

**Self-healable and freeze-resistance polyelectrolyte based on EG anchor chain and
dual dynamic reversible interaction as highly sensitive ionic skins**

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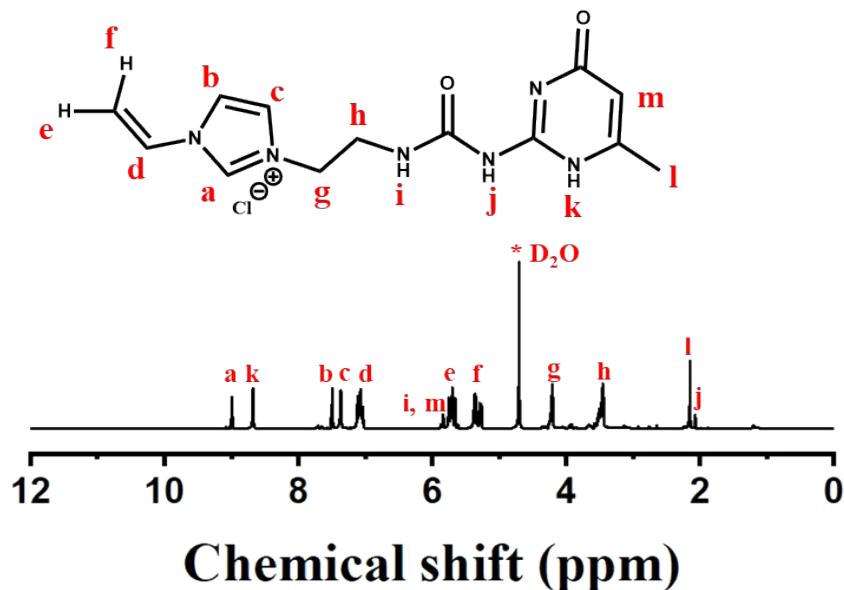
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Supporting information:



Chemical shift (ppm)

Fig. S1. ¹H-NMR spectra of UPy-VIm

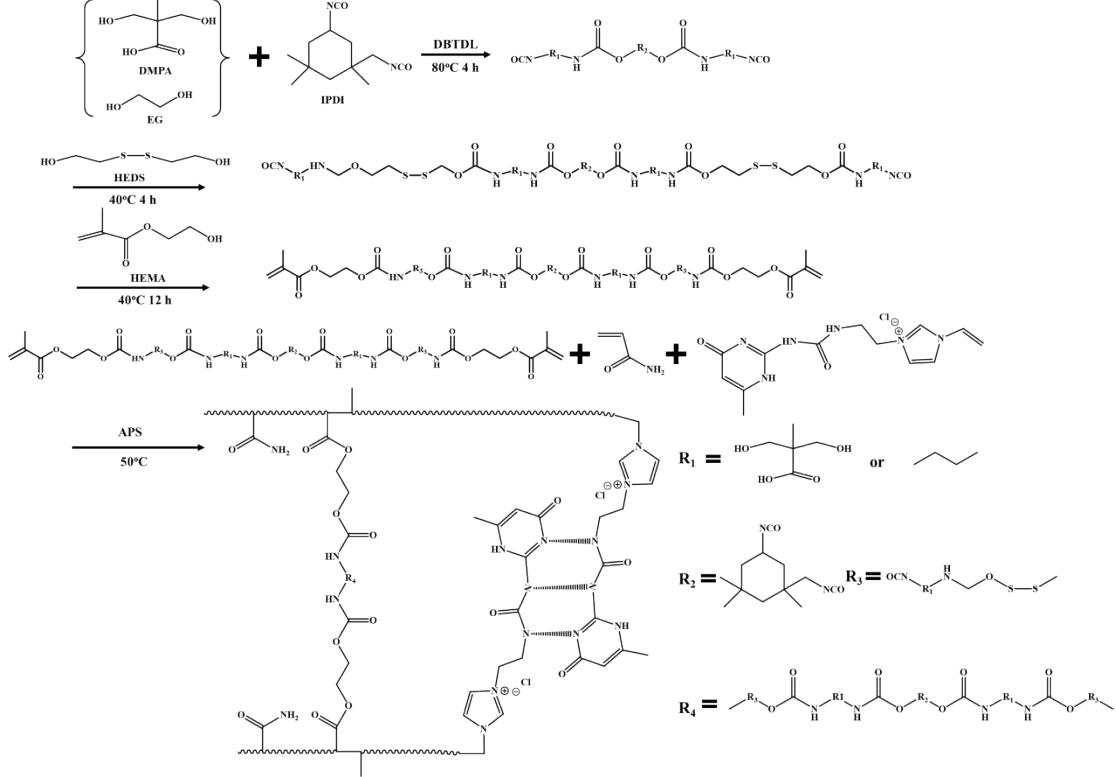


Fig. S2. Synthesis routine of EG based dual functional waterborne anionic polyurethane acrylates (DB-waPU) and DF-hydrogel

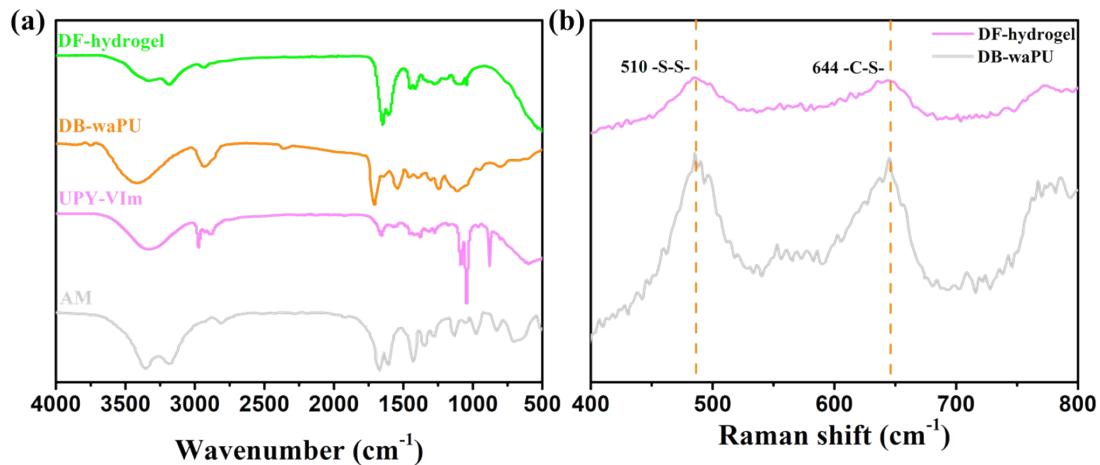


Fig. S3. (a) FTIR spectra of AM monomer, DB-waPU and DF-hydrogel, (b) Raman spectra of DB-waPU and DF-hydrogel

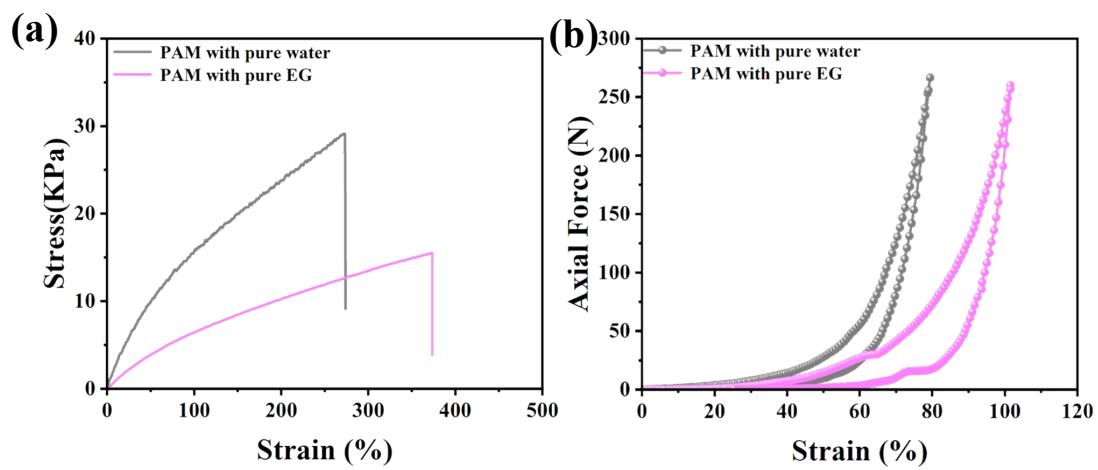


Fig. S4. Mechanical comparison of PAM with pure water and PAM with pure EG: (a) stretchability, (b) compressibility

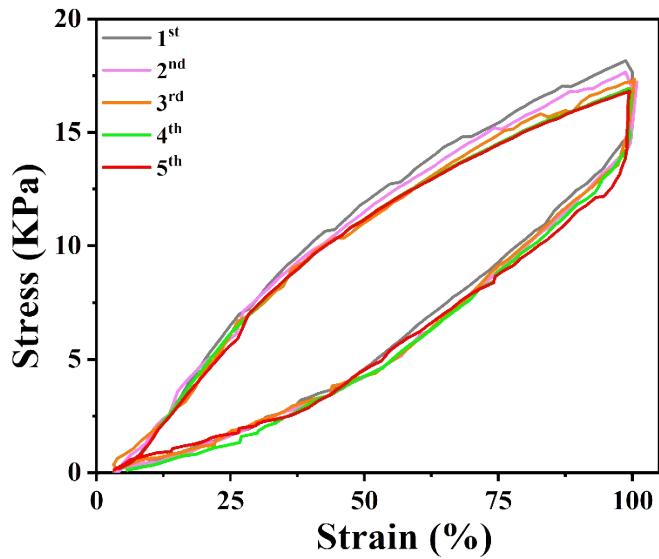


Fig. S5. Cycles of loading-unloading tests for DF-hydrogel

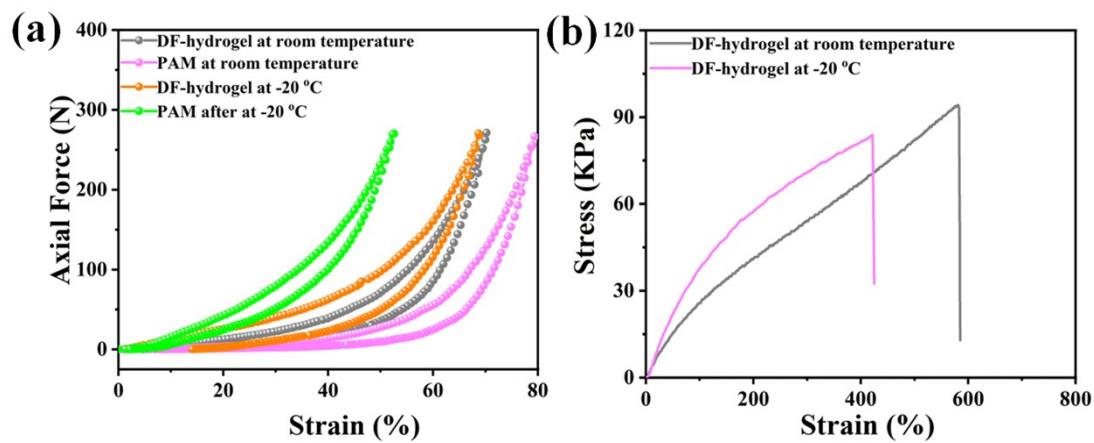


Fig. S6. (a) Compression tests with different hydrogel samples, (b) Anti-freezing performances for DF-hydrogel

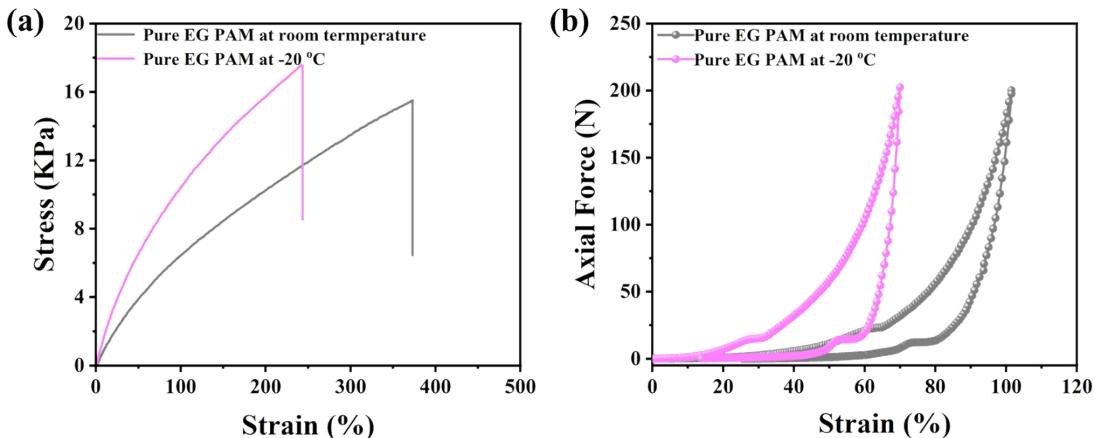


Fig. S7. Anti-freezing of mechanical properties for PAM organogel: (a) stretchability and (b) compressibility

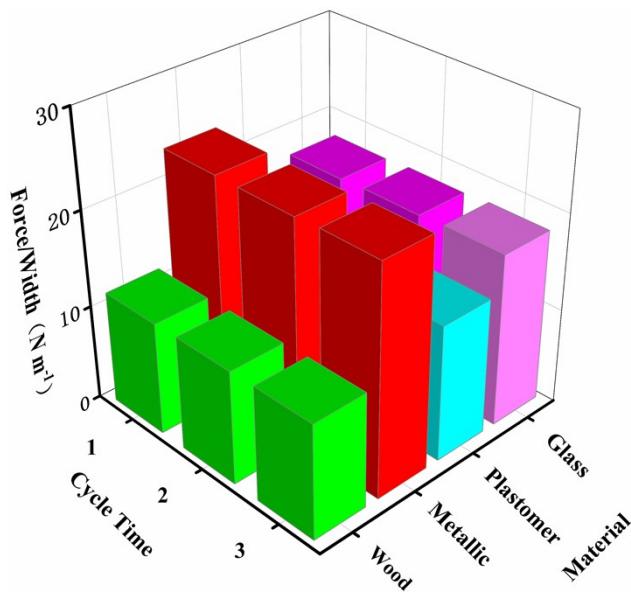


Fig. S8. Adhesion performance of DF-hydrogel to different material substrates

Fig. S8 illustrates that the DF-hydrogel has good adhesion properties to different materials, in which the strongest adhesion is to metal substrates (23.8 N m^{-1}), and the difference in the adhesion properties to other substrates is not significant (mainly maintained between 14 and 18 N m^{-1}). This is because the -OH on the substrate surface can form hydrogen bonds with DF-hydrogel to impart adhesion properties.

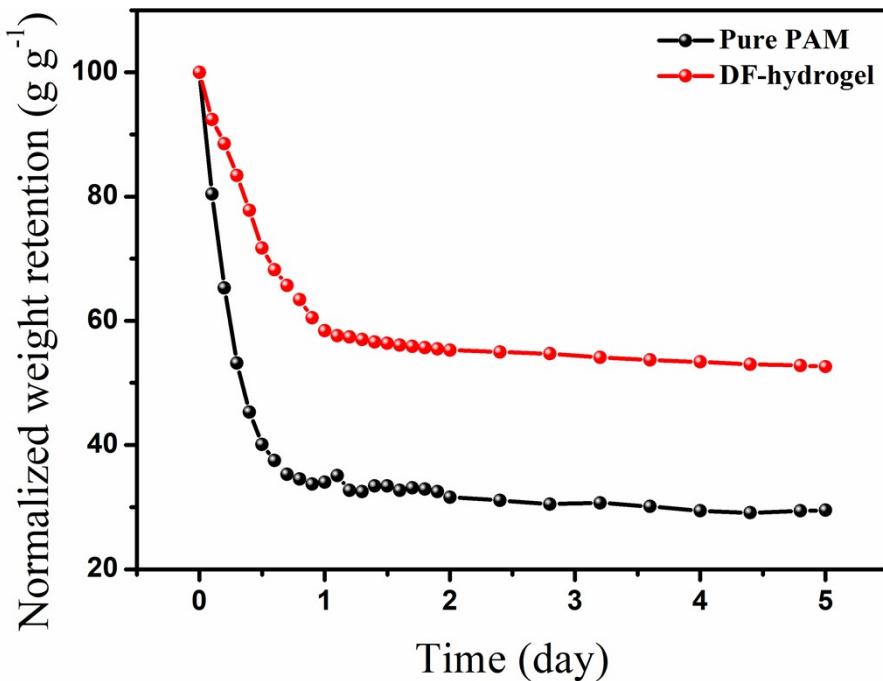


Fig. S9. Water retention of DF-hydrogel and pure PAM hydrogel

Water retention is an important indicator of the durability of hydrogels, as the performance of hydrogels always requires activation in an aqueous environment. Fig. S6 shows that the rate of water evaporation in DF-hydrogel is significantly less than in pure PAM hydrogel, and their water retention rates after 5 days are 56.2% and 26.9% respectively.

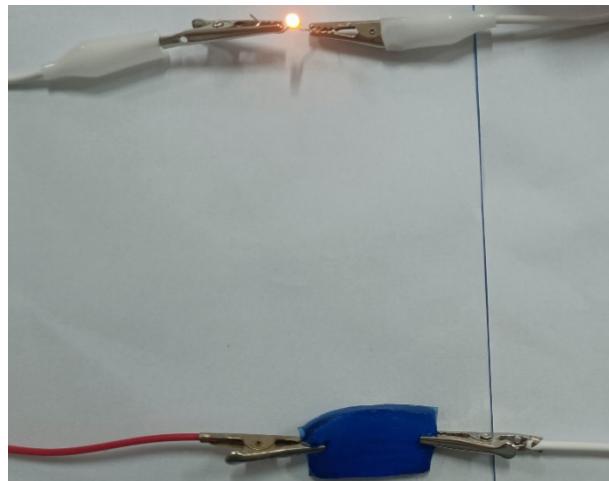


Fig. S10. Photo of DF-hydrogel as wire for lighting the LED

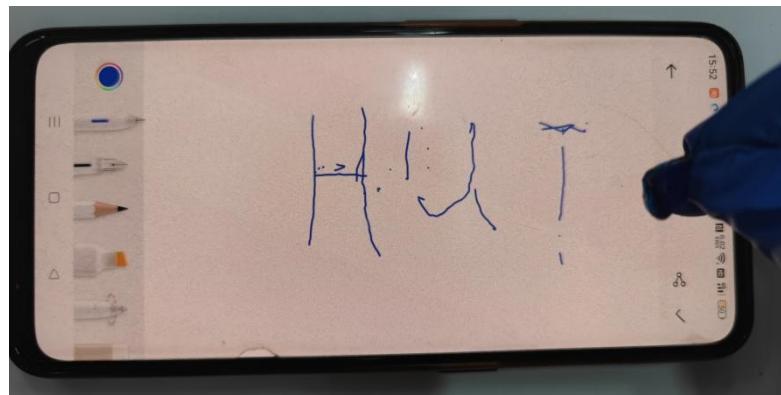


Fig S11. Photo of DF-hydrogel ionic skin acted as stylus to write text on the screen of smartphone

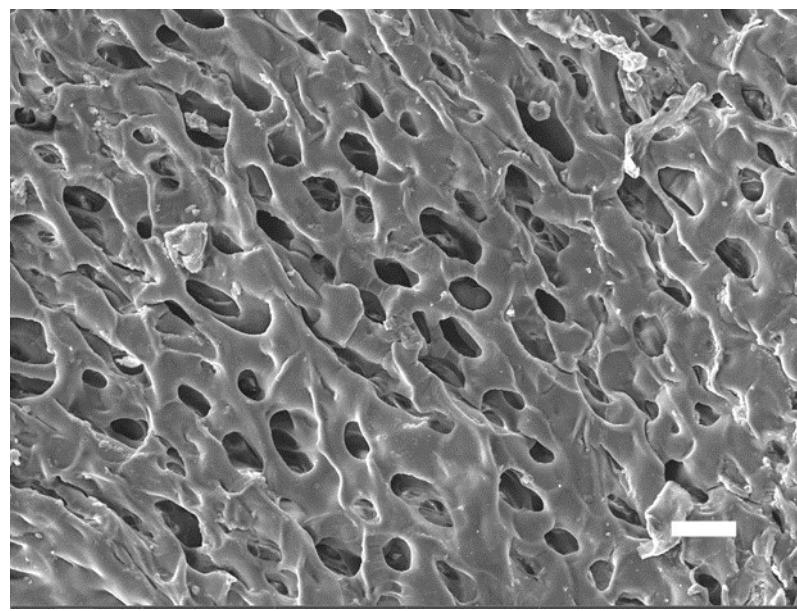


Fig. S12. SEM image of the freeze-dried DF-Hydrogel, scale bar: 20 μm .

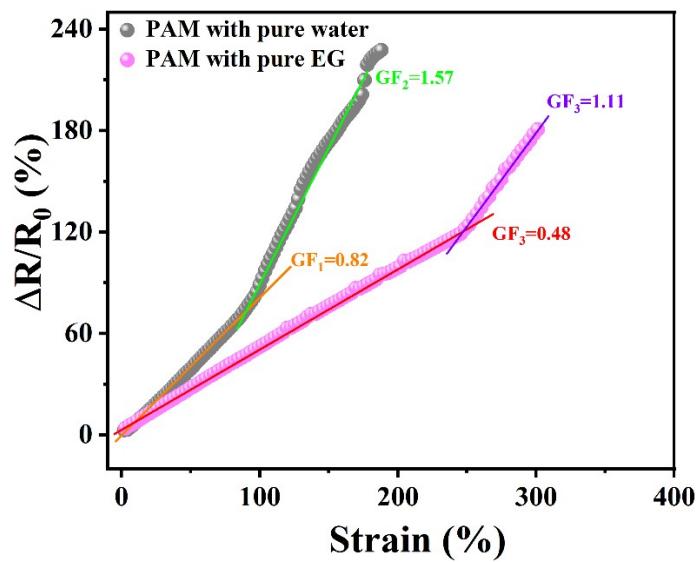


Fig. S13. Relative resistance changes of PAM hydrogel and PAM organogel as a function of strain.



Fig. S14. Photos of the conductivity of hydrogels at low temperature: (a) pure PAM and (b) DF-hydrogel

Table S1. Comparison of comprehensive performances of the hydrogel-based sensors

Ionic skin	Self-healing capability	Freeze-resistant capability	Gauge factor	Sensitive strain	Ref.
DF-Hydrogel	YES	YES	1.99	0-300%	This work
PVA/TA@talc/AlCl ₃ /EG	NA	YES	1.3	0-100%	Ref. S1
PAAM/SA/NaCl	YES	NA	2.66	0.3-1800%	Ref. S2
PAM/NaCl	NA	NA	NA	0-500%	Ref. S3
PAM/LiCl/PDMS	NA	NA	0.84	0-50%	Ref. S4
PDA-PPy/PAAM	NA	NA	NA	NA	Ref. S5
PVA/Gly/CB/CNT	NA	YES	2.01	0-700%	Ref. S6
PAAM/SA/CNT/CaCl ₂	NA	YES	3.125	0-400%	Ref. S7
PVA/PAA/CNT/FeCl ₃ /PEDOT-PSS/EG	YES	YES	1.6	0-100%	Ref. S8
PVA/SWCNT	YES	NA	1.51	0-1000%	Ref. S9
PANI/PSS-UPy	YES	NA	3.4	0-300%	Ref. S10
PANI/P(AAm-co-HEMA)	YES	NA	1.48	0-300%	Ref. S11

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