

Supporting information

A Mussel-inspired Semi-interpenetrating Structure Hydrogel with Superior stretchability, Self-adhesive, and pH Sensitivity for Smart Wearable Electronics

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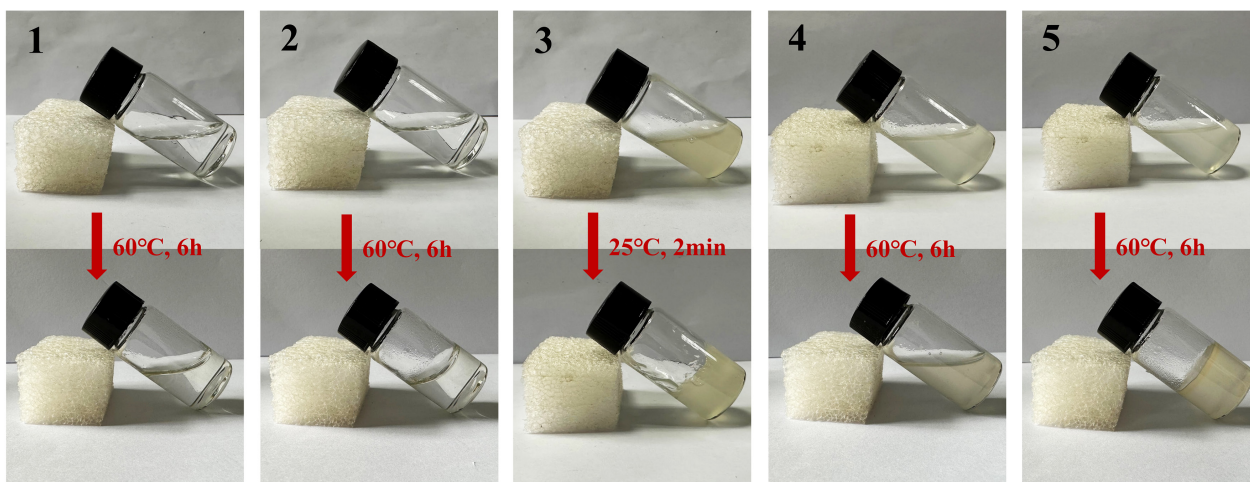


Fig. S1 Graph of the result of comparative tests

	AA (ml)	Al(NO ₃) ₃ ·9H ₂ O (mg)	CMC (g)	APS (mg)	H ₂ O (ml)
Sample 1	0.75	/	/	7.5	3.75
Sample 2	0.75	26.25	/	7.5	3.75
Sample 3	/	26.25	0.09	/	4.5
Sample 4	0.75	26.25	0.09	/	3.75
Sample 5	0.75	26.25	0.09	7.5	3.75

Table. S2 Recipes for comparative tests

To further investigate the ionic coordination in hydrogels, five comparative experiments were carried out with the formulations shown in Table S2. As shown in Fig S1 and samples 1-2, APS would trigger the polymerization of AA monomer at 60°C, so that AA would polymerize from monomer to

PAA molecular chains. After 6 hours of reaction, the viscosity of the samples increased due to the intertwining of molecular chains. Under the same reaction conditions, after the further introduction of Al^{3+} , new chain entanglement sites would be formed due to the coordination interaction between Al^{3+} and the carboxyl groups on the PAA molecular chains, which would make the entanglement between the PAA molecular chains tighter and subsequently the viscosity of the samples would be even higher. At the same time, significant amounts of carboxyl groups were also present on the CMC molecular chains, which would rapidly form coordination interactions with Al^{3+} at room temperature, causing the samples to gel (sample 3). However, in the presence of AA monomers, this physical cross-linking was inhibited, making it difficult to cross-link the molecular chains. This was attributed to the fact that the carboxyl groups on the CMC chains were difficult to ionize under acidic conditions and the molecular chains were in a contracted state, so the coordination interaction with Al^{3+} was weakened (sample 4).

From this, it could be speculated that in the fully gelled sample 5, CMC was penetrated in the PAA network as a linear macromolecule, while Al^{3+} existed mainly in ionic coordination with the carboxyl groups on the PAA molecular chains, i.e., hydrogels with the semi-interpenetrating network and multiple chain entanglement sites were formed.

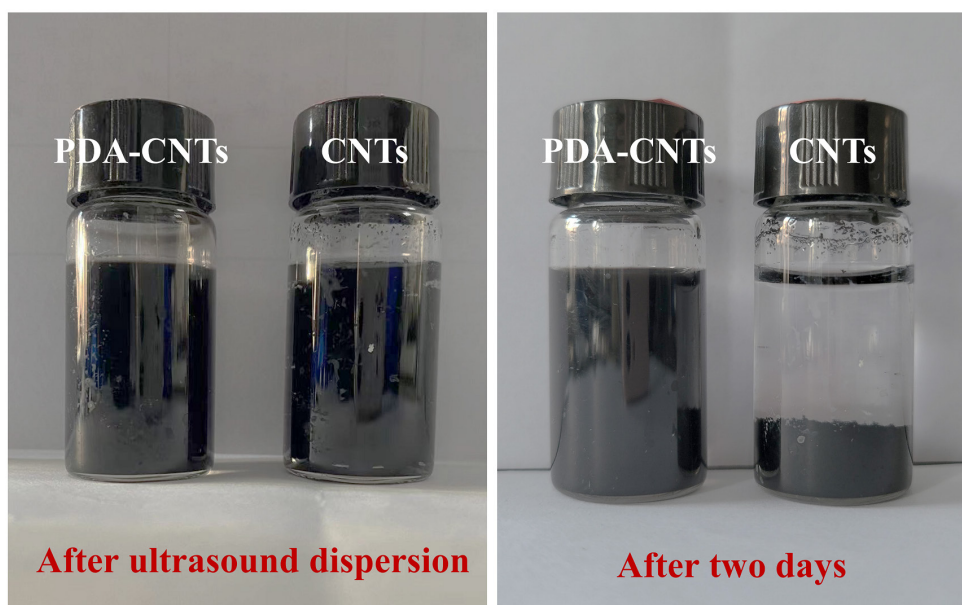


Fig. S2 Images of the dispersion characterization of CNTs and PDA-CNTs.

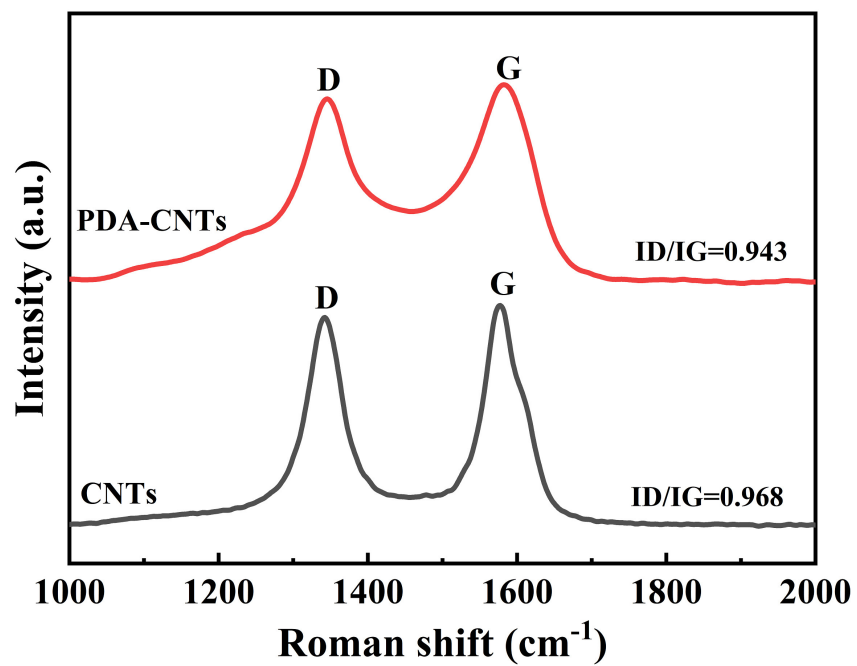


Fig. S3 The Raman spectra of CNTs and PDA-CNTs.

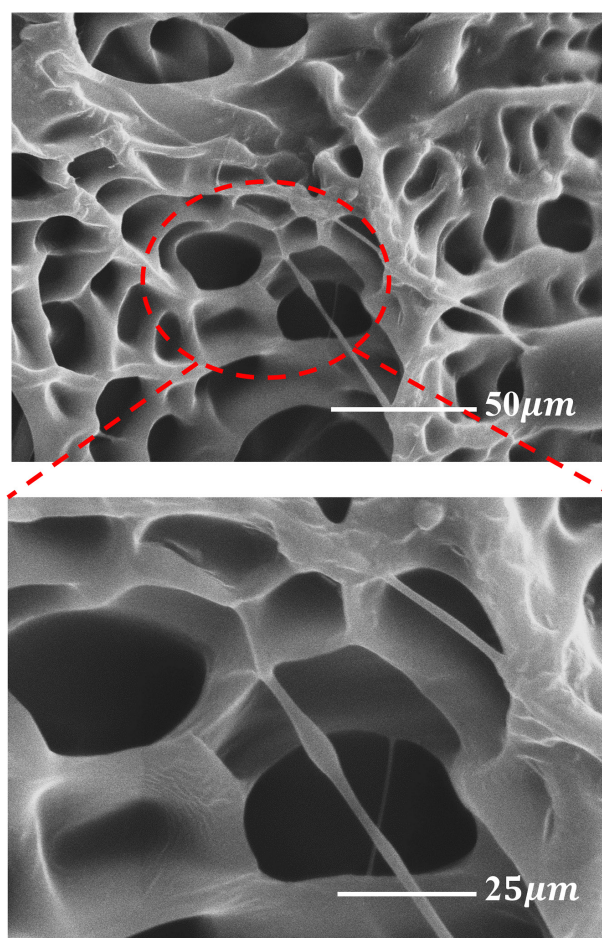


Fig. S4 The SEM images of CMC/PAA hydrogel.

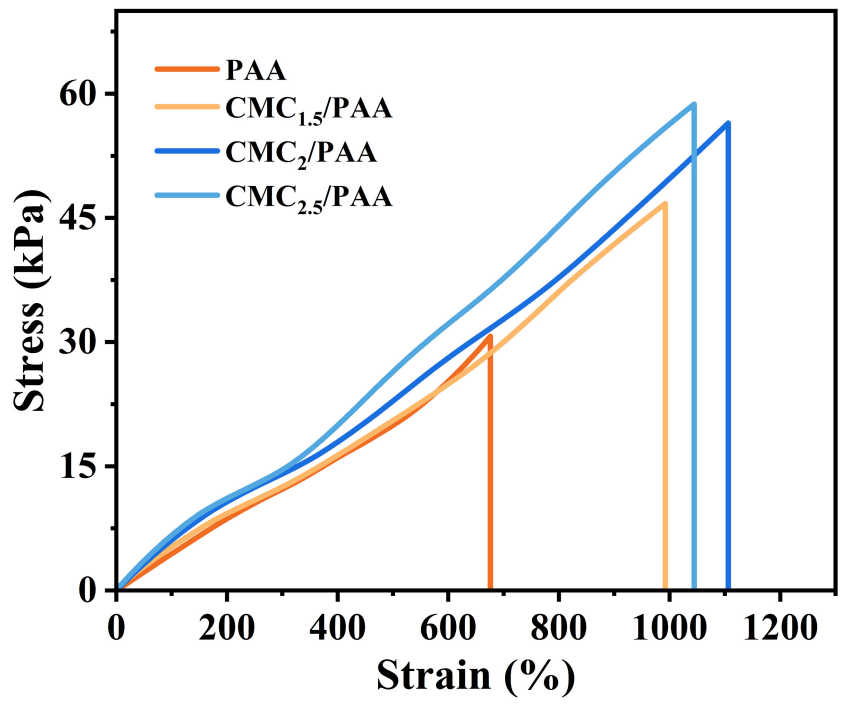


Fig. S5 The tensile-strain curves of PAA and CMC/PAA hydrogels (tested with CMC as the variable).

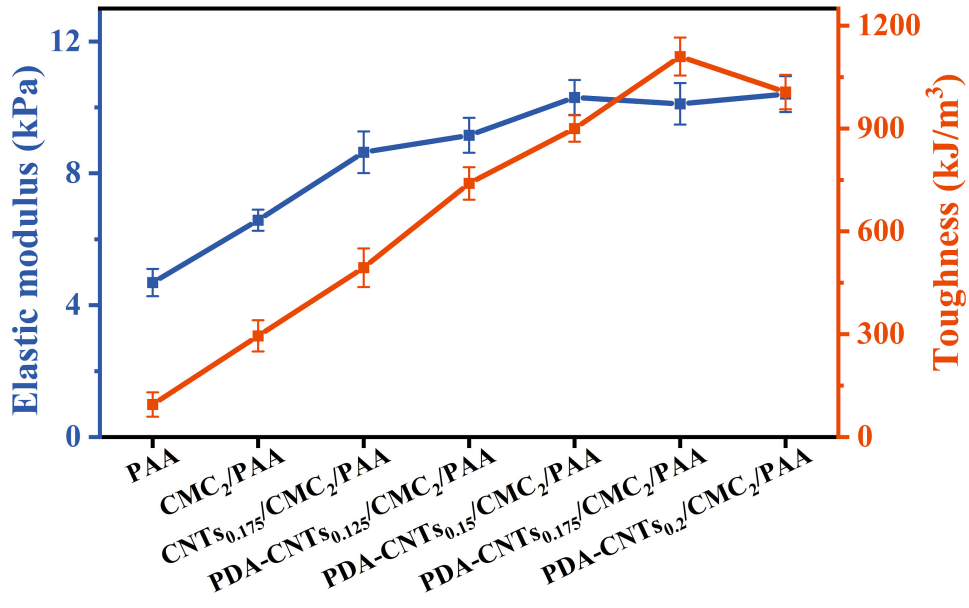


Fig. S6 The elastic modulus and toughness of PAA, CMC₂/PAA, CNT_{0.175}/CMC₂/PAA, and PDA-CNTs/CMC₂/PAA hydrogels.

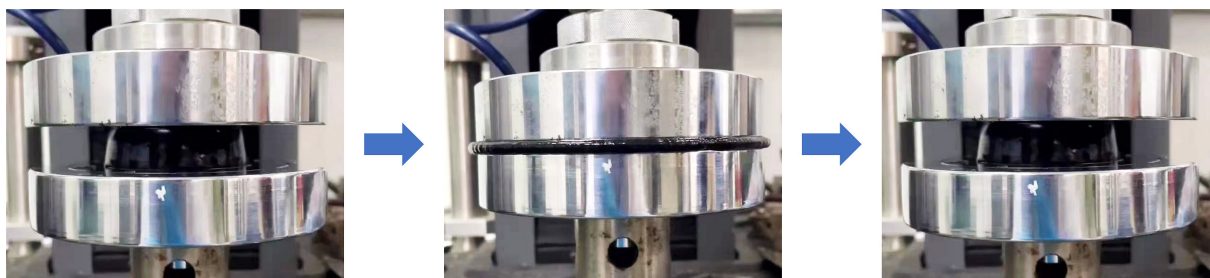


Fig. S7 The digital photos of PDA-CNTs/CMC/PAA hydrogel compression test.

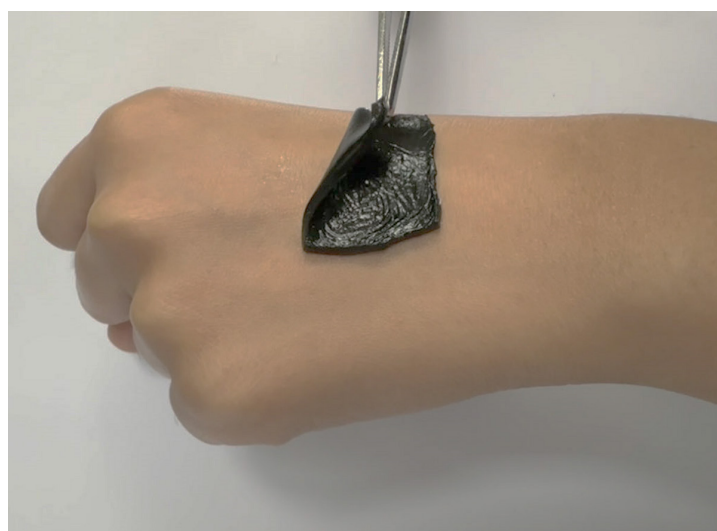


Fig. S8 Hydrogel detaching from human skin.

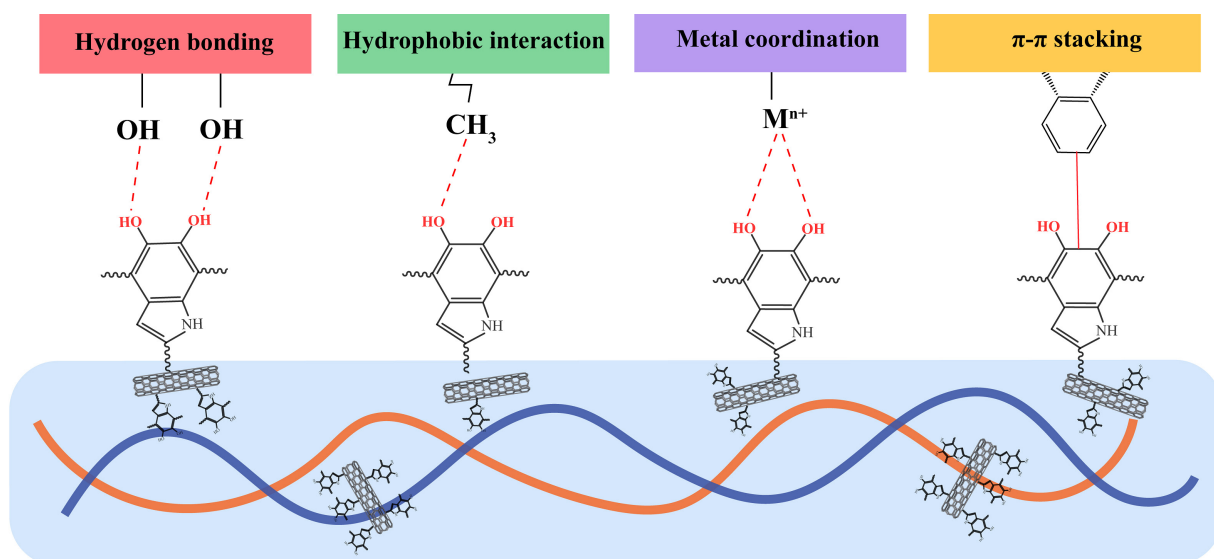


Fig. S9 Adhesion mechanism between the hydrogel and various substrates.

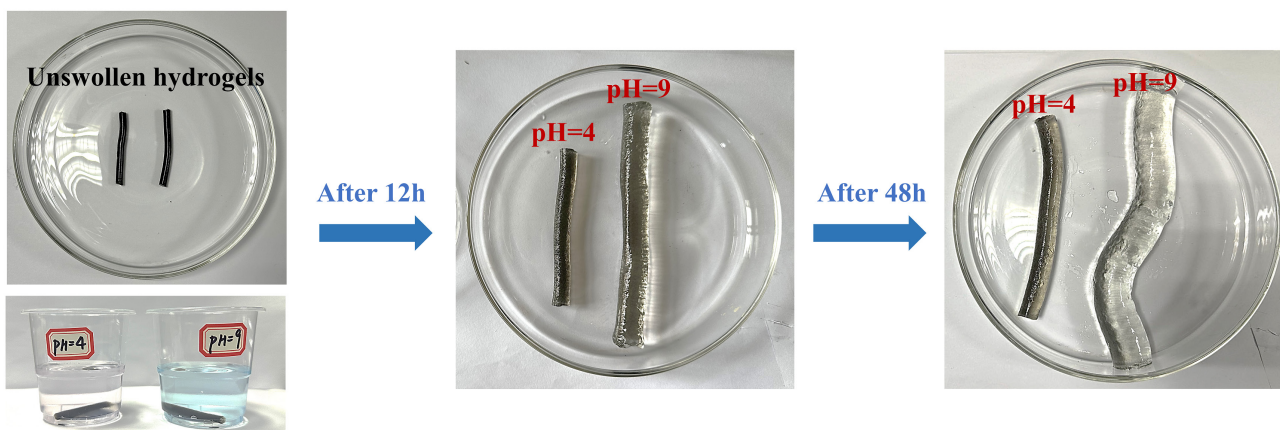


Fig. S10 Photographs of PDA-CNT_{0.175}/CMC₂/PAA hydrogels before swelling, and after swelling in different pH solutions for 12h and 48h, respectively.

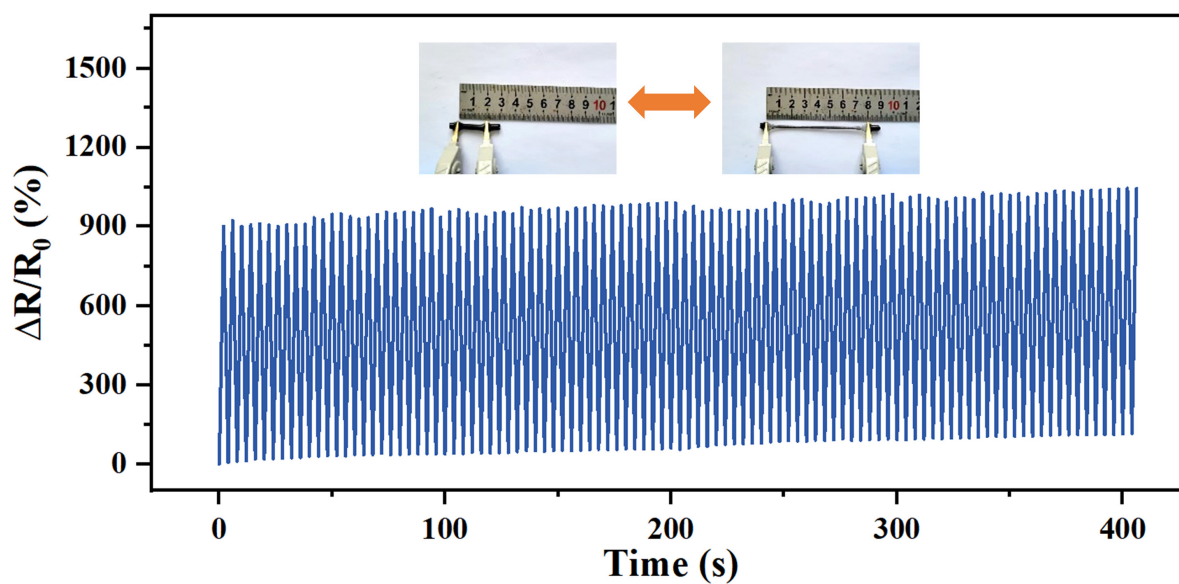


Fig. S11 The 100 repeated load-unload cycles at 300% tensile strain.