

Supplementary Materials

Interfacially integrated Janus bilayer hydrogel for spatially decoupled wet adhesion and hydrated lubrication

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Quantification of Anti-adhesion Efficiency

The anti-adhesion efficiency was determined from the reported adhered cell/bacterial number, colony number, or OD value according to the equation:

$$\text{Anti-adhesion efficiency (\%)} = \left(1 - \frac{A_{\text{sample}}}{A_{\text{control}}}\right) \times 100\%$$

Supplementary Table 1. Formulation of samples synthesized in this study

Sample	ATAC (g)	PEA (g)	A-Keto (g)	MBAA (g)	HA-CHO	CMC-NH ₂
AP-HC-1	4.36	8.97	0.02	0.01	1% (w/v)	1% (w/v)
AP-HC-2	2.90	10.26	0.02	0.01	1% (w/v)	1% (w/v)
AP-HC-3	2.19	10.89	0.02	0.01	1% (w/v)	1% (w/v)

Optimization of AP and AP-HC formulations

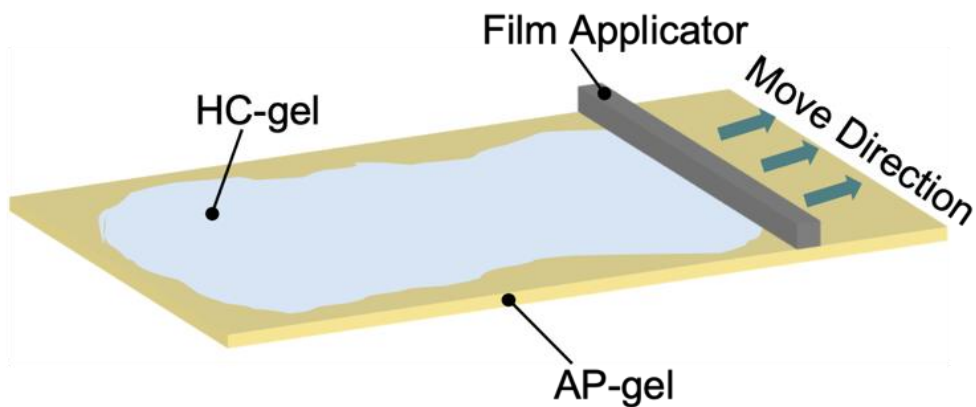
Based on the comprehensive evaluation of adhesion performance, mechanical properties, and structural stability, AP-2 was selected as the optimized AP formulation, while AP-HC-2 was selected as the corresponding Janus bilayer hydrogel formulation. Therefore, all experiments involving AP gel were performed using AP-2, and all experiments involving AP-HC bilayer hydrogel were conducted using AP-HC-2 unless otherwise specified. The other formulations were only used for comparative optimization studies.

Supplementary Table 2. Oxidation degree and amination degree of HA-CHO and CMC-NH₂

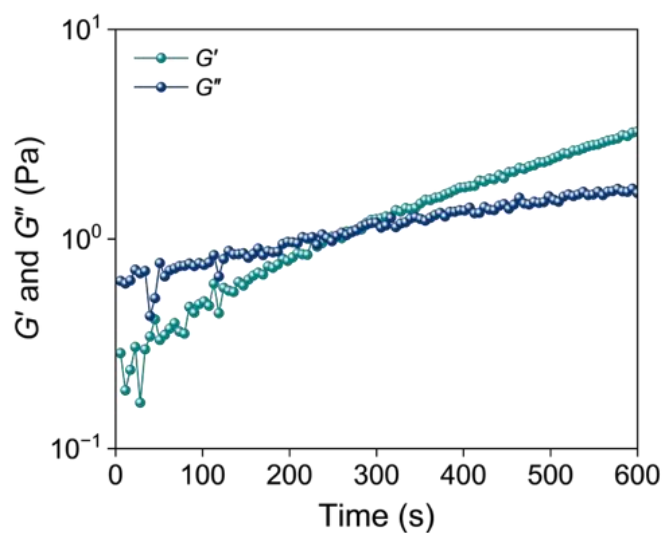
	Oxidation degree	Modification degree
HA-CHO	34.8%	-
CMC-NH ₂	-	27.1%

Reactive-group stoichiometry

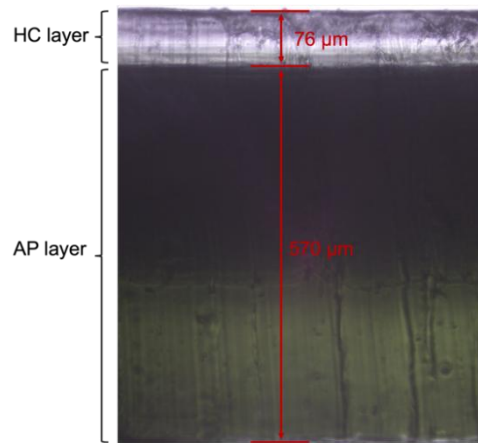
Since HA-CHO and CMC-NH₂ solutions (both 1% w/v) were mixed at an equal volume ratio, the corresponding mass ratio was 1:1. Based on the functionalization degrees of HA-CHO (34.8%) and CMC-NH₂ (27.1%), the theoretical molar ratio of aldehyde groups to amino groups (CHO/NH₂) was calculated to be approximately 1:1.25.



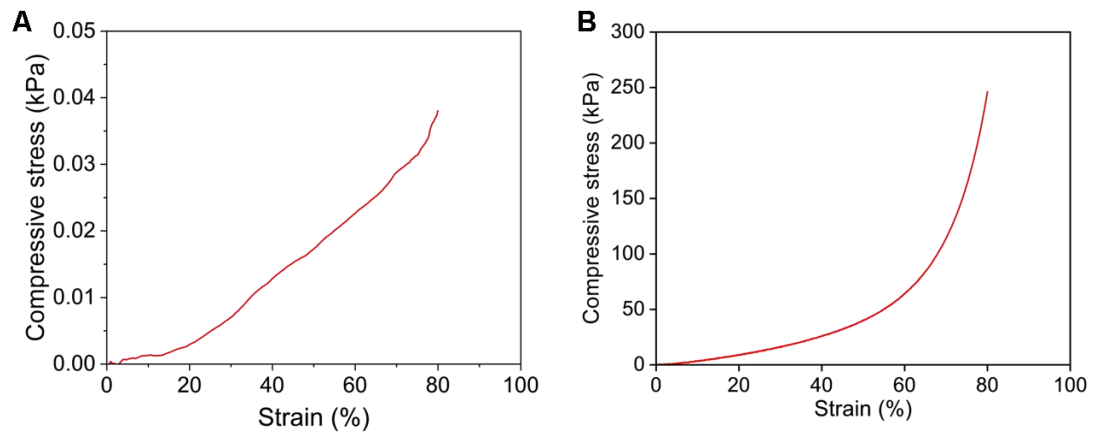
Supplementary Figure 1. Schematic illustration of the fabrication of the AP-HC bilayer hydrogel by in situ blade-coating of the HC layer onto the AP surface.



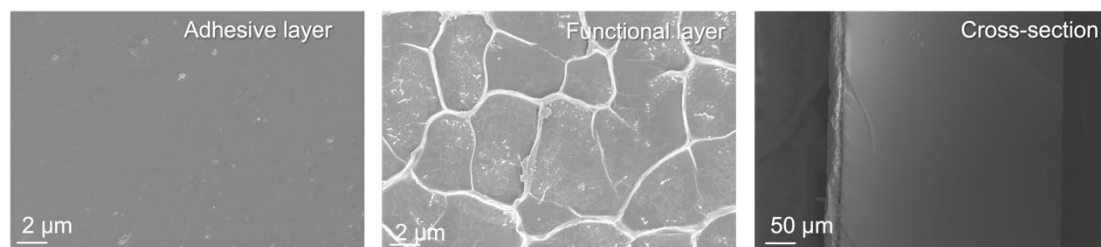
Supplementary Figure 2. Rheological characterization of the gelation behavior of HC-gel. Time-dependent evolution of storage modulus (G') and loss modulus (G'') after mixing HA-CHO and CMC-NH₂ solutions.



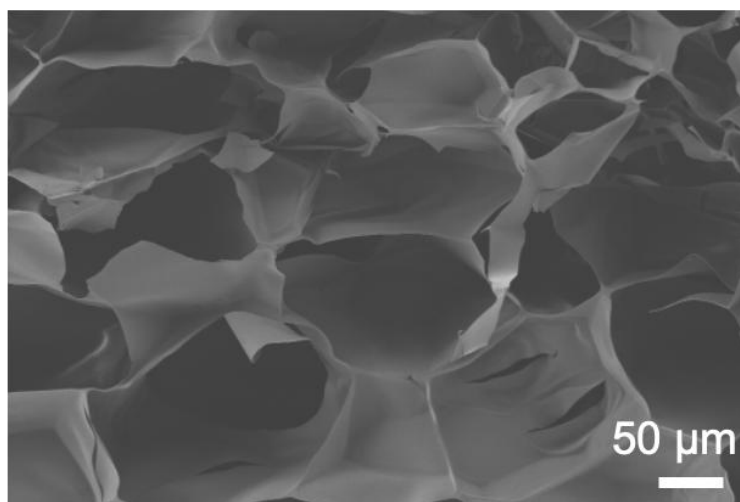
Supplementary Figure 3. Cross-sectional optical microscopy image of the AP-HC hydrogel.



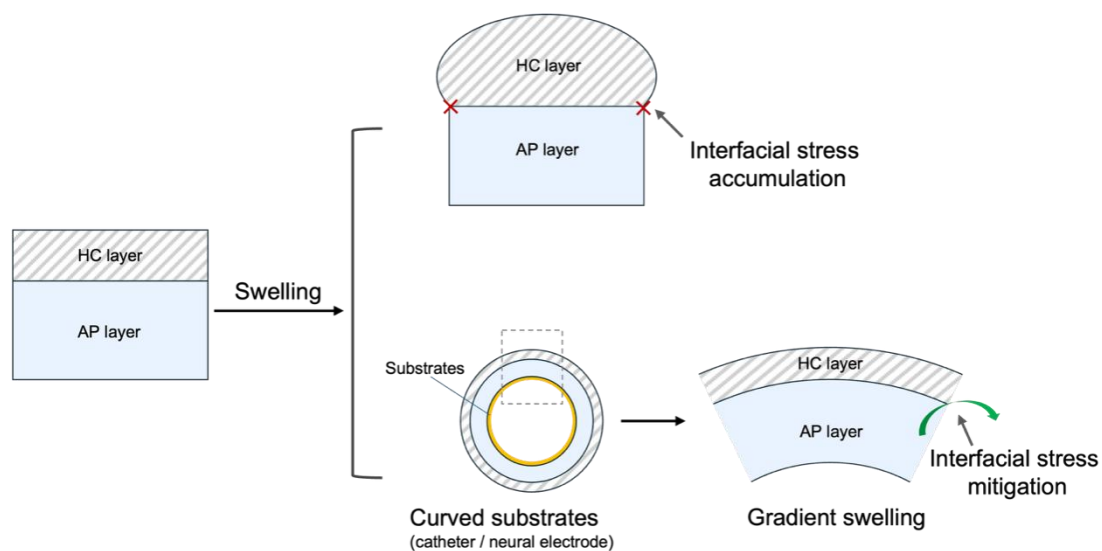
Supplementary Figure 4. (A) Compression curve of HC-gel. (B) Compression curve of AP-gel.



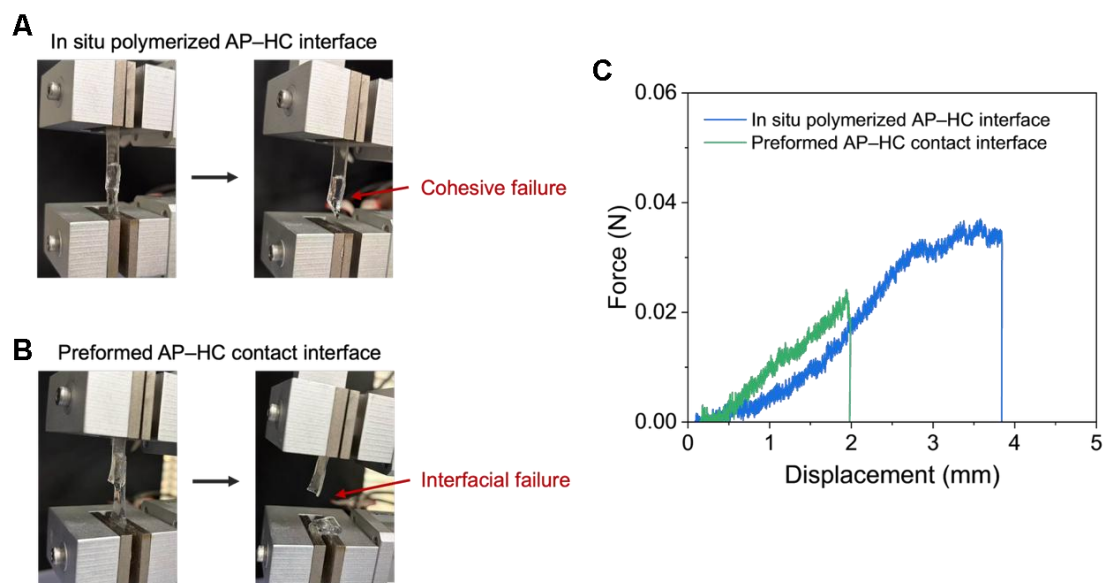
Supplementary Figure 5. SEM images of the adhesive side, lubricating side, and cross section of the AP-HC bilayer hydrogel



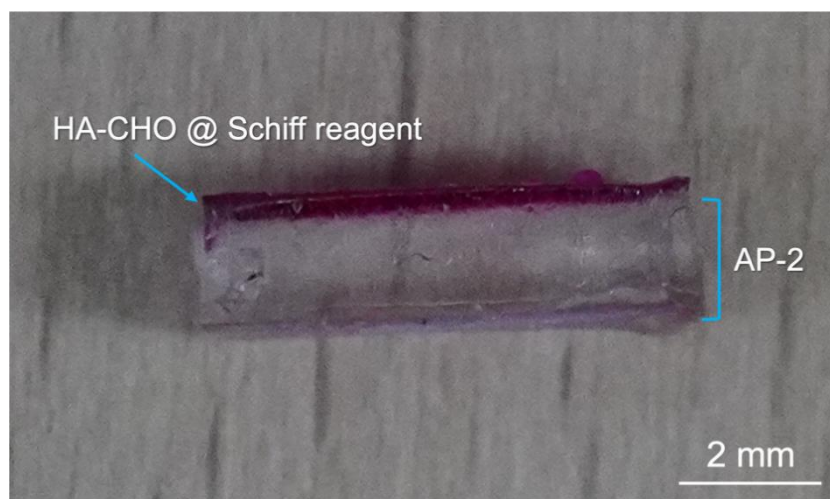
Supplementary Figure 6. SEM image showing the highly porous microstructure of HC-gel.



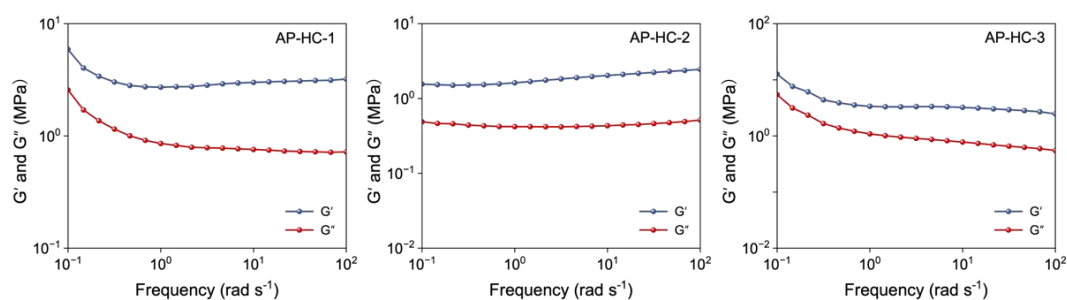
Supplementary Figure 7. Schematic illustration of gradient swelling–mediated interfacial stress relaxation in AP–HC bilayer hydrogels on curved substrates.



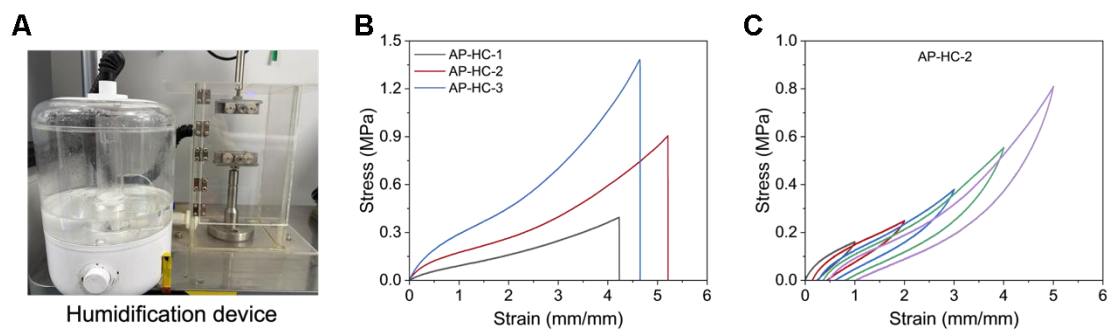
Supplementary Figure 8. Interfacial bonding performance of AP–HC hydrogels evaluated by lap-shear tests. Photographs showing (A) cohesive failure of the hydrogel with an in situ polymerized AP–HC interface and (B) interfacial failure of the preformed AP–HC contact interface after lap-shear testing. (C) Representative force–displacement curves of the in situ polymerized and preformed AP–HC interfaces.



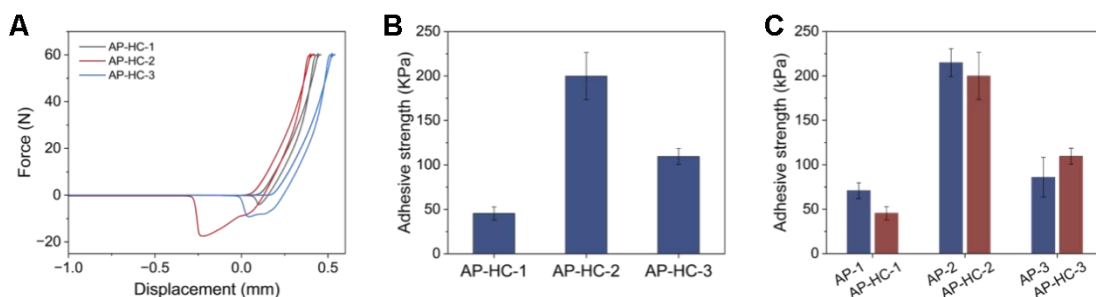
Supplementary Figure 9. Qualitative visualization of HA-CHO penetration into the superficial region of the AP network using Schiff reagent staining.



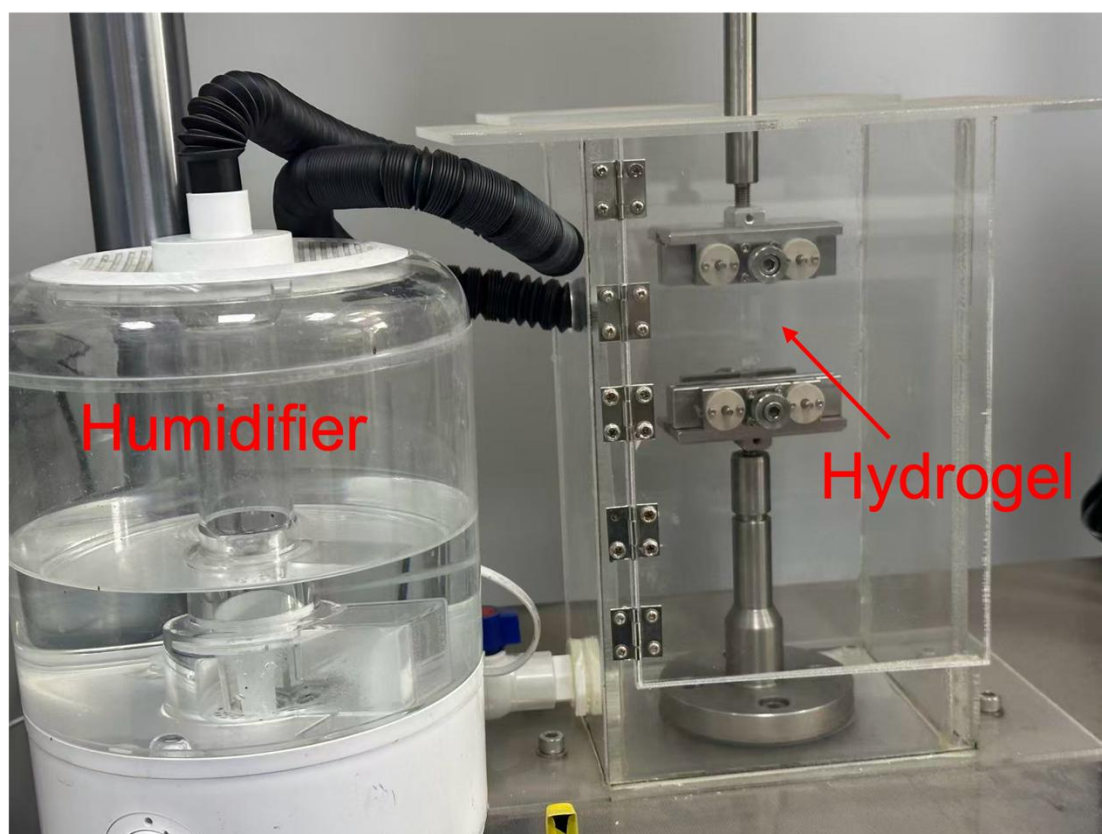
Supplementary Figure 10. Frequency sweep curves of the storage and loss moduli of samples with different compositions.



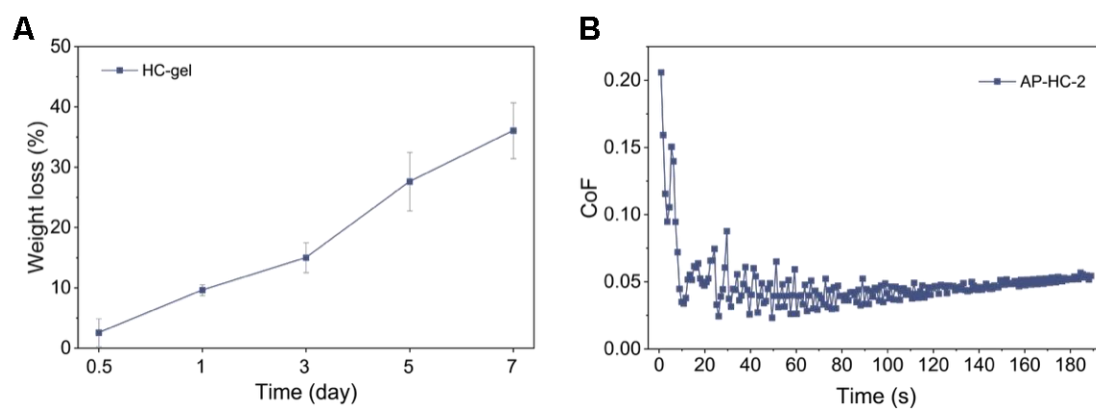
Supplementary Figure 11. Mechanical characterization of AP-HC hydrogels under a humidified environment. (A) Photograph of the tensile testing setup equipped with a humidification device. (B) Uniaxial tensile stress-strain curves of AP-HC-1, AP-HC-2, and AP-HC-3 under humidified conditions. (C) Cyclic tensile stress-strain curves of AP-HC-2 at different strain amplitudes under humidified conditions.



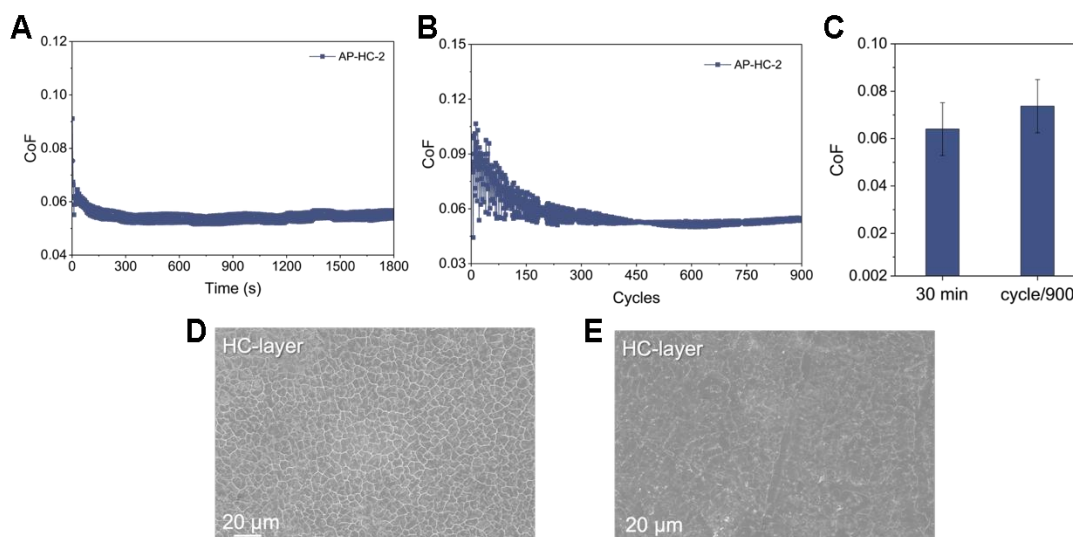
Supplementary Figure 12. Probe tack adhesion performance of the AP-HC Janus hydrogel. (A) Representative force-displacement curve obtained from the probe tack test of AP-HC. (B) Quantified adhesion strength of AP-HC. (C) Comparison of the adhesion strengths of AP-gel and AP-HC hydrogel.



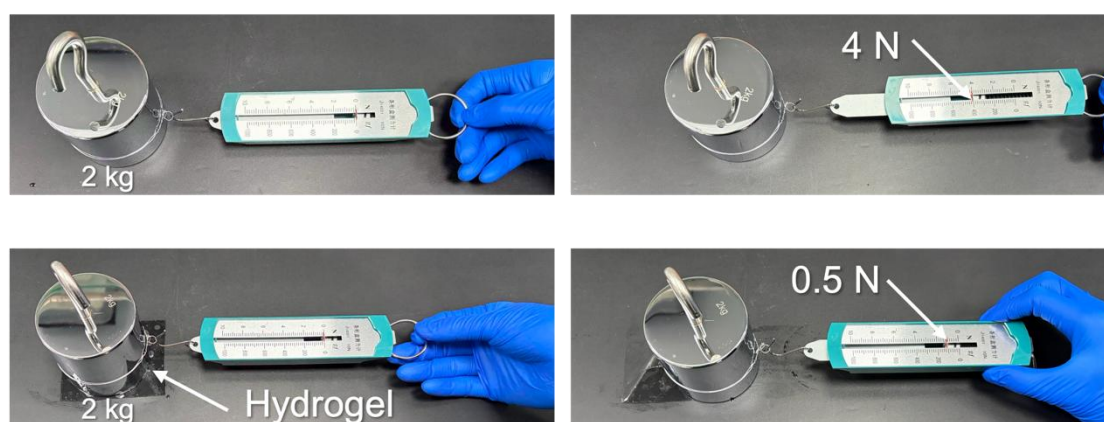
Supplementary Figure 13. Schematic of the setup for 1000-cycle tensile fatigue testing on a fatigue testing machine.



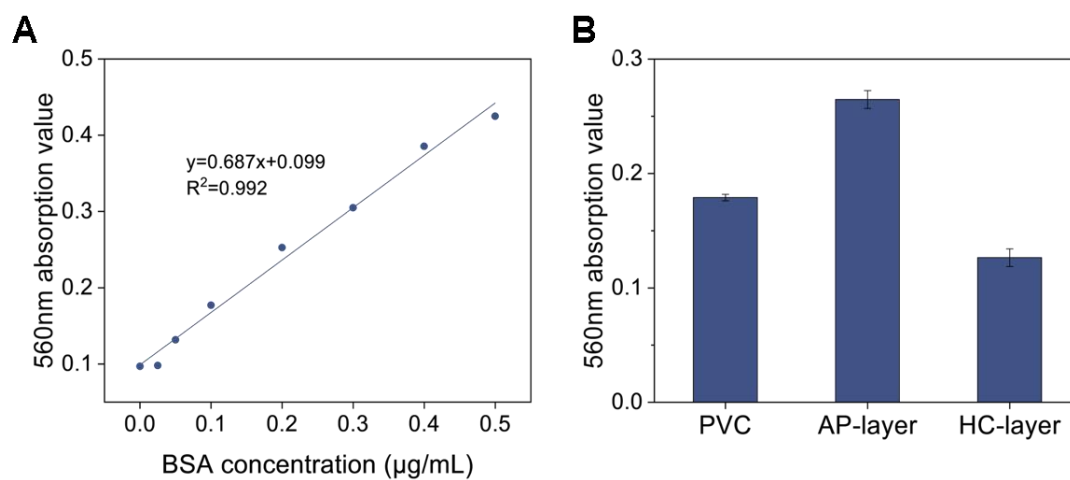
Supplementary Figure 14. Degradation and long-term lubrication performance of the hydrogels. (A) Degradation behavior of the HC gel in simulated body fluid at 37 °C. (B) Friction curve of the AP-HC hydrogel after immersion in simulated body fluid for 48 h.



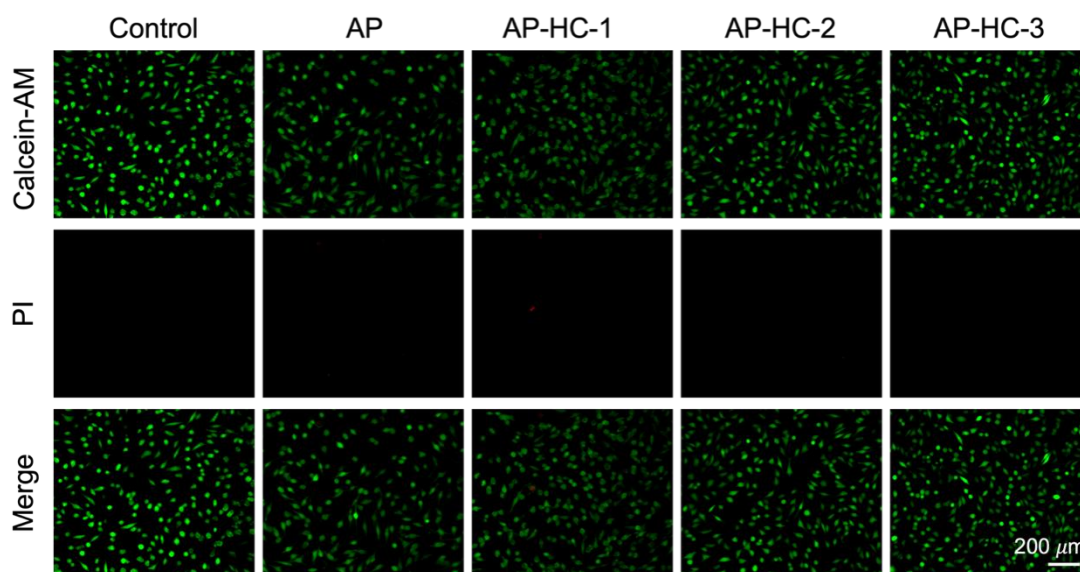
Supplementary Figure 15. Long-term friction performance of the hydrogel. Coefficient of friction measured (A) over 30 min and (B) during 900 friction cycles. (C) Statistical comparison of the coefficients of friction obtained after 30 min and 900 cycles. SEM images of the HC side (D) before friction and (E) after 30 min of friction.



Supplementary Figure 16. Demonstration of the lubrication performance of the AP-HC lubricating layer through weight-dragging tests on a desktop and an AP-HC-coated surface.



Supplementary Figure 17. Anti-Protein-Adhesion Assay (A) Standard curve of BSA determined by the BCA protein assay. (B) Optical density (OD) values of adsorbed proteins on PVC, AP-layer, and HC-layer samples after BSA incubation



Supplementary Figure 18. Calcein AM/PI fluorescence staining images and cell viability of L929 cells after co-culture with AP-HC.