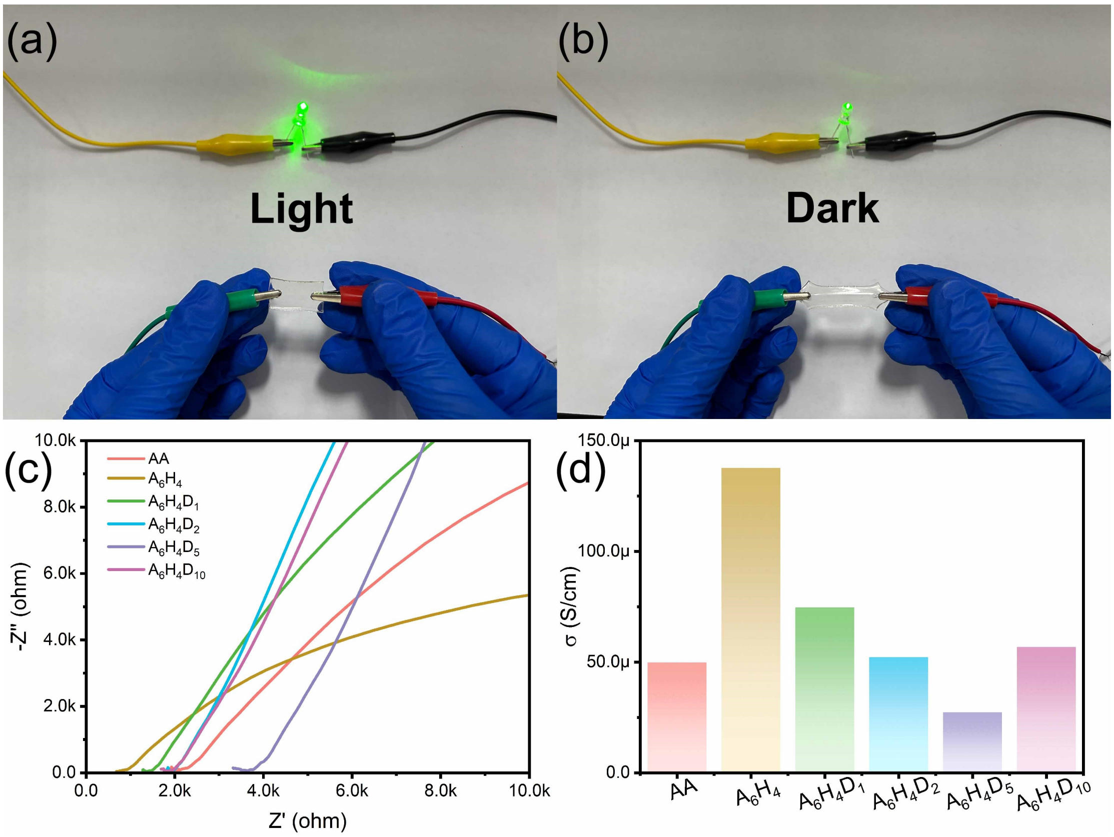

Fig. S7. LED brightness of $A_6H_4D_5$ eutectogel in (a) initial and (b) stretched states. AA eutectogel, AH eutectogel, and AHD eutectogel with different pDMAPS contents are shown in (c) EIS curves and (d) statistical plots of ionic conductivity.

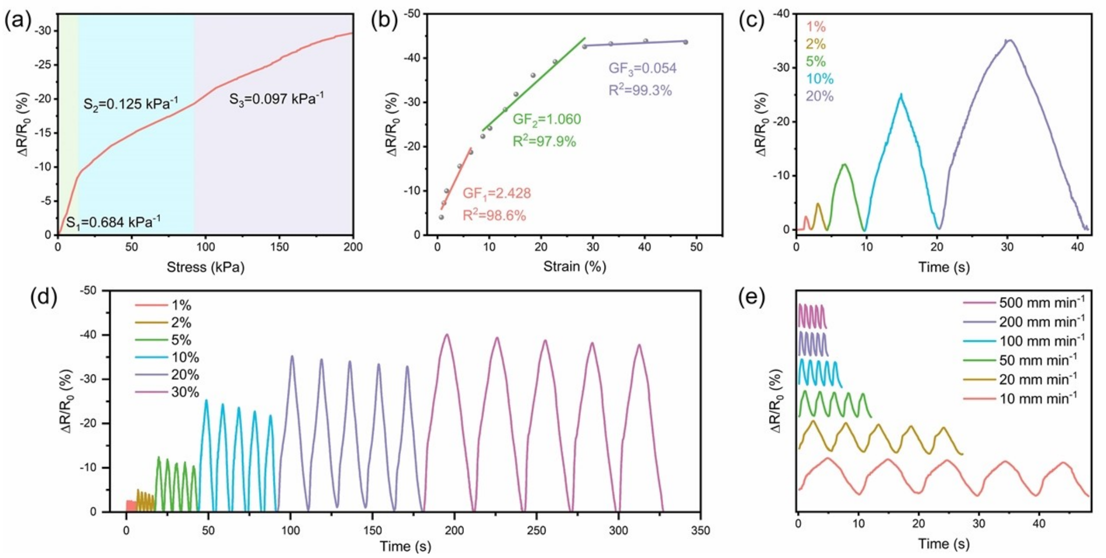

Fig. S8. (a) The pressure sensitivity of $A_6H_4D_5$ eutectogel-based sensors under various pressures. (b) The gauge factor of $A_6H_4D_5$ eutectogel-based sensors at different compressive strains. (c) Relative resistance changes of $A_6H_4D_5$ eutectogel-based sensors during continuous loading-unloading at different compressive strains (1%-20%). (d) Relative resistance changes of $A_6H_4D_5$ eutectogel-based sensor during continuous loading-unloading at a fixed compressive strain. (e) Relative resistance changes of $A_6H_4D_5$ eutectogel-based sensor when subjected to 5 cycles of 10% compressive strain at different tensile rates.

Table S1. Comparison of various properties between AHD eutectogel and previously reported eutectogels.

Material	Transmittance (%)	Stress (MPa)	Strain (%)	Toughness (MJ m^{-3})	Elastic modulus (MPa)	Adhesion strength (kPa, of glass)	Reference
PAA/MEA/CQS	92	0.10	1780	N/A	N/A	~75	[1]
SL-Fe^{3+}-DES	93.5	0.21	700	N/A	N/A	N/A	[2]
Zn-PAM-H_2O	N/A	0.49	1347	N/A	N/A	N/A	[3]
poly(NAGA/ChCl/Gly/H_2O)	N/A	<0.6	310	N/A	N/A	N/A	[4]
PHEAA/gelatin/MXene	N/A	0.5	940	1.39	0.03035	36.27	[5]
PVDF-HFP/DES	N/A	~0.6	~300	N/A	N/A	N/A	[6]
pDMAPS/p(AA-co-HEMAA)	89	0.569	637	2.529	0.204	76.9	This work

Reference

1. Y. Wu, L. Yang, J. Wang, S. Li, X. Zhang, D. Chen, Y. Ma and W. Yang, *ACS Appl. Mater. Interfaces*, 2023, **15**, 36759-36770.
2. Y. Feng, J. Yu, D. Sun, C. Dang, W. Ren, C. Shao and R. Sun, *Nano Energy*, 2022, **98**, 107284.
3. Y. Wu, Y. Deng, K. Zhang, J. Qiu, J. Wu and L. Yan, *ACS Appl. Energy Mater.*, 2022, **5**, 3013-3021.
4. Y. Liu, X. Zhang, B. Li, H. Chen, H. Li, J. Chen and H. Dong, *Chem. Eng. J.*, 2023, **461**, 141965.
5. B. Guo, M. Yao, S. Chen, Q. Yu, L. Liang, C. Yu, M. Liu, H. Hao, H. Zhang, F. Yao and J. Li, *Adv. Funct. Mater.*, 2024, **34**, 2315656.
6. Z. Li, S. Zhang, Z. Jiang, D. Cai, C. Gu and J. Tu, *Materials Chemistry and Physics*, 2021, **267**, 124701.