

[Supplementary Data]

Hexagonal Metal Complex Based Mechanically Robust Transparent Ultrathin Gold μ ECoG for Electro-Optical Neural Interfaces

Duhee Kim^{1†}, Murali Bissannagari^{1,2,3†}, Boil Kim^{4†}, Nari Hong^{1,2}, Jaew Park⁵, Hyeongtae Lim¹, Junhee Lee¹, Jungha Lee¹, Yoon Kyoung Kim⁴, Youngjae Cho⁶, Kwang Lee⁴, Junghyup Lee¹, Jong-Hyeok Yoon¹, Jae Eun Jang¹, David Tsai⁷, Sanghoon Lee⁵, Hyuk-Jun Kwon¹, Han Kyoung Choe^{4*}, and Hongki Kang^{3,8,9,10*}

¹ Department of Electrical Engineering and Computer Science, Daegu Gyeongbuk Institute of Science and Technology (DGIST), Daegu 42988, Republic of Korea

² Information and Communication Engineering Research Center, Daegu Gyeongbuk Institute of Science and Technology (DGIST), Daegu 42988, Republic of Korea

³ Institute of Medical and Biological Engineering, Medical Research Center, Seoul National University, Seoul 03080, Republic of Korea

⁴ Department of Brain Sciences, Daegu Gyeongbuk Institute of Science and Technology (DGIST), Daegu 42988, Republic of Korea

⁵ Department of Robotics and Mechatronics Engineering, Daegu Gyeongbuk Institute of Science and Technology (DGIST), Daegu 42988, Republic of Korea

⁶ School of Undergraduate Studies, College of Transdisciplinary Studies, Daegu Gyeongbuk Institute of Science and Technology (DGIST), Daegu 42988, Republic of Korea

⁷ Graduate School of Biomedical Engineering, UNSW Sydney, NSW 2052, Australia

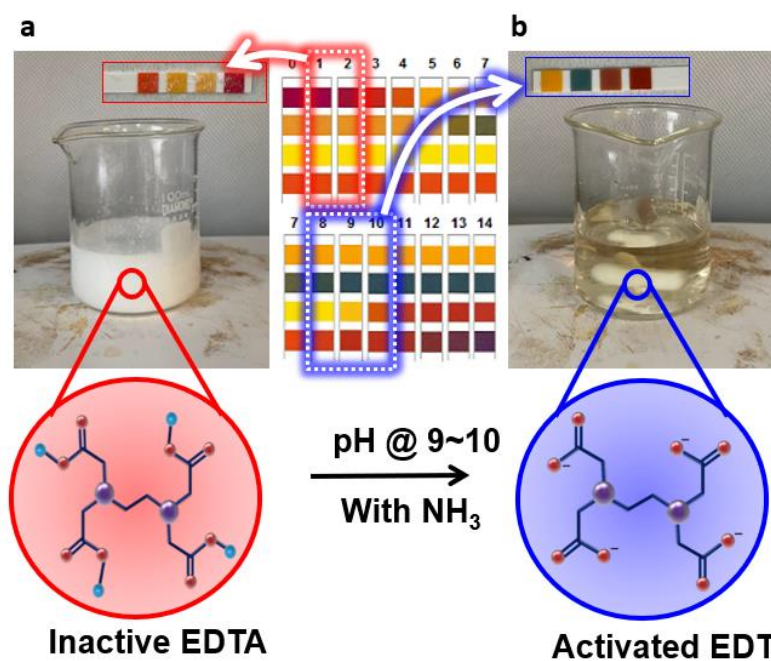
⁸ Department of Biomedical Engineering, Seoul National University College of Medicine, Seoul 03080, Republic of Korea

⁹ Interdisciplinary Program in Bioengineering, College of Engineering, Seoul National University, Seoul 08826, Republic of Korea

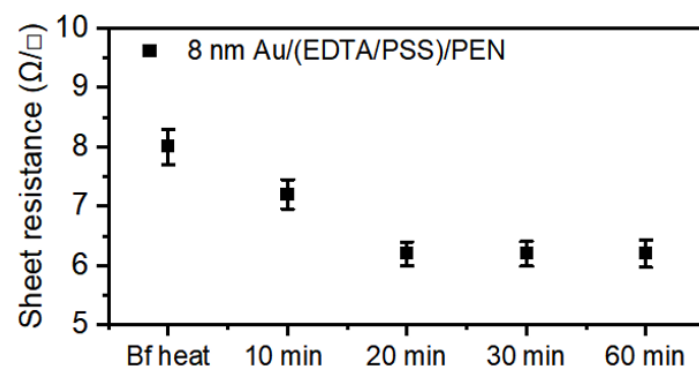
¹⁰ Department of Biomedical Engineering, Seoul National University Hospital, Seoul, 03080, Republic of Korea

[†] These authors contributed equally

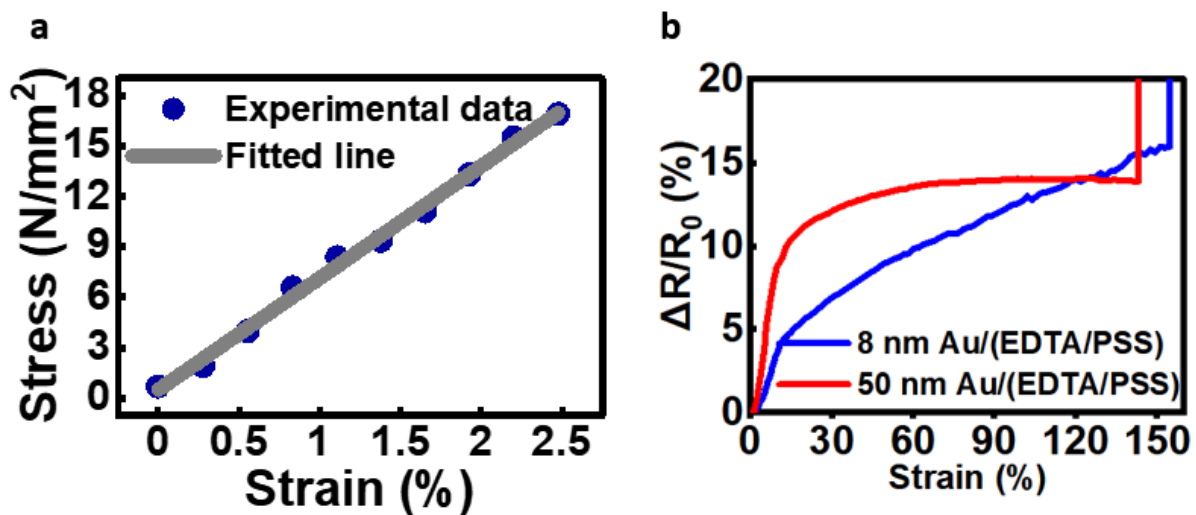
*Correspondence to: choehank@dgist.ac.kr, hongki.kang@snu.ac.kr



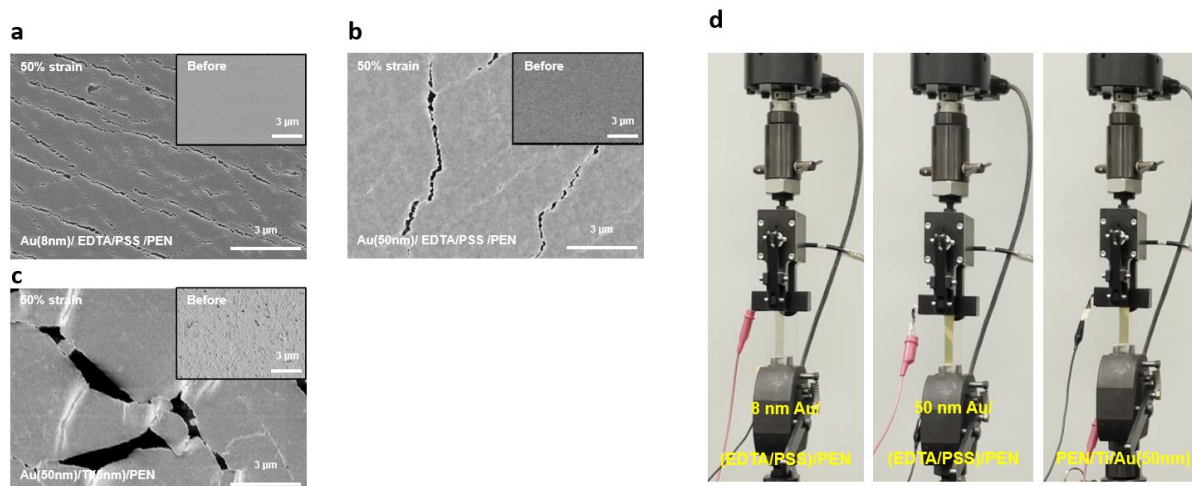
Supplementary Figure 1. The chemical reaction mechanism of EDTA (a) Inactive (deprotonated $[EDTA]^0$. (b) The pH adjusted active (protonated $[EDTA]^{4-}$) chemical structures.



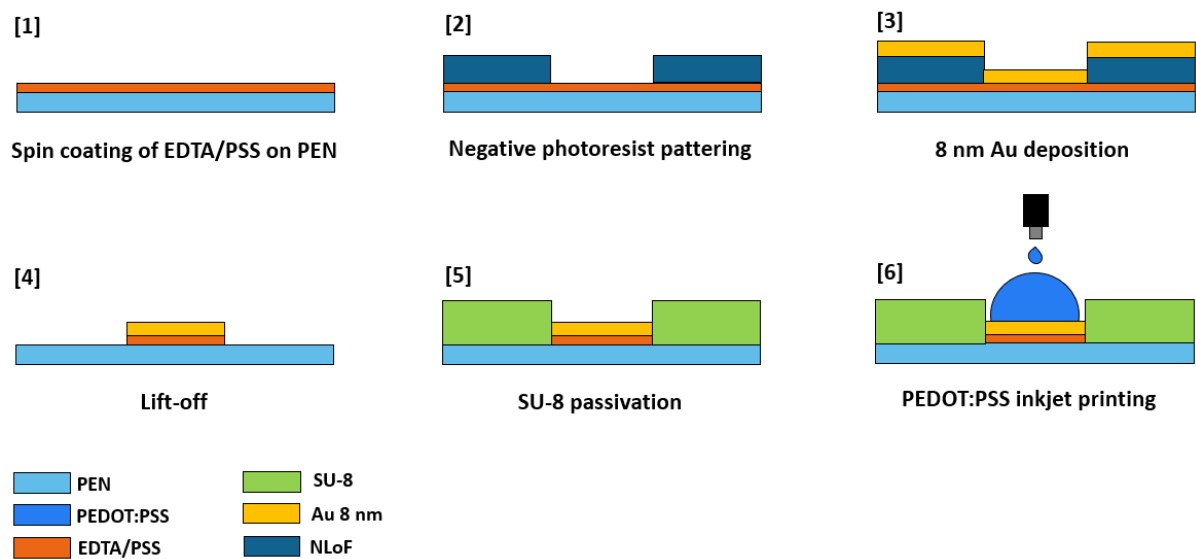
Supplementary Figure 2. Heat treatment induced conductivity optimization of 8 nm Au/(EDTA/PSS)/PEN film.



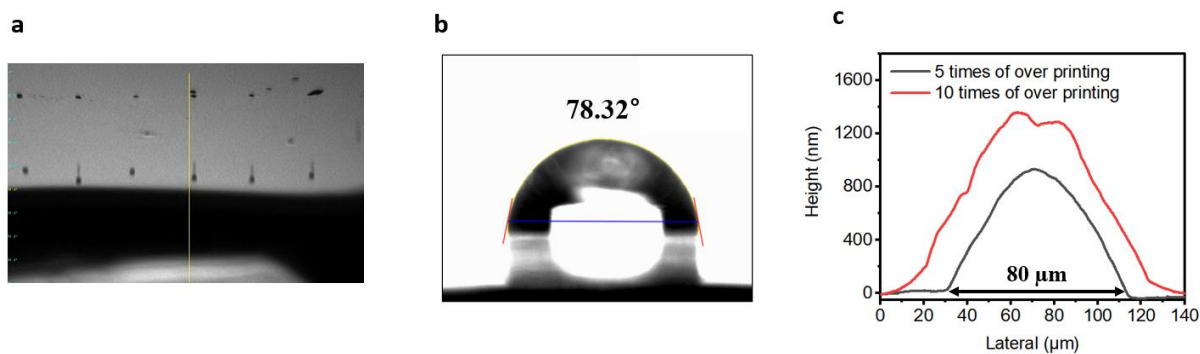
Supplementary Figure 3. Mechanical robustness of the transparent, ultrathin Au film. (a) magnified results of Stress-strain curve and (b) electrical resistance change during the strain test.



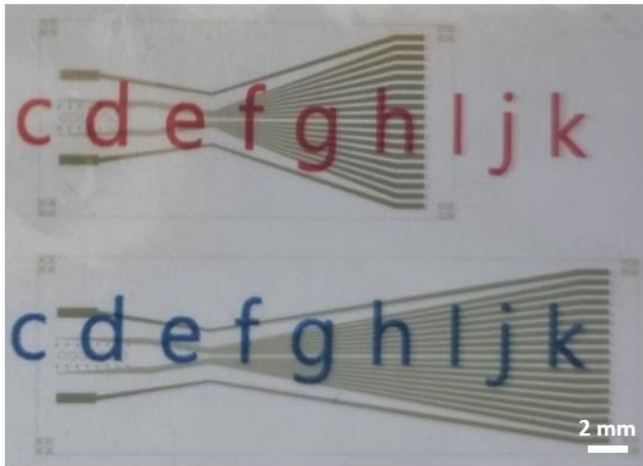
Supplementary Figure 4. SEM images before and after applying 50% tensile strain on the films. (a-c) 8 nm Au/(EDTA/PSS)/PEN, 50 nm Au/(EDTA/PSS)/PEN, and Au/Ti (50 nm/5 nm)/PEN, respectively. (d) strain-stress test setup of each metal film.



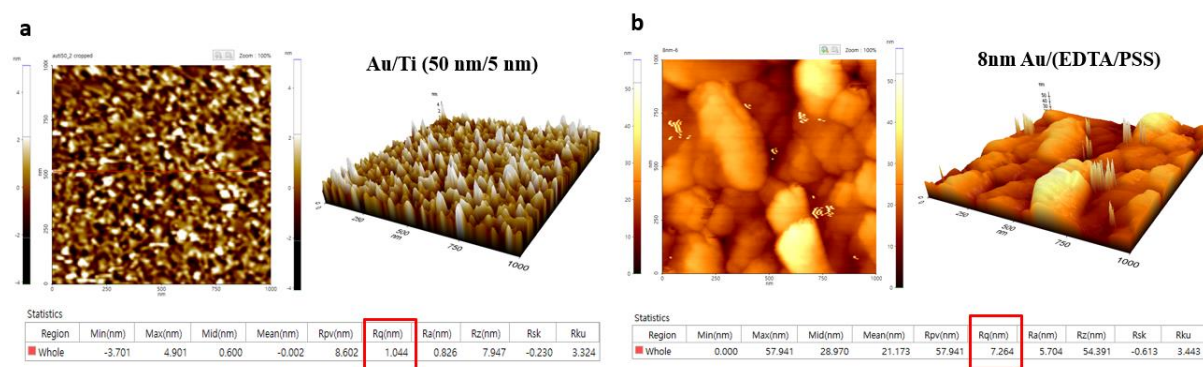
Supplementary Figure 5. Fabrication steps of EDTA seed layer based transparent, low impedance μ ECoG



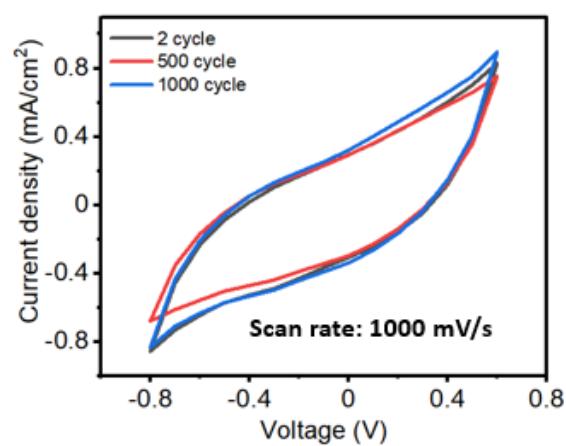
Supplementary Figure 6. Optimization of PEDOT:PSS inkjet printing on μ ECoG interface. (a) Single drop jetting image of PEDOT:PSS ink. **(b)** Contact angle of Au 8nm/EDTA-PSS film with DI drop. **(c)** Vertical morphology of 5 times/10 times inkjet-printed PEDOT:PSS μ ECoG interface.



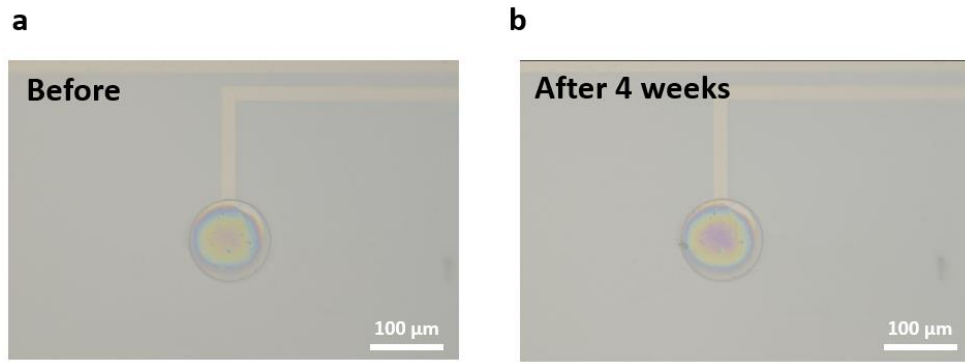
Supplementary Figure 7. Fabrication results of transparent μ ECoG (Top: 1 cm of interconnection line, bottom: 2 cm of interconnection line, Scale bar: 2 mm)



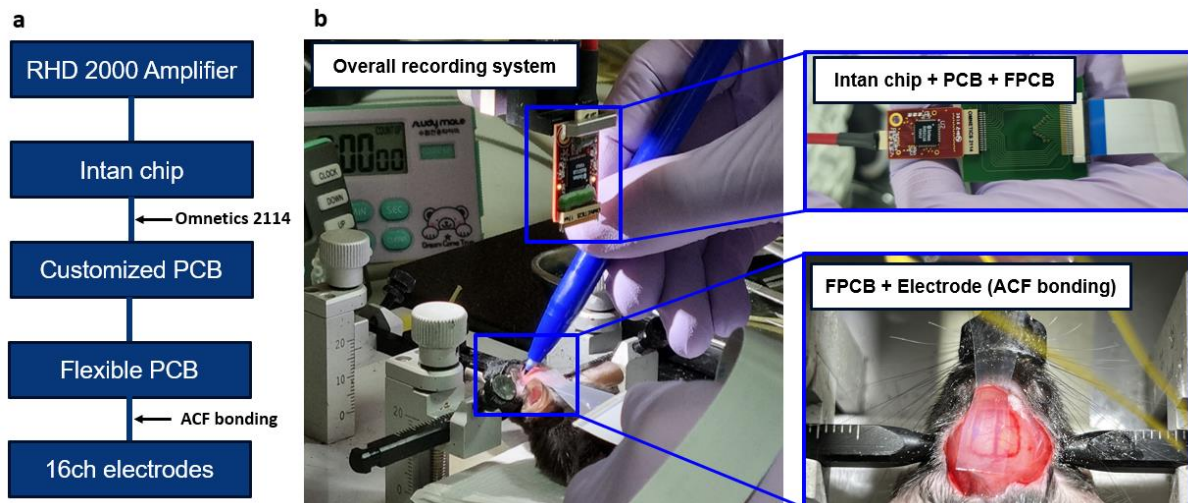
Supplementary Figure 8. AFM scanning results of μ ECoG interface. (a) Au/Ti (50 nm/5 nm) and (b) 8nm Au/EDTA-PSS film. (Sample scanning size: 1 μ m by 1 μ m. 256-bit by 256-bit).



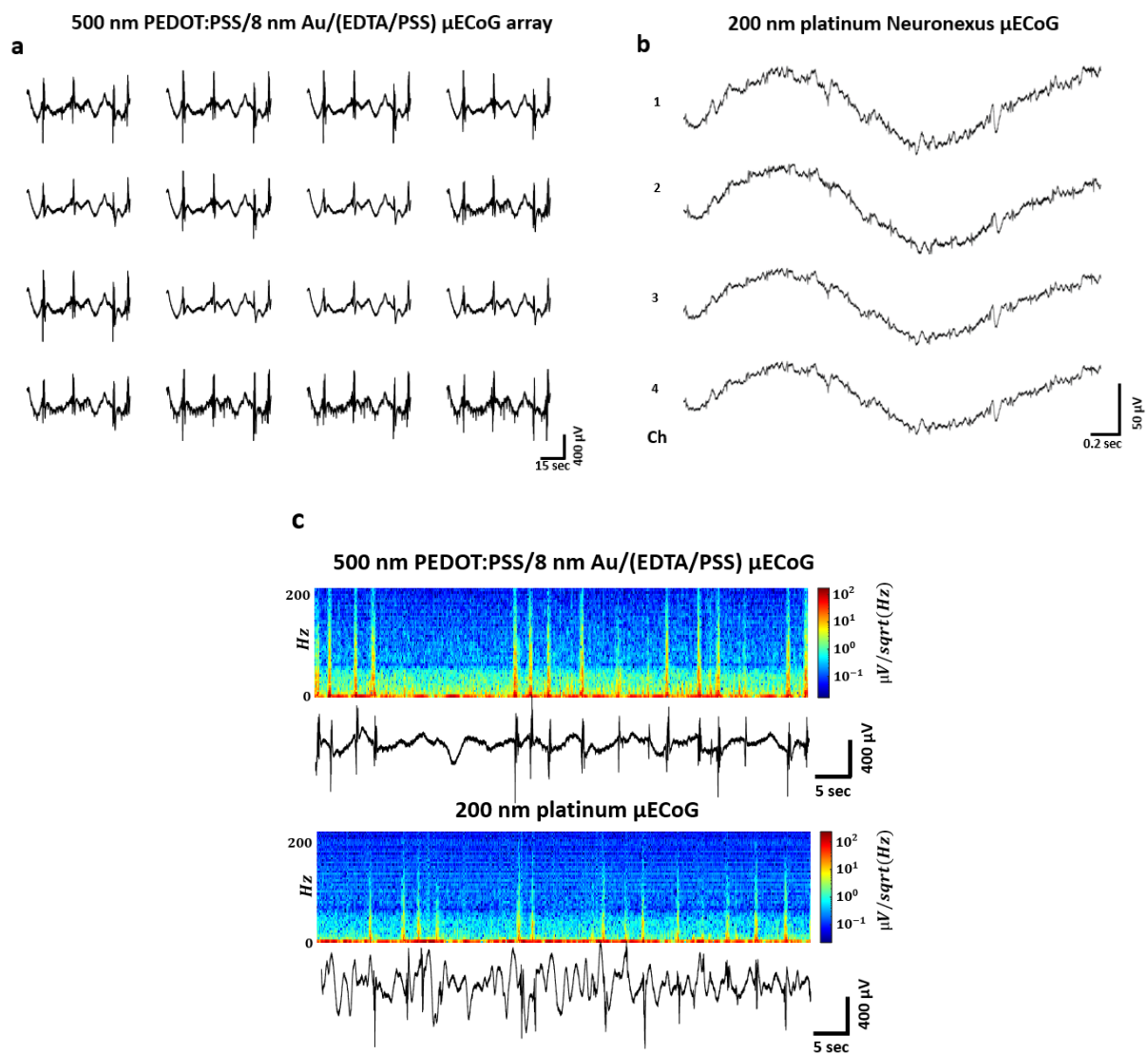
Supplementary Figure 9. Cyclic voltammetry durability test up to 1000 cycles of the 500 nm PEDOT:PSS/8 nm Au/(EDTA/PSS) μ ECoG array at a voltage scan range of -0.8V to 0.6V and a scan rate of 1000 mV/s.



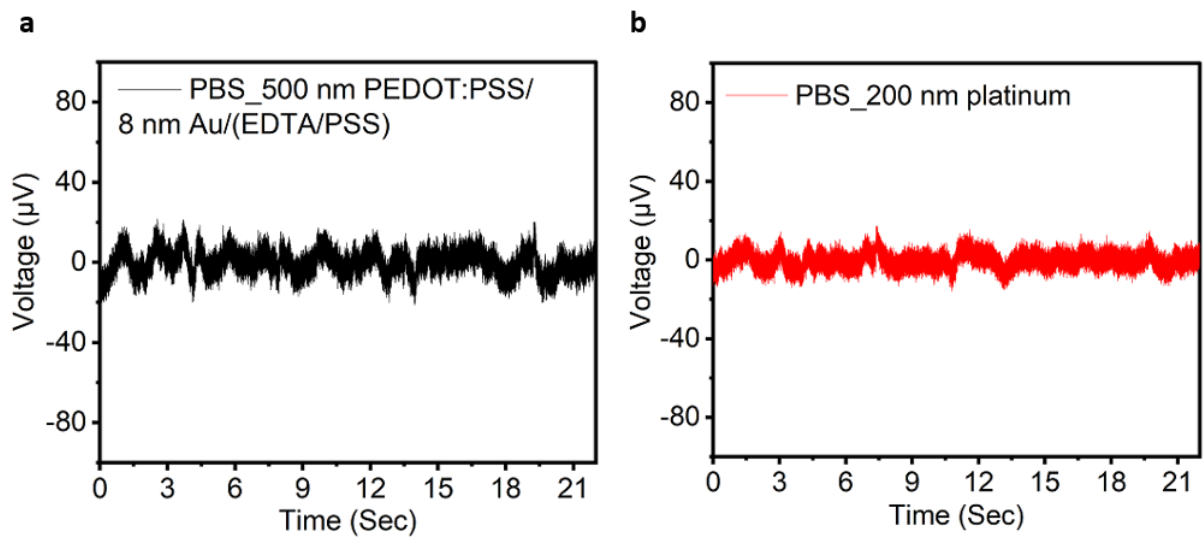
Supplementary Figure 10. Long term durability test of μ ECoG interface. (a) Optical images before immersing the electrode in PBS solution. **(b)** Optical images after immersing the electrode in PBS solution for 4 weeks.



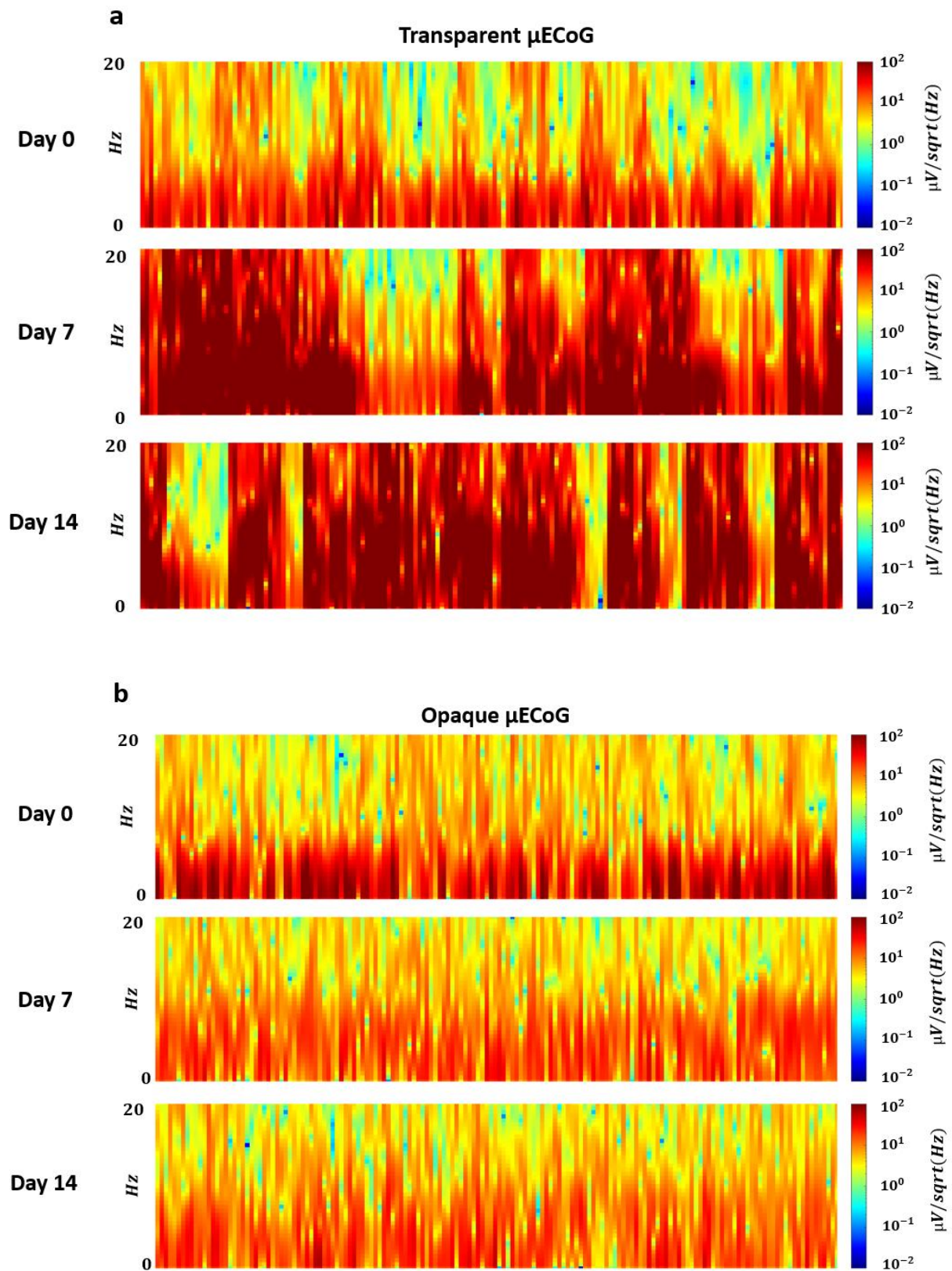
Supplementary Figure 11. *In vivo* neural recording system setup. (a) Schematic of in vivo recording system. **(b)** Specific real images for each part of the *in vivo* recording system.



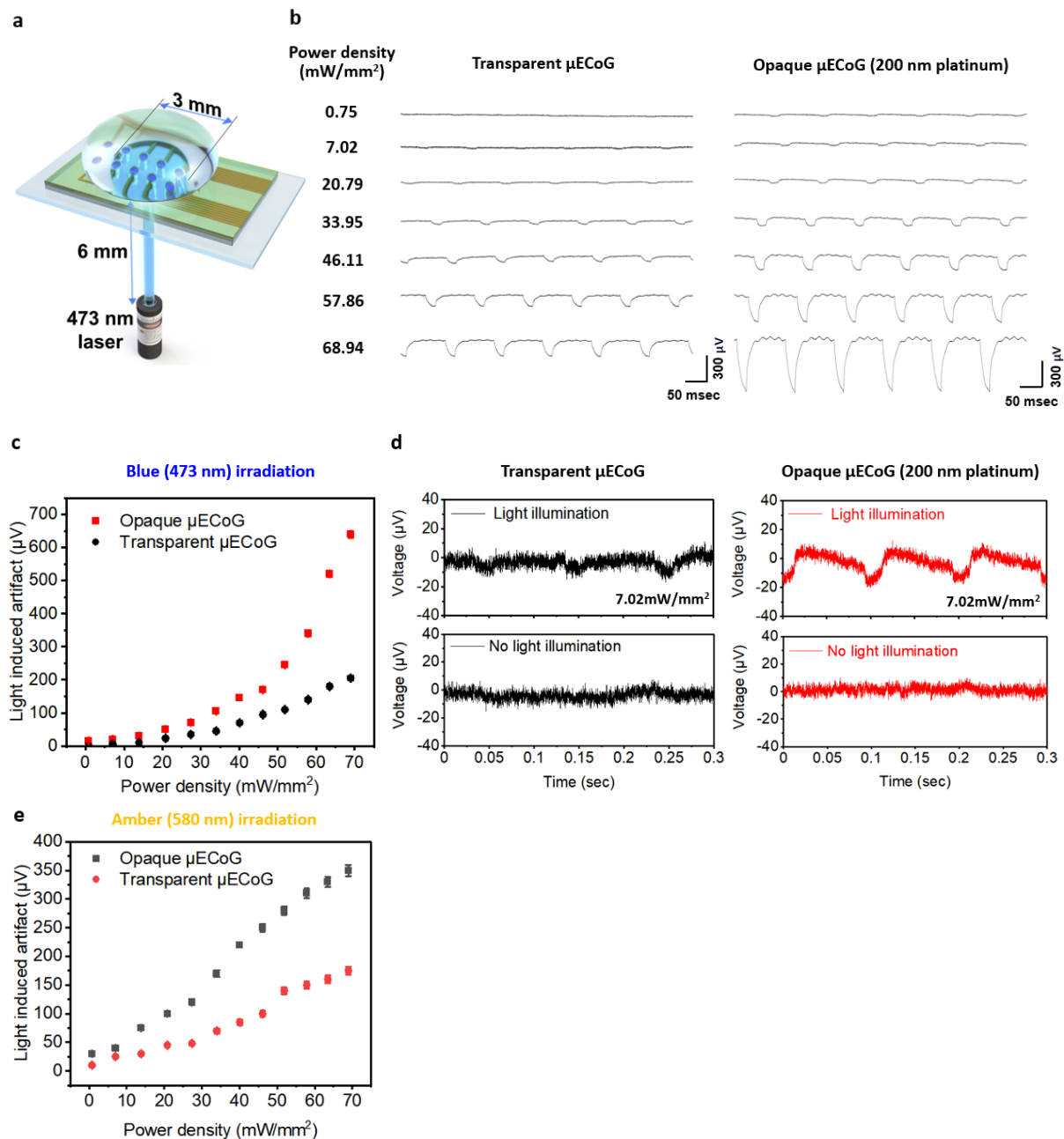
Supplementary Figure 12. *In vivo* neural recordings. (a) μ ECoG neural signals measured from the 16-channel 500 nm PEDOT:PSS/8 nm Au/(EDTA/PSS) μ ECoG array. (b) LFPs in four channels (Ch 1-4) of 200 nm platinum μ ECoG array which were low-pass filtered (200 Hz) from original neural signals. (c) Time-domain analysis of the short-time Fourier transform for each μ ECoG array recording in the 0-200 Hz frequency range.



Supplementary Figure 13. Results of pre-electrical recording in PBS solution. Free-recording results of immersed in PBS solution of **(a)** 500 nm PEDOT:PSS/8 nm Au/(EDTA/PSS) μECoG array and **(b)** platinum 200 nm μECoG array. Both recordings are performed during 22 seconds and there was no spontaneous spikes which we characterized during *in vivo* brain signal recording.

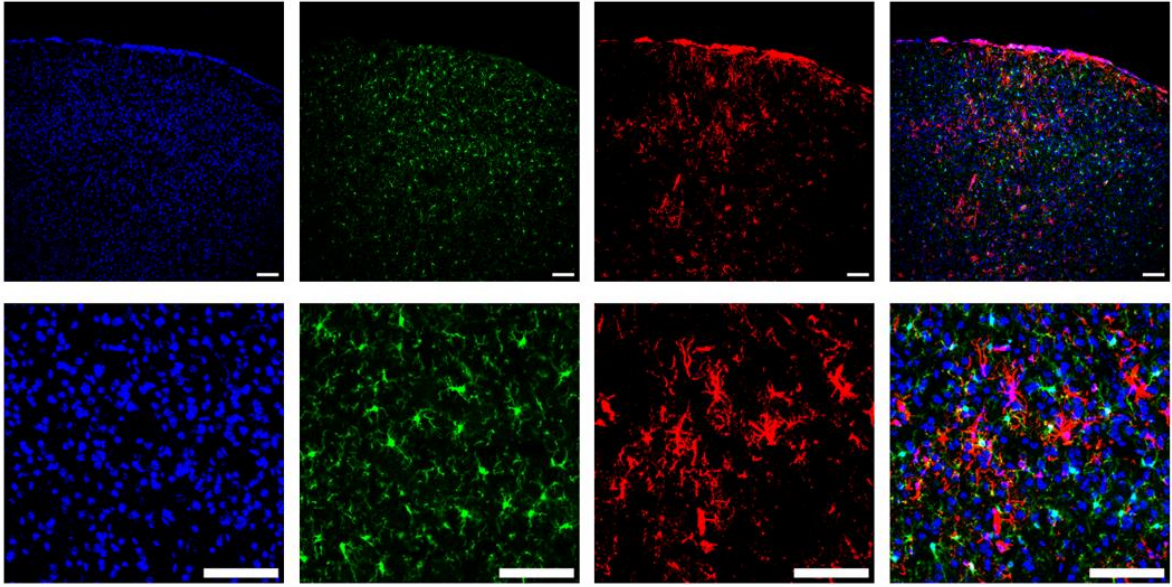


Supplementary Figure 14. PSD results of chronic neural recording. (a) Transparent μ ECoG array and **(b)** opaque μ ECoG array over 14 days, matched with the recorded signal shown in Figure 5g.

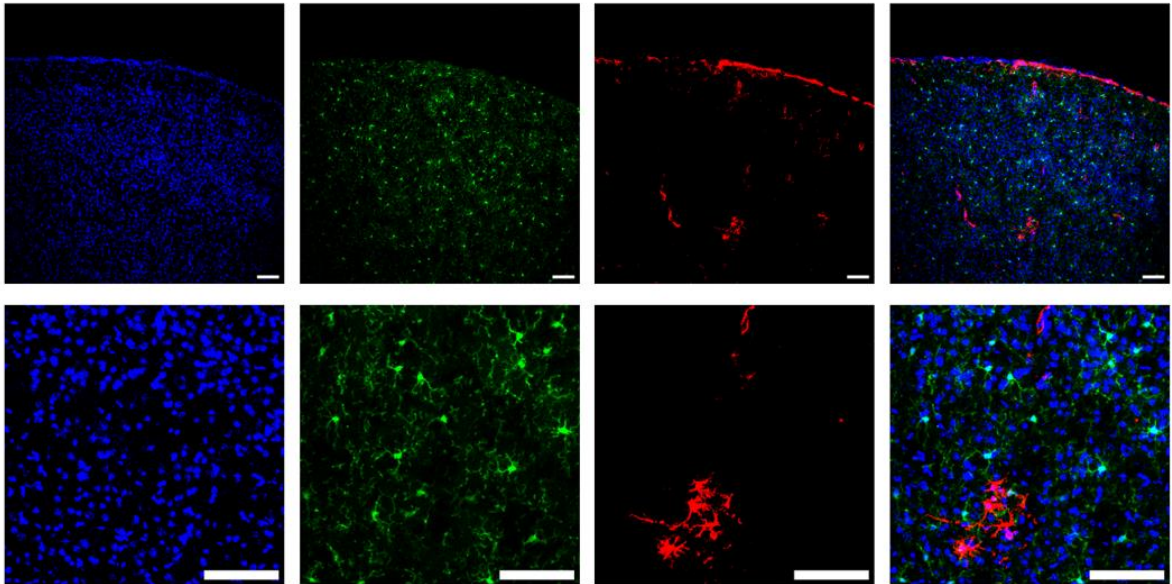


Supplementary Figure 15. Minimized Light-Induced Artifact for Optical Neural Recording and Stimulation in PBS solution. (a) Schematic of light induced artifact measurement setup. The distance from laser tip to electrode was 6 mm, and the diameter of laser was 3 mm. (b) Photoelectric artifacts for a transparent electrode (left) and opaque (right) at various laser power densities. Optical images of commercial, opaque electrodes on mouse brain. Neural recording electrode (right zoom, scale bar: 100 μm). (c) Light induced artifact quantification with blue laser (473 nm) illumination of transparent (red) and opaque (black) electrodes for various power densities. (d) Photoelectric artifacts for illuminated (black) and not illuminated (red) spots at 7.02 mW/mm² power density with repeated on/off (20 s/80 s) pulsed laser. (e) Light induced artifact quantification with amber laser (580 nm) illumination of transparent (red) and opaque (black) electrodes for various power densities.

a Opaque μ ECoG array - implanted cortex



b Opaque μ ECoG array - nonimplanted cortex



Supplementary Figure 16. Comparative immunohistochemical analysis of microglia and astrocyte in implanted and non-implanted Cortices. (a) DAPI, Iba1, and GFAP staining in the implanted cortex with the opaque μ ECoG array, displaying cellular responses in the implanted region. Scale bar = 100 μ m. **(b)** DAPI, Iba1, and GFAP staining in the non-implanted cortex for the opaque μ ECoG array, serving as a comparison to highlight minimal microglial activation and increased astrocyte response in implanted regions. Scale bar = 100 μ m.