

Light-Regulated Biomaterial Interfaces Enable Flexible Neural Bioelectronics with Stable Adhesion and On-Demand Detachment for Chronic Electrophysiology

Zili Li^{1,2}, Binbin Hu¹, Quansheng He¹, Zhongzhong Wang¹, Liting Sun¹, Haoyi Yang¹, Xiaoxue Zhang¹, Hui Guo³, Yousheng Shu¹, Miao Tang^{1*}

¹Department of Pain Management, Huadong Hospital, State Key Laboratory of Brain Function and Disorders, Institute for Translational Brain Research, MOE Frontiers Center for Brain Science, Fudan University, Shanghai 200032, China

²College of Future Information Technology, Fudan University, 2005 Songhu Road, Shanghai, 200438, China

³Department of Neurosurgery, Shanghai Deji Hospital-The 9th Clinical College, Qingdao University, Shanghai, China

Supplementary Video S1. Conformal adhesion of the bPDMS@Fe film to human skin. The bPDMS@Fe film conforms closely to the skin surface and remains attached during gentle manual peeling, demonstrating stable adhesion under dry conditions.

Supplementary Video S2. UV-triggered detachment of the bPDMS@Fe film from human skin. Following UV irradiation at 365 nm, the bPDMS@Fe film can be readily detached from the skin without macroscopically visible residue, demonstrating on-demand light-triggered release.

Supplementary Video S3. Synchronized behavioral video and LFP recording during non-ictal activity. Representative synchronized recording of a freely moving mouse and the corresponding nine-channel LFP traces during behavior without electrographic seizure activity.

Supplementary Video S4. Synchronized behavioral video and LFP recording during 4-AP-induced seizure. Representative synchronized video and nine-channel LFP recording following intracortical administration of 4-AP, showing the onset and progression of electrographic seizure activity, associated convulsive movements, and subsequent seizure termination.

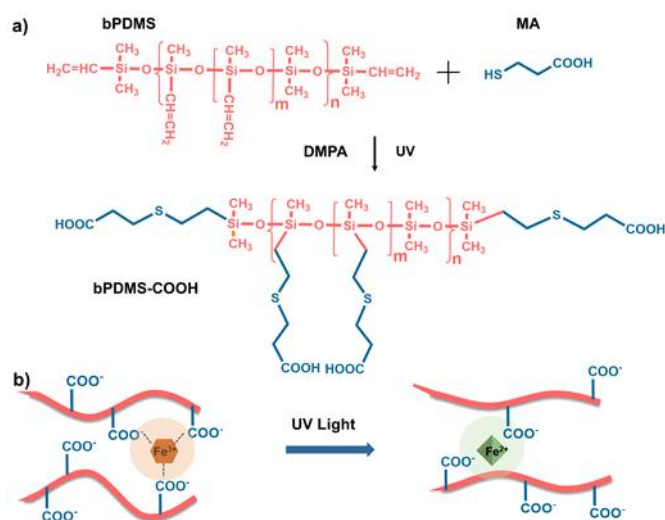


Figure S1. Synthesis and UV-responsive Fe³⁺/Fe²⁺ coordination mechanism of bPDMS@Fe. (A) Synthetic route used to prepare carboxyl-functionalized branched polydimethylsiloxane (bPDMS-COOH) through the thiol-ene addition of 3-mercaptopropionic acid and vinyl-containing PDMS. The red structural segments represent the PDMS backbone, whereas the blue structural segments represent the grafted carboxyl-containing thioether groups. (B) Schematic illustration of the UV-induced transition of the coordination network. The orange circle represents Fe³⁺-carboxylate coordination before irradiation, the green diamond represents Fe²⁺-

associated coordination after photoreduction, the red curved lines represent polymer chains, and the blue groups represent carboxylate ligands. The blue arrow indicates the UV-induced conversion from Fe^{3+} to Fe^{2+} .

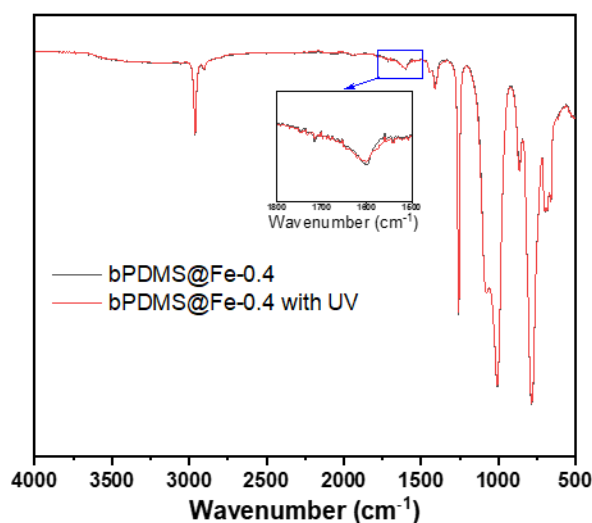


Figure S2. FTIR spectra of bPDMS@Fe before (black curve) and after (red curve) UV irradiation. UV irradiation weakens carboxylate coordination in the bPDMS@Fe network. The inset shows an enlarged view of the carboxylate-related band region. The reduced asymmetric carboxylate stretching band near 1603 cm^{-1} after irradiation is consistent with weakened Fe^{3+} -carboxylate coordination. The suffix 0.4 denotes an $\text{Fe}^{3+}/\text{COOH}$ molar feed ratio of 0.4.

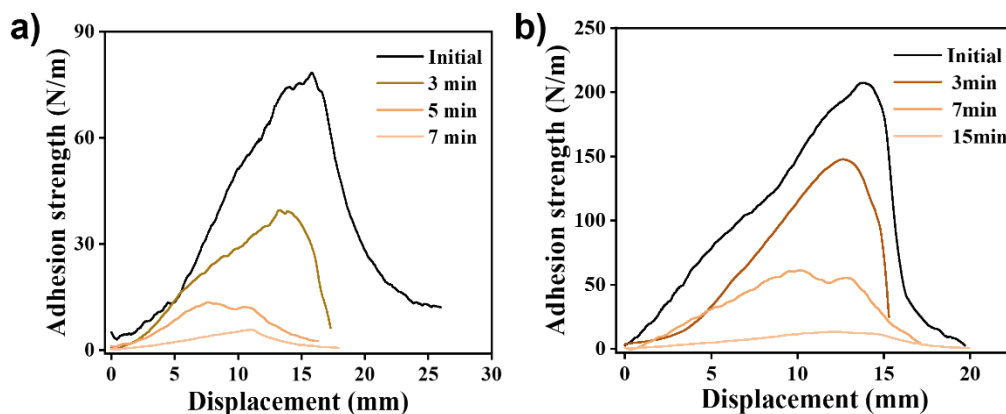


Figure S3. UV irradiation progressively decreases the peel adhesion of bPDMS@Fe on copper. (a) Peel adhesion strength-displacement curves of bPDMS@Fe-0.05 before UV irradiation (black curve) and after irradiation for 3, 5, and 7 min (progressively lighter orange curves). (b) Peel adhesion strength-displacement curves of bPDMS@Fe-0.2 before UV irradiation (black curve) and after irradiation for 3, 7, and 15 min (progressively lighter orange curves). The suffixes 0.05 and 0.2 denote the corresponding $\text{Fe}^{3+}/\text{COOH}$ molar feed ratios. UV, ultraviolet.

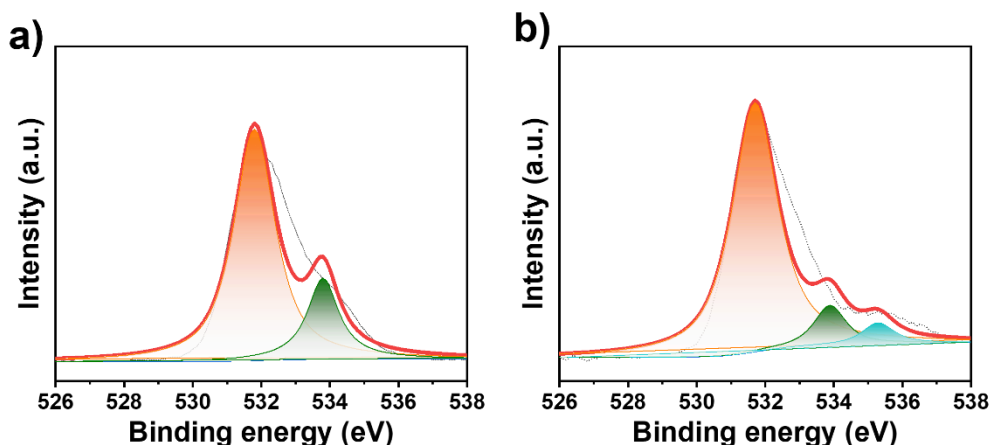


Figure S4. UV irradiation alters the O1s chemical states of bPDMS@Fe. High-resolution O1s XPS spectra of bPDMS@Fe (a) before and (b) after UV irradiation. The black traces represent the experimental spectra, the red curves represent the overall fitted spectra, and the colored shaded peaks represent the individual fitted O1s components. The peaks at 531.8 and 533.6 eV are assigned to Si-O/C=O and C-O species, respectively.

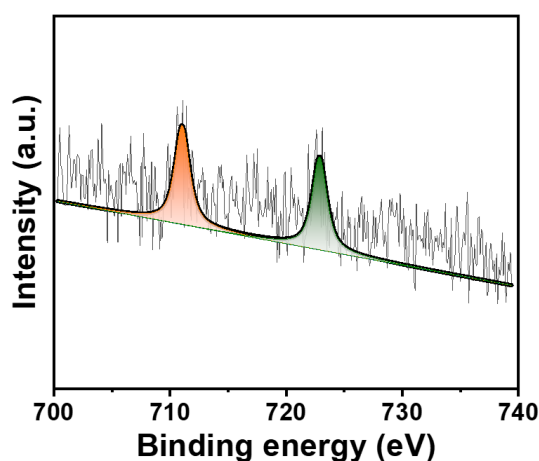


Figure S5. Fe2p spectra of bPDMS@Fe before UV irradiation. Fe^{3+} dominates the Fe2p spectrum of bPDMS@Fe before UV irradiation. High-resolution Fe2p XPS spectrum of bPDMS@Fe before UV irradiation. The black trace represents the experimental spectrum, the orange and green shaded peaks represent the fitted $\text{Fe}2p_{3/2}$ and $\text{Fe}2p_{1/2}$ components, respectively, and the dark green line represents the fitted background. The characteristic Fe^{3+} contribution is centered near 710.4 eV; the Fe^{2+} contribution near 708.9 eV becomes evident after UV irradiation, as shown in Figure 2I.

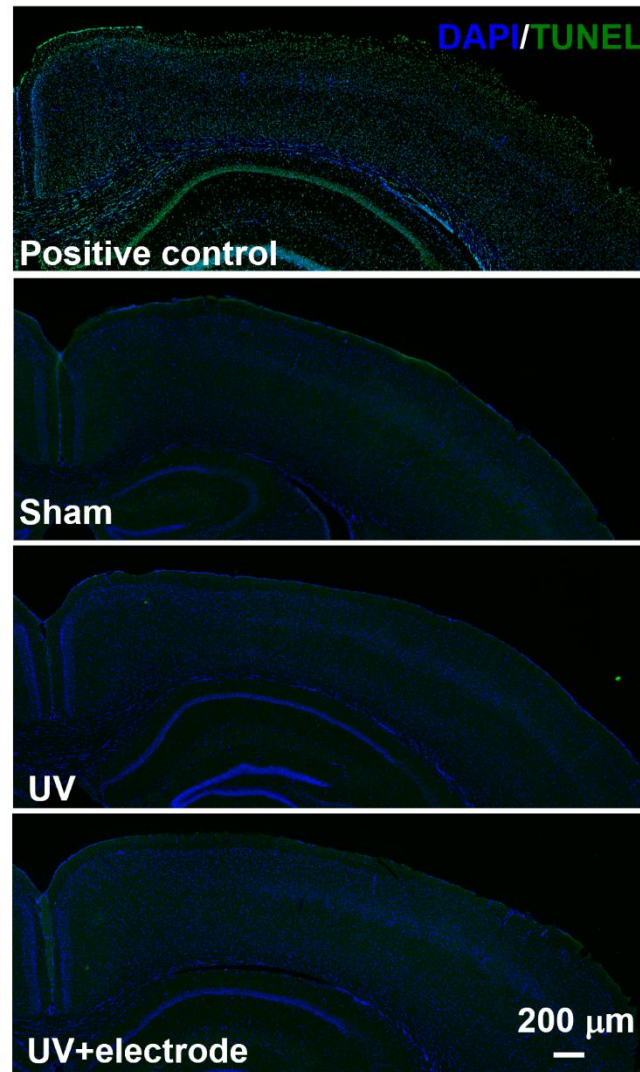


Figure S6. Representative TUNEL staining images of cortical tissue from the positive control and the sham, direct UV exposure (UV), and electrode placement followed by UV-assisted removal (UV+electrode) groups. Tissue collection was initiated 1 h after the corresponding treatment. TUNEL labeling is shown in green, and DAPI-stained nuclei are shown in blue. A positive-control section was included to verify TUNEL labeling. Three mice were examined per experimental group. Scale bar: 200 μm .

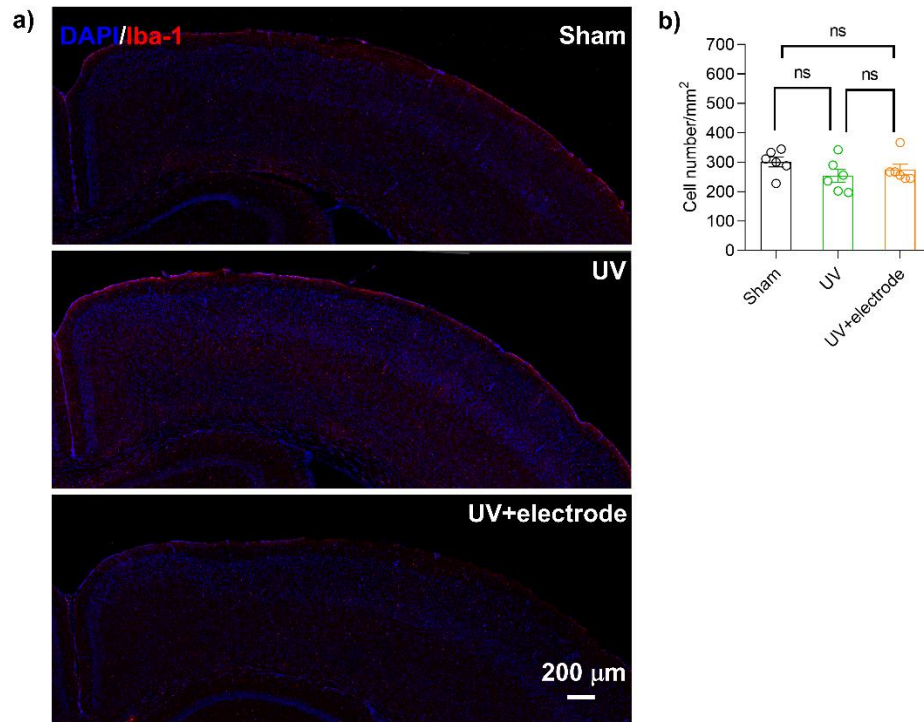


Figure S7. Direct UV exposure and UV-assisted electrode removal do not induce an acute increase in cortical microglial density. (a) Representative immunofluorescence images of cortical tissue from the sham, direct UV exposure (UV), and electrode placement followed by UV-assisted removal (UV+electrode) groups. Tissue collection was initiated 1 h after the corresponding treatment. Iba-1 (red) labels microglia, and DAPI (blue) stains cell nuclei. Scale bar: 200 μm . (b) Quantitative analysis of Iba-1-positive cell density in the cortical region of interest. Three mice were examined per group. In (b), the black-, green-, and orange-outlined bars represent the sham, UV, and UV+electrode groups, respectively. Open circles represent the individual plotted measurements, and bars and error bars show the mean $\pm$ SD.

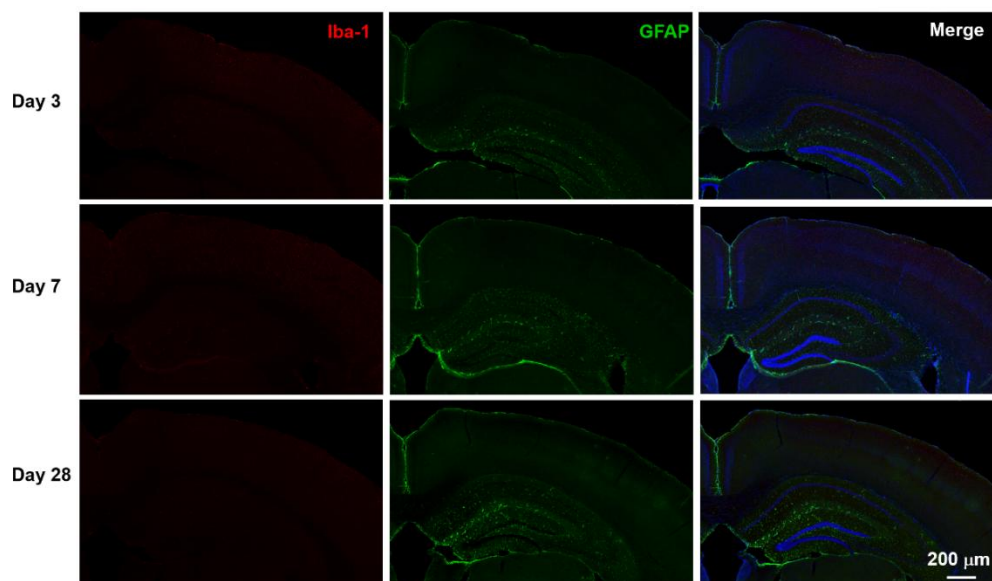


Figure S8. Representative immunofluorescence images of brain tissue sections collected at 3, 7, and 28 days after placement of Fe-free bPDMS-COOH samples. Iba-1 (red) labels microglia/macrophages, GFAP (green) labels astrocytes, and DAPI (blue) stains cell nuclei. Merged images are shown in the right column. Scale bar: 200 μm .

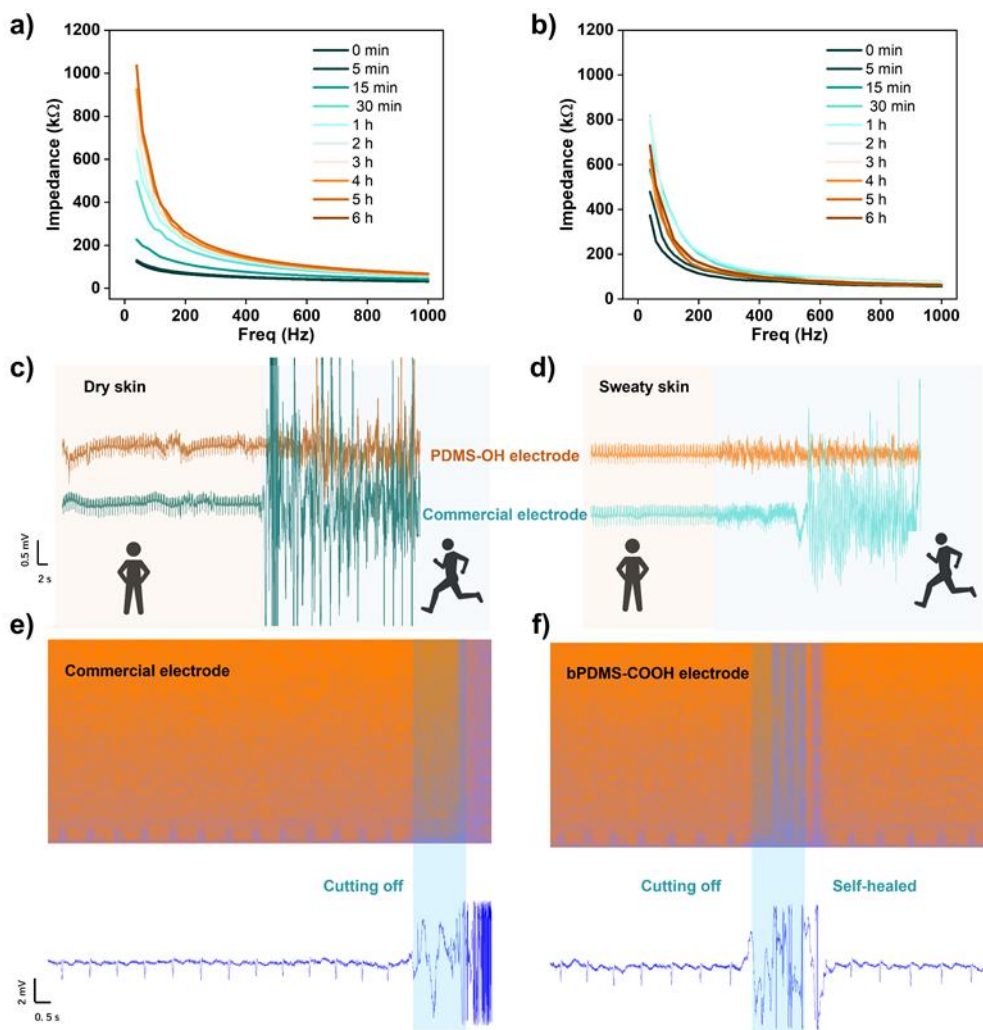


Figure S9. The bio-interfacing performance of the bPDMS@Fe adhesive electrode. The impedance of the (a) commercial electrodes and (b) bPDMS@Fe elastomer-based electrode attached to the skin for different durations. The ECG waveforms of people in rest and exercise states with a commercial electrode and the bPDMS@Fe elastomer-based electrode after attachment to the skin under (c) dry skin and (d) sweaty skin conditions. Representative ECG signals recorded with (e) a commercial electrode

before and after deliberate cutting and (f) a self-healing bPDMS@Fe elastomer-based electrode.

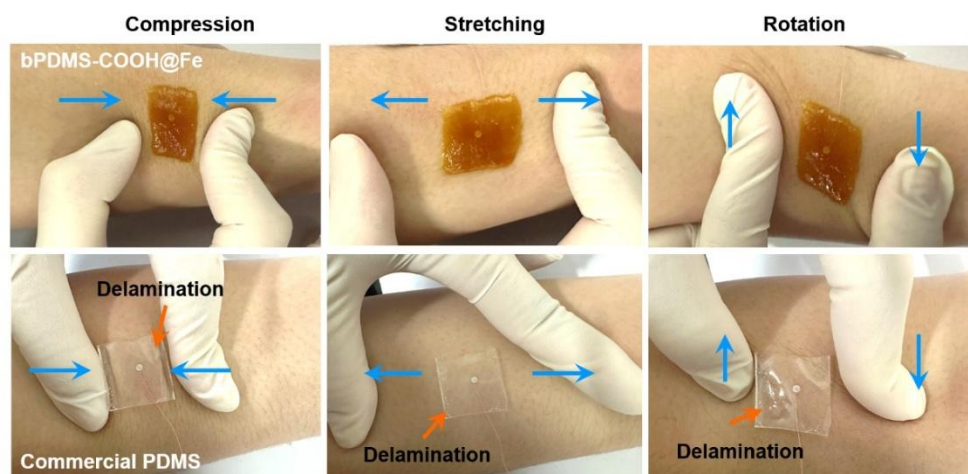


Figure S10. Dynamic adhesion stability of the bPDMS@Fe electrode (upper panel) and commercial electrode (lower panel) on human skin under various mechanical deformations. The bPDMS@Fe electrode maintains intimate and conformal contact with the skin surface without any delamination during compression, stretching, and rotation. In sharp contrast, the commercial PDMS electrode shows obvious edge delamination under the same mechanical deformations, demonstrating the superior dynamic adhesion performance of our material.

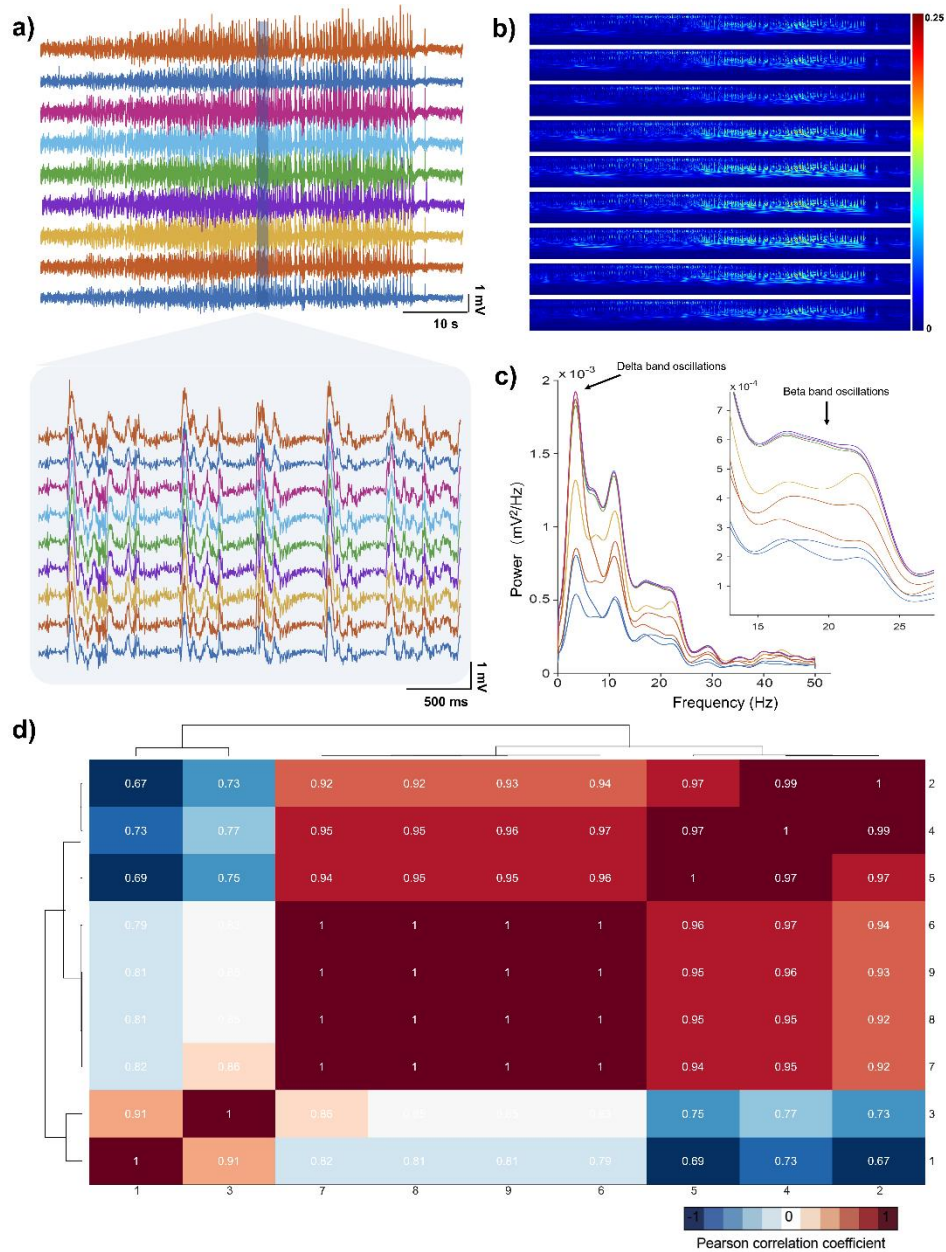


Figure S11. Representative 9-channel LFP recording and epileptic-network analysis in a freely moving mouse after intracortical 4-AP administration. (a) Raw 9-channel LFP traces and magnified ictal segment. (b) Time-frequency spectrograms showing the evolution of seizure-associated oscillatory activity across different channels and different frequency bands. (c) Average power spectral curves highlighting enhanced delta- and beta-band oscillations during ictal activity. (d) Pearson correlation coefficient matrix calculated from the 9-channel LFP signals during the epileptic seizure.

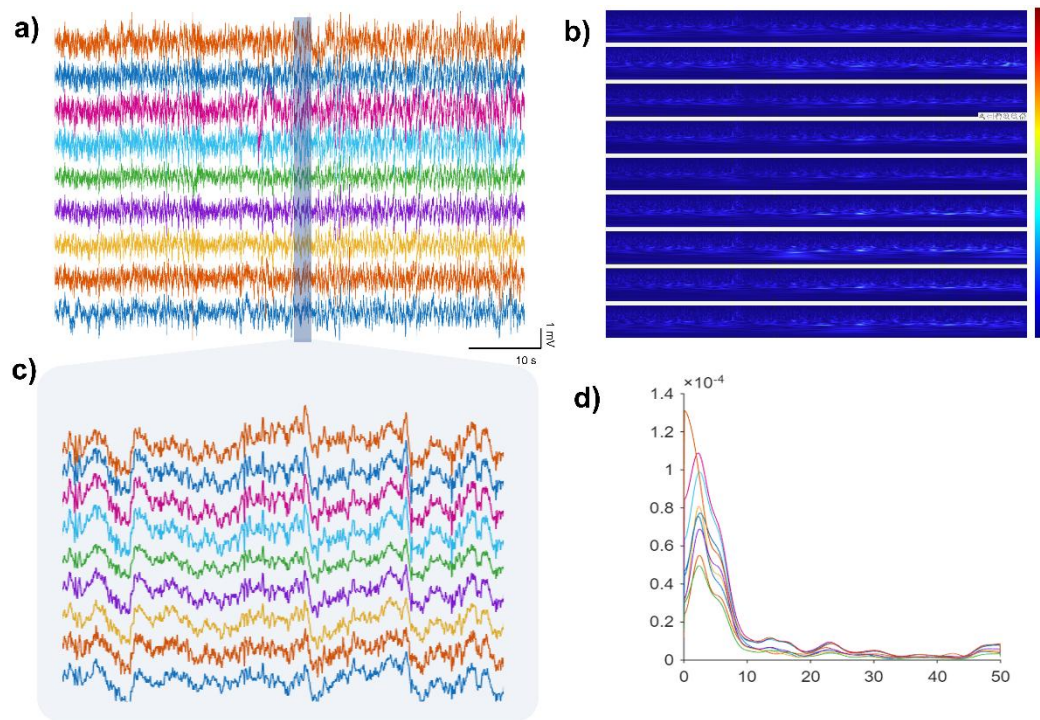


Figure S12. Additional 9-channel LFP recording analysis from freely moving epileptic mice. (a) Representative multichannel raw LFP traces with the analyzed ictal segment highlighted. (b) Time-frequency spectrograms of the corresponding 9-channel LFP signals. (c) Magnified view of synchronized epileptiform discharges within the highlighted ictal window. (d) Average power spectral curves showing seizure-associated spectral enhancement across channels.

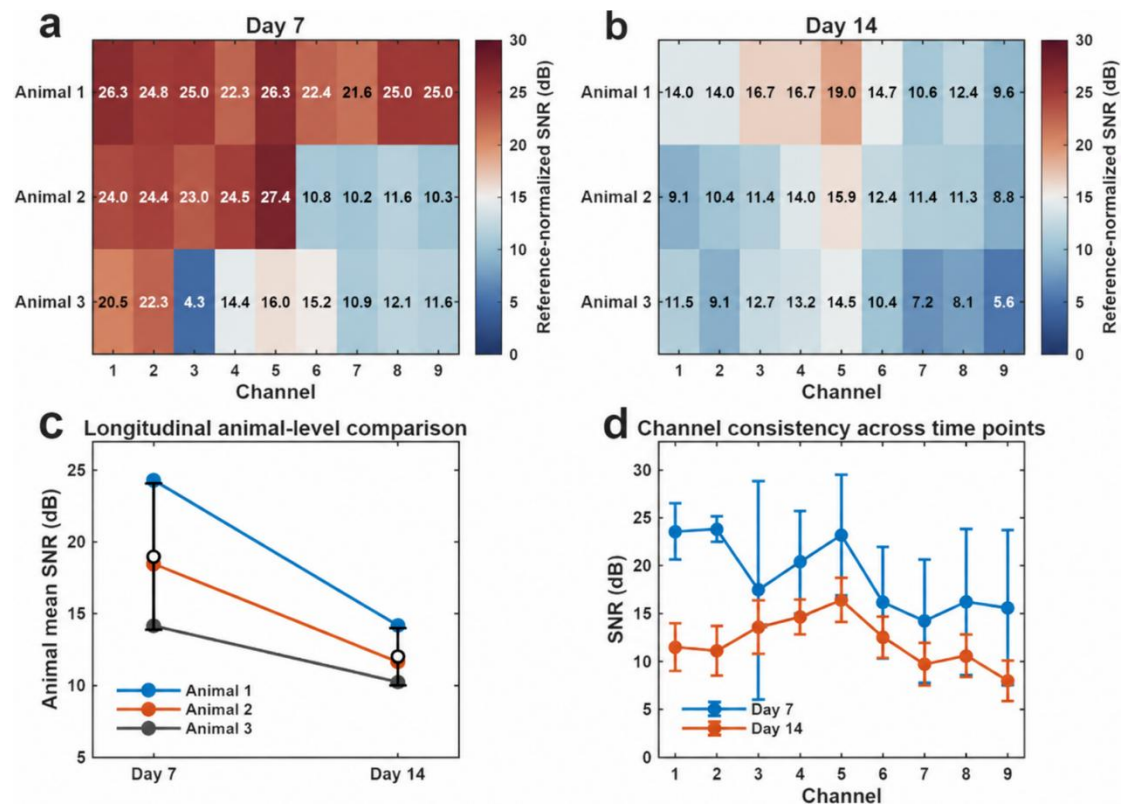


Figure S13. SNR across animals, channels, and time points. Channel-resolved SNR_{ref} at (a) day 7 and (b) day 14. (c) Paired animal-level values and group mean $\pm$ SD. (d) SNR across the three animals for each channel.

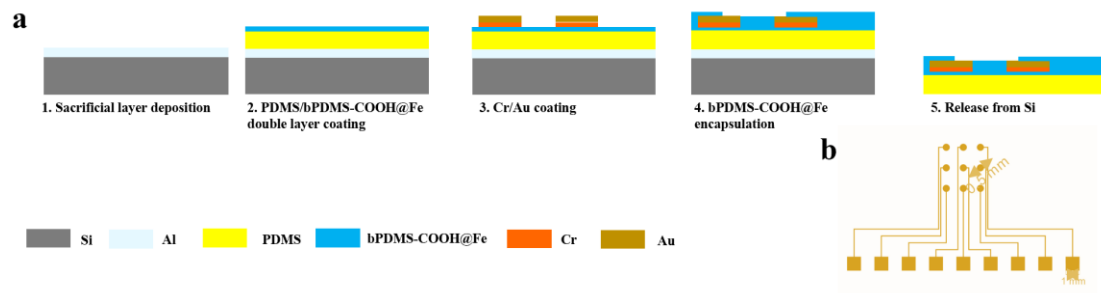


Figure S14. Fabrication and layout of the bPDMS@Fe flexible electrode array. (a) Schematic illustration of the preparation process, including sacrificial-layer deposition, PDMS/bPDMS@Fe coating, Cr/Au patterning, bPDMS@Fe encapsulation, and device release. (b) Top-view layout of the 9-channel Cr/Au electrode pattern.

Table S1. Performance comparison of this work with state-of-the-art bioadhesive neural interfaces and flexible bioelectronics.

Reference	Material system	On-demand debonding	Self-healing	Fluid/sweat stability	Bioelectronic/animal model	Key limitation versus this work
This Work	Hydrophobic elastomer (bPDMS-COOH@Fe)	Yes; UV irradiation	Yes	Excellent; hydrophobic, anti-fluid	Awake freely moving mice; epilepsy LFP mapping	-
REF.1	PAAc-NHS@biopolymer	No	No	Works on wet tissue; water-absorbing/swelling mechanism	Wet tissue/device fixation; rat, porcine, and mouse models	Strong, but not reversible, not electronic, and not self-healing
REF.2	PVA/PAA/NHS	Yes; SBC/GSH solution	No	Wet tissue adhesion; hydrogel after hydration	In vitro tissue sealing/wound closure models	Reversible but chemical-solution-triggered; not a neural/electrode platform
REF.3	PVA/PAA/NHS@GO	Yes; triggering solution	No	Wet dynamic tissues; hydrogel after hydration	In vitro porcine and in vivo rat recording/stimulation	Reversible but chemical-solution-triggered
REF.4	Cationic QDM gel	Yes; voltage	No	Hydrogel/tissue wet environment	In vitro bovine aorta, cornea, lung, and cartilage repair models	Reversible, but requires electrical field; not electrode-integrated neural mapping
REF.5	Electro-oxidized alginate-dopa/PAA	Limited; electrical oxidation	No	Wet tissue adhesion; hydrogel after hydration	In vitro porcine skin	Very strong but not truly on-demand detachable after fixation; not electronic
REF.6	Hydrogel hybrid neural probe	No	No	Long-term implanted hydrogel matrix	Freely moving mice; single-unit neural recording >6 months	Strong neural relevance, but no adhesive/debonding/self-healing function
REF.7	dPEDOT-CA-PDA-PAM conductive bioadhesive hydrogel BMI	Non-destructive removal reported	No	Ultrasoft hydrogel; brain-compatible	ECoG/BMI on brain tissue	Adhesive brain interface, but not reversible detachment and self-healing
REF.8	Non-swelling PAACP hydrogel	Yes; sodium chloride and glutathione	No; regenerable adhesion	Non-swelling hydrogel	Muscle fatigue monitoring; electrode array/strain sensor	Strong anti-swelling hydrogel, but hydrophilic and not neural LFP mapping

solution						
REF.9	Silicone bioadhesive (BioAdheSil)	No	No	Wet tissue adhesion; Good hydrated/PBS stability	Rat model and chest	Hydrophobic/silicone-relevant, but not detachable or neural/electrical
REF.10	CNF-DA/PAA@Fe hydrogel	Yes; UV/photo-Fenton-like process	Not clearly self-healing	Conductive hydrogel; skin electronics	Self-powered e-skin/ human motion monitoring	UV-detachable, but hydrophilic hydrogel and not implantable neural LFP
REF.11	PAAm/cellulose hydrogel	No	No	Excellent on sweating skin	Human EMG on persistent sweating skin	Strong sweat adaptation, but no on-demand debonding/self-healing/neural implant use

References

1. Yuk H, *et al.* Dry double-sided tape for adhesion of wet tissues and devices. *Nature* **575**, 169-174 (2019).
2. Chen X, Yuk H, Wu J, Nabzdyk CS, Zhao X. Instant tough bioadhesive with triggerable benign detachment. *Proc Natl Acad Sci* **117**, 15497-15503 (2020).
3. Deng J, *et al.* Electrical bioadhesive interface for bioelectronics. *Nat Mater* **20**, 229-236 (2021).
4. Borden LK, Gargava A, Raghavan SR. Reversible electroadhesion of hydrogels to animal tissues for suture-less repair of cuts or tears. *Nat Commun* **12**, 4419 (2021).
5. Xue B, *et al.* Hydrogel tapes for fault-tolerant strong wet adhesion. *Nat Commun* **12**, 7156 (2021).
6. Park S, *et al.* Adaptive and multifunctional hydrogel hybrid probes for long-term sensing and modulation of neural activity. *Nat Commun* **12**, 3435 (2021).
7. Wang X, *et al.* Bioadhesive and conductive hydrogel-integrated brain-machine interfaces for conformal and immune-evasive contact with brain tissue. *Matter* **5**, 1204-1223 (2022).

8. Tian G, *et al.* A nonswelling hydrogel with regenerable high wet tissue adhesion for bioelectronics. *Adv Mater* **35**, 2212302 (2023).
9. Singh M, *et al.* A tunable soft silicone bioadhesive for secure anchoring of diverse medical devices to wet biological tissue. *Adv Mater* **36**, 2307288 (2024).
10. Zhang L, *et al.* Cellulose nanofiber-mediated manifold dynamic synergy enabling adhesive and photo-detachable hydrogel for self-powered E-skin. *Nat Commun* **15**, 3859 (2024).
11. Wang S, Wang Z, Zhang L, Song Z, Liu H, Xu X. Sweat-adaptive adhesive hydrogel electronics enabled by dynamic hydrogen bond networks. *Chem Eng J* **492**, 152290 (2024).