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Supplementary Materials for

Mixed-conducting particulate composites for soft electronics

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The PDF file includes:

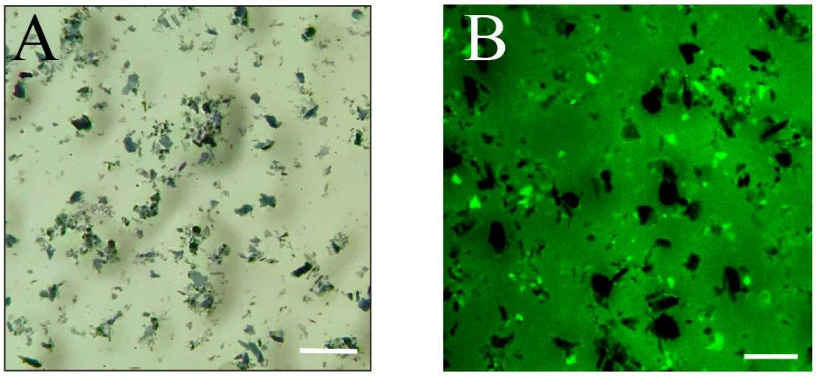
Figs. S1 to S14
Legend for movie S1

Other Supplementary Material for this manuscript includes the following:

(available at advances.sciencemag.org/cgi/content/full/6/17/eaaz6767/DC1)

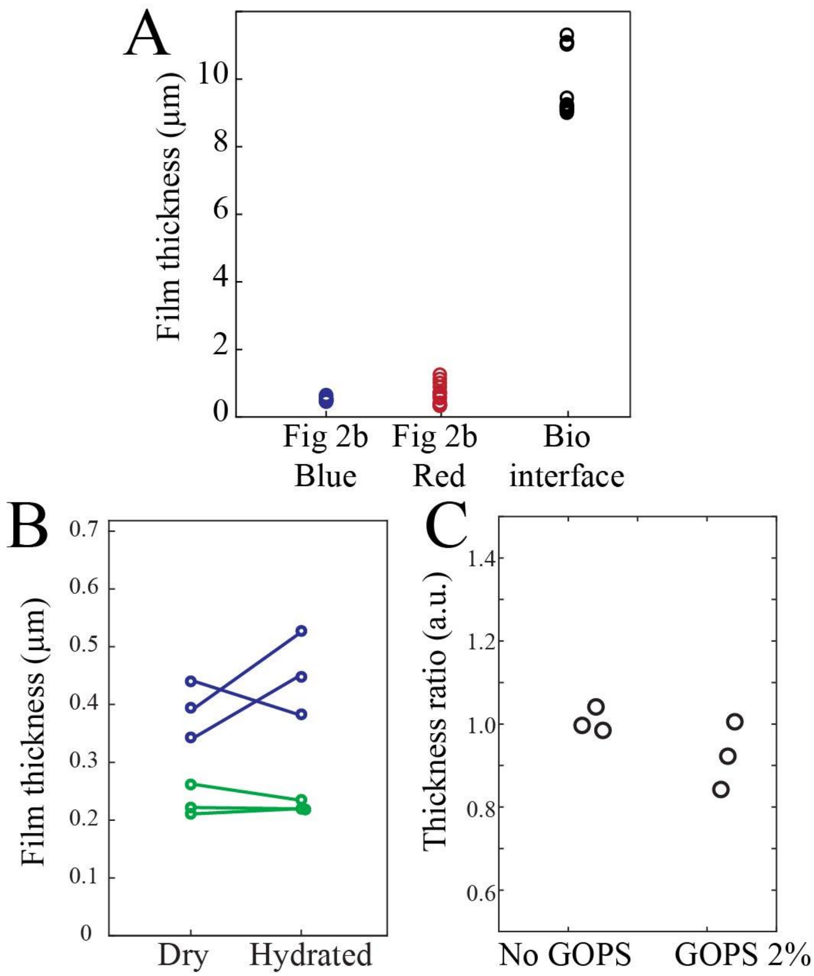
Movie S1 (.mov format)

Supplementary Materials



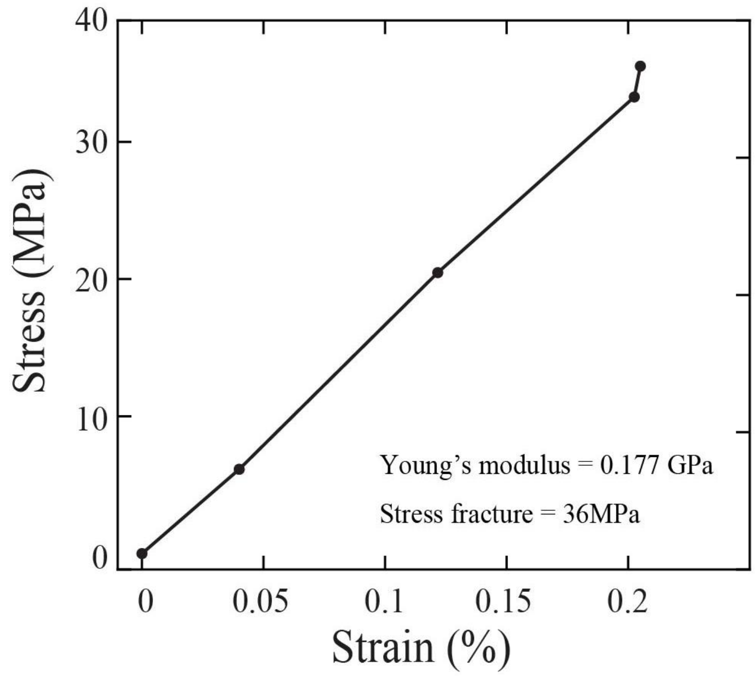
Supplementary Figure 1: Optical properties of MCP.

A. Sample micrograph of blade-coated MCP film (CS/PEDOT:PSS wt:wt ratio 1:3, particle size = 10 – 40 µm, 200 µm blade gap). Scale bar = 100 µm.
B. Fluorescence micrograph of MCP using a green excitation fluorescence filter. Scale bar = 100 µm.



Supplementary Figure 2: Consistency of MCP film thickness and aqueous stability.

A. Film thickness is consistent for various MCP formulations. "Bio-interface" corresponds to thickness of film used in ECG or EMG measurements.
B. Film thickness for CS (green) and CS+2% v/v GOPS (blue) before and after 10 s of hydration in DI water.
C. Thickness ratio for CS and CS+2% v/v GOPS. [Ratio = (thickness after hydration)/(thickness before hydration)]



Supplementary Figure 3: Stress vs strain for MCP CS+PVA film (thickness = 70 um, area under strain = 35 mm^2).

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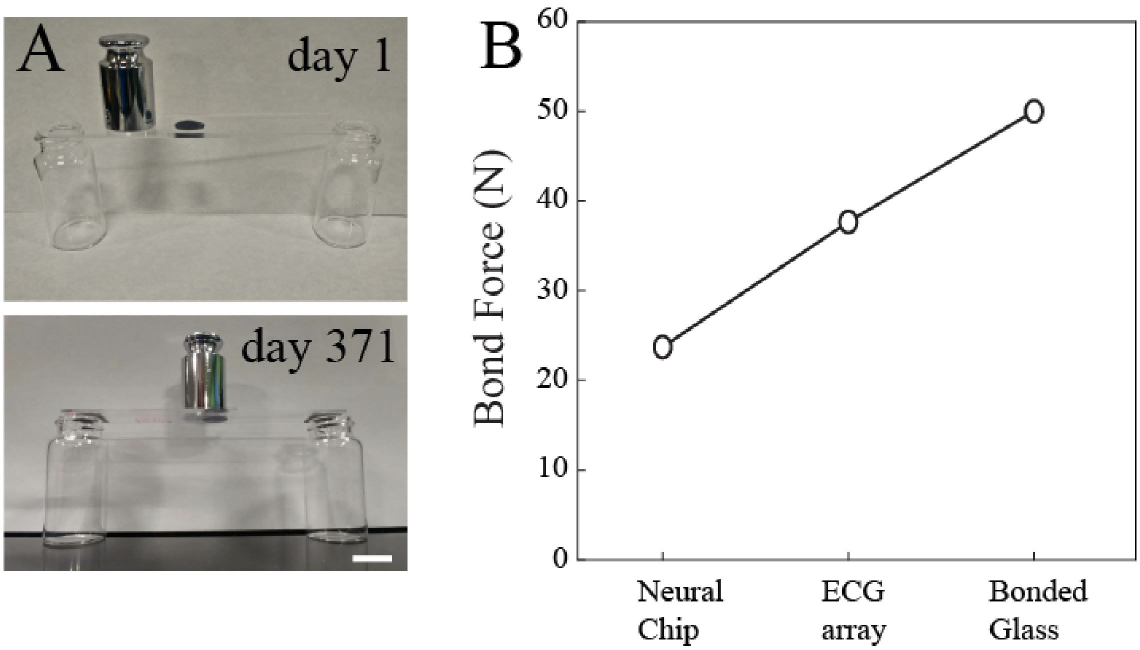
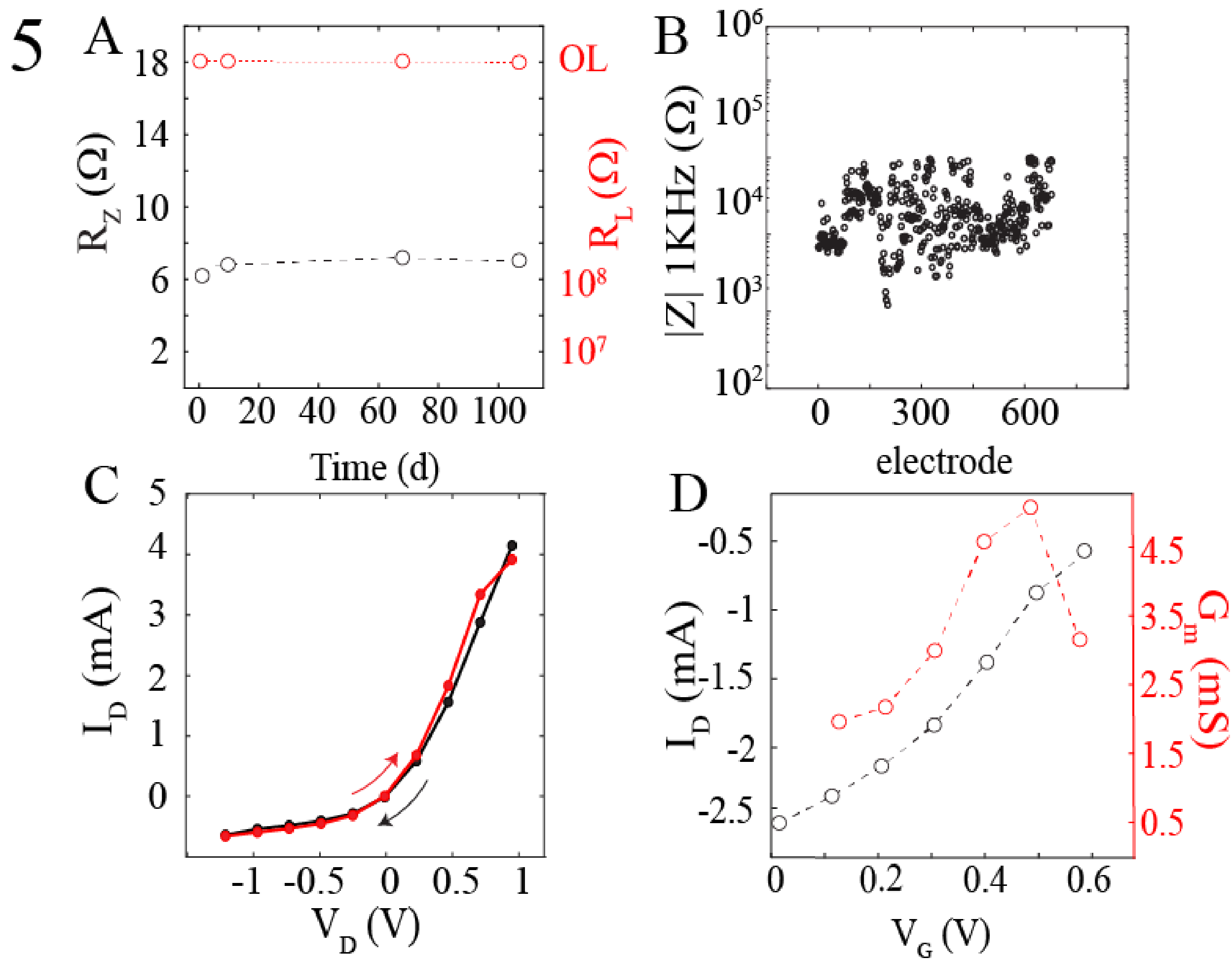


Supplementary Figure 4: MCP creates high bonding strength interfaces.

(A) MCP establishes a strong mechanical bond. MCP (30 µm particles) of area 1.3 cm^2 bonds two glass slides that form a bridge capable of supporting a 100 g weight, both initially, and 371 days after the bond was formed. Scale bar 10 mm. Photo Credit: Patricia Jastrzebska-Perfect, Columbia University

(B) MCP withstands strong mechanical stresses. "Bonded glass" represents the horizontal stress applied to the bond visualized in A. "ECG array" and "neural chip" represent extrapolations of the forces that the ECG array (10 x 10 mm^2) and neural chip (7 x 9 mm^2) could withstand, based on their bonded surface areas.



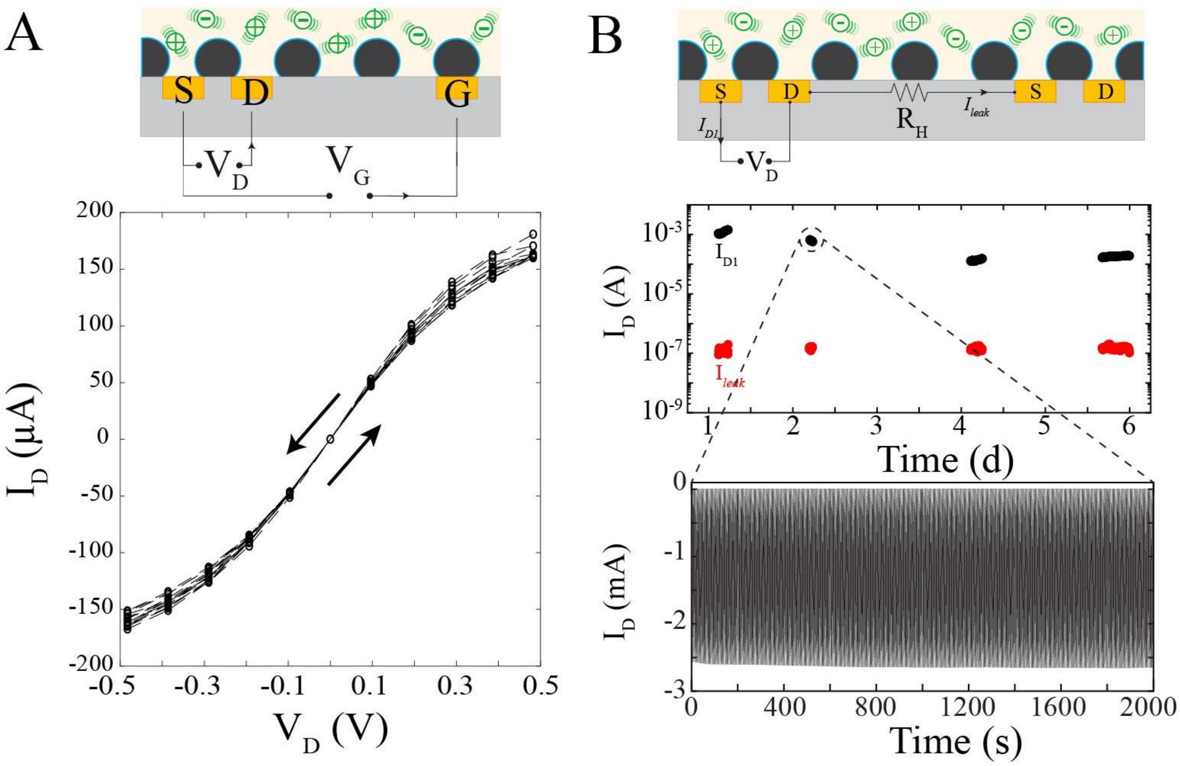
Supplementary Figure 5: Performance of MCP-based devices.

(A) MCP creates a stable, highly anisotropic contact for 100 days (particle size = 40 µm, electrode pitch = 250 µm). OL = overload.

(B) Impedance of individual electrodes for a high-density neural interface device bonded with MCP. MCP enables high electrode yield. Electrode size = 20 x 20 µm^2.

(C) Forward and reverse current of an MCP-based diode (particle size = 10 µm, L = 200 µm, W = 700 µm) displays minimal hysteresis. Voltage was swept from 1 to -1.2 V, and then reversed.

(D) Transfer curve for MCP-based transistor shown in **Figure 3** (particle size = 30 µm) at V_D = -0.6 V (left axis), and corresponding transconductance (G_m = 4.82 mS).



Supplementary Figure 6: MCP-based integrated circuits display low leakage in a physiological environment over extended time period.

A. MCP-based transistor (particle size = 10 μm, L = 30 μm, W = 500 μm) at zero gate voltage displays minimal hysteresis. Voltage was swept from 0.5 to -0.5 V, and then reversed.

B. Schematic of two adjacent MCP-based transistors. I_{leak} shows the leakage current due to horizontal resistance $R_{H.}$ Drain (I_{D1}) vs. leakage (I_{leak}) current for two adjacent MCP-based transistors (particle size = 30 μm, 200 μm channel length and transistor spacing) over six days in PBS solution. Gate voltage was applied in 1 s pulses. Note that there is no voltage across the channel of transistor 2 (top). Drain current modulation is consistent across time, at the given time point (bottom).

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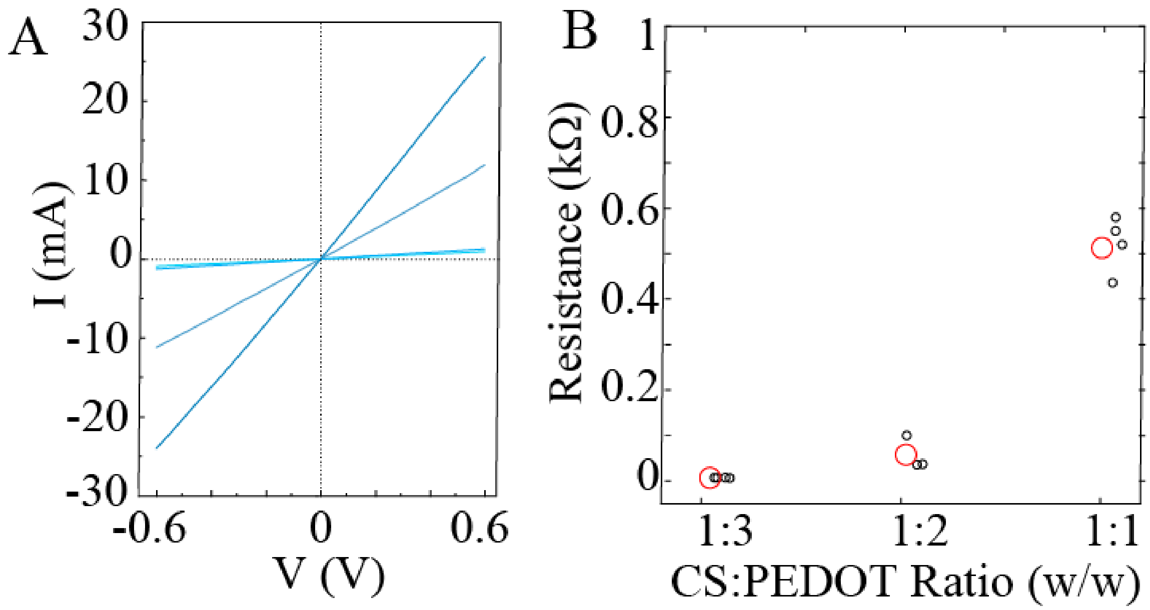
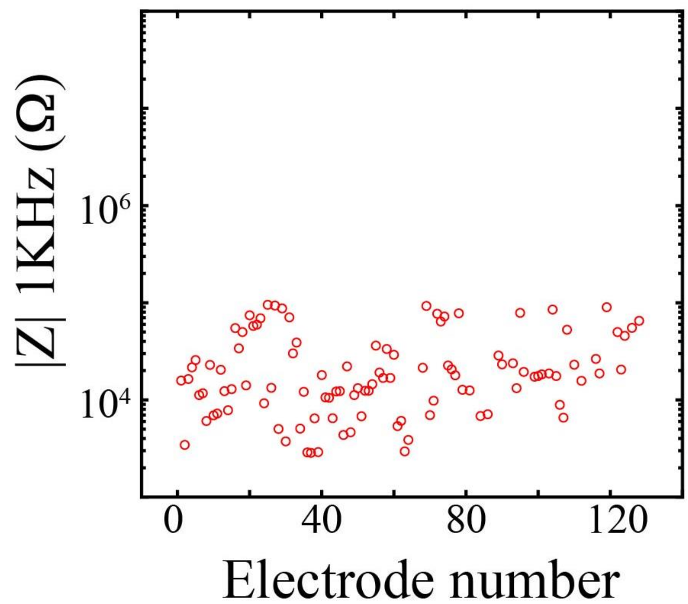
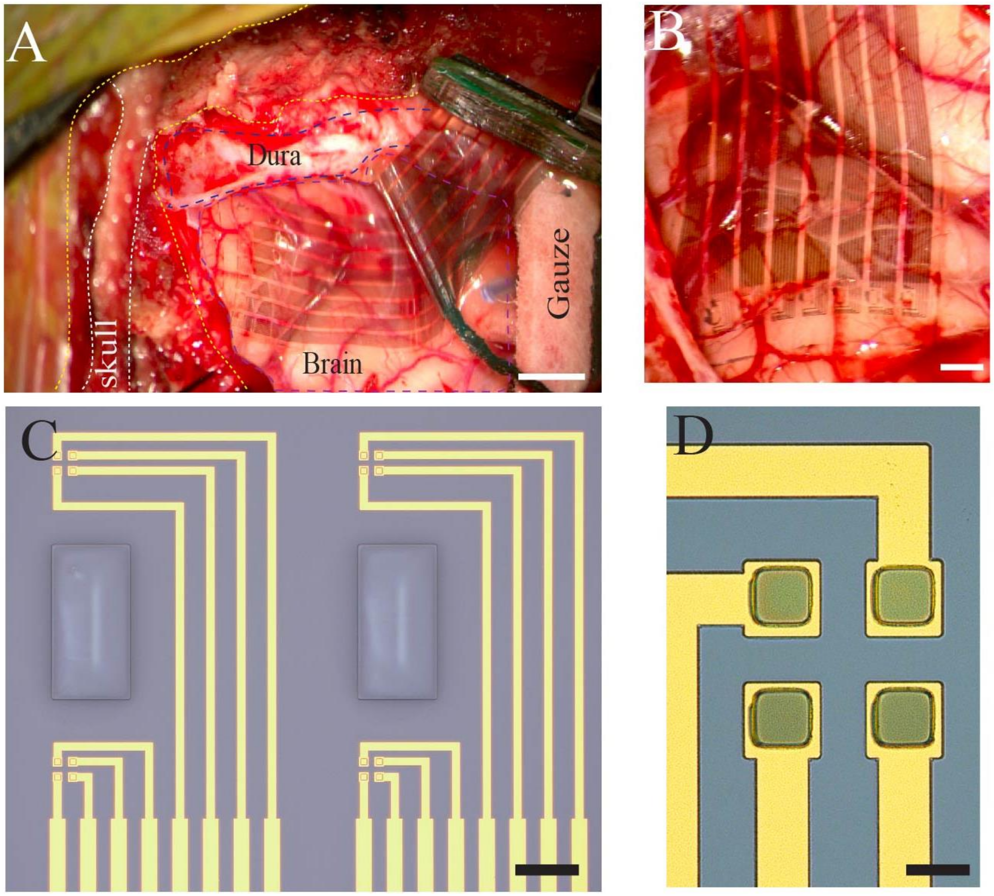


Supplementary Figure 7: Varying conducting polymer particle density results in highly consistent, inversely proportional changes in resistance.

A. IV characteristics of MCP film (L = 250 µm, W = 5 mm) for various particle concentrations (CS/PEDOT:PSS wt:wt ratio 1:3 (light blue) and 1:1 (dark blue), particle size = 30 µm).
B. Resistance of MCP varies with conducting polymer particle density and is highly consistent (independent MCP sample measurements (black circles) and mean value (red circles).

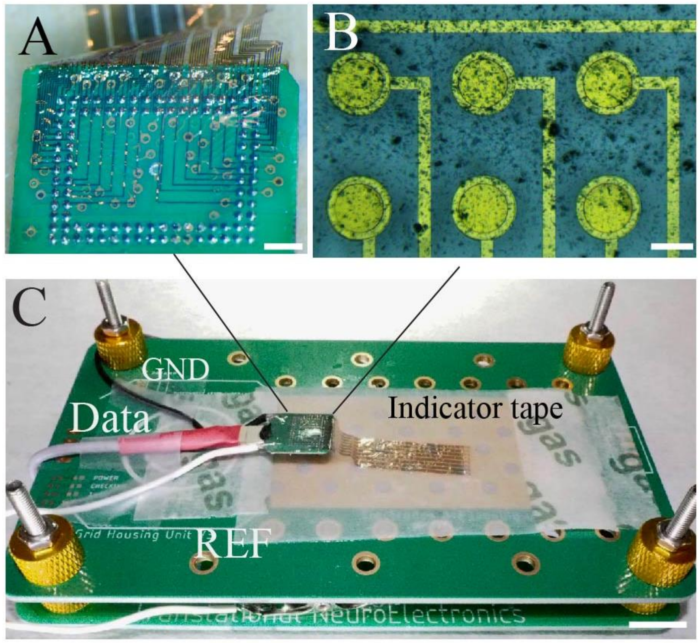


Supplementary Figure 8: Electrochemical impedance values at 1 kHz frequency for each electrode of a neural interface device with 128 electrodes.



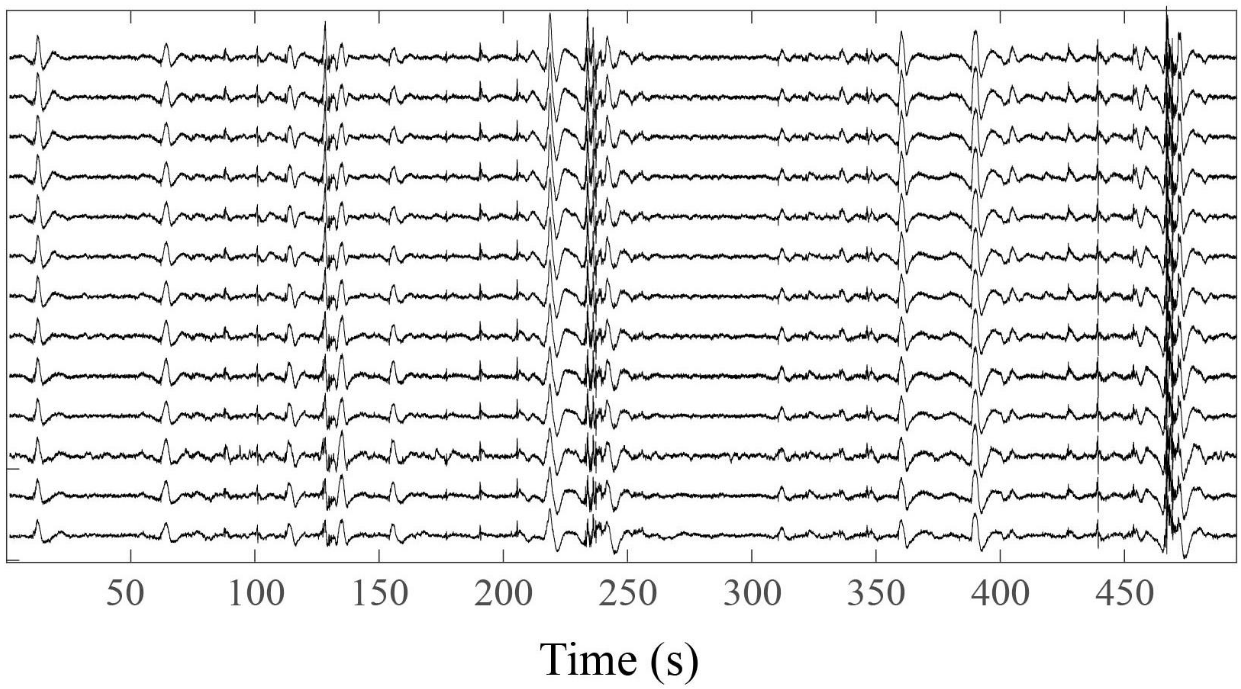
Supplementary Figure 9: MCP enables intra-operative recordings from the surface of the human brain.

A. Photograph of a NeuroGrid device and its associated electronics during an intra-operative procedure. Scale bar, 5 mm. Photo Credit: Dion Khodagholy, Columbia University
B. Micrograph of the front-end electrodes of the NeuroGrid conforming on the curvilinear surface of human cortex. Scale bar 1 mm. See also **Supplementary Video 1**. Photo Credit: Dion Khodagholy, Columbia University
C. Micrograph of the tetrode (four electrode) structure of the NeuroGrid with large perforations to facilitate cerebrospinal fluid circulation. Scale bar 80 µm. Photo Credit: Dion Khodagholy, Columbia University
D. Micrograph of single tetrode with PEDOT:PSS coating. Scale bar 20 µm. Photo Credit: Dion Khodagholy, Columbia University

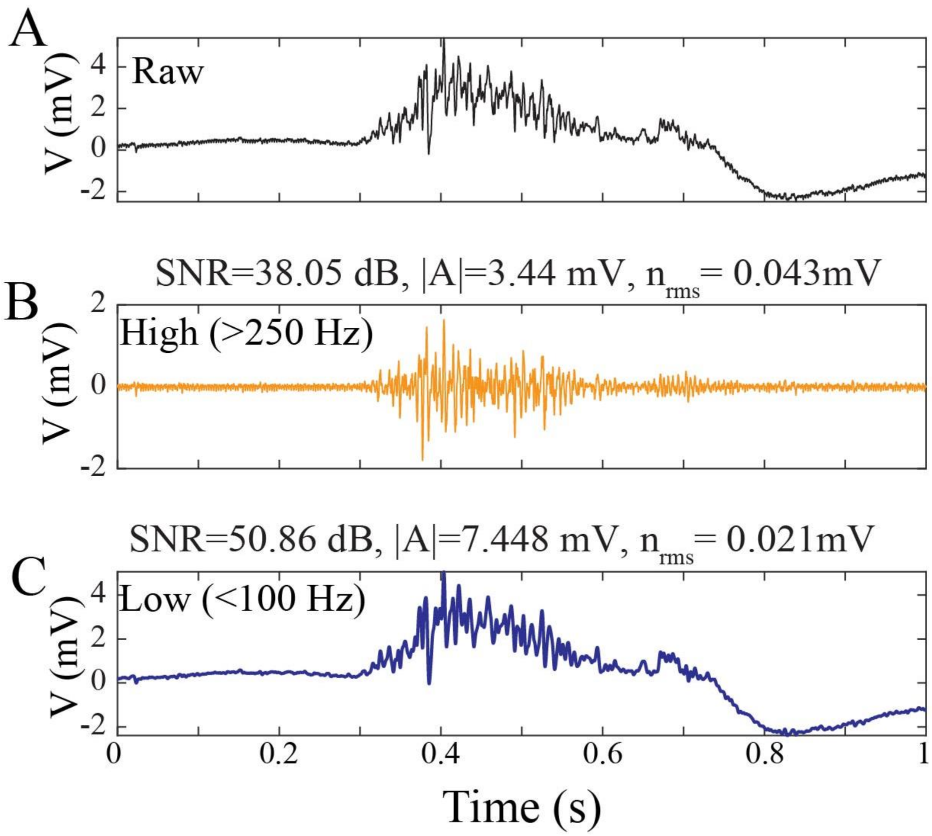


Supplementary Figure 10: MCP film creates high quality electrophysiological interface that is unaffected by sterilization with ethylene oxide gas.

A. Conformable NeuroGrid bonded to rigid back-end amplifier circuit using MCP. Scale bar 1 mm. Photo Credit: Dion Khodagholy, Columbia University
B. Micrograph of MCP deposited on Au-based electrode array. Scale bar 250 µm. Photo Credit: Patricia Jastrzebska-Perfect, Columbia University
C. Sterilization packaging of the MCP-based NeuroGrid. Scale bar 10 mm. Photo Credit: Dion Khodagholy, Columbia University



Supplementary Figure 11: Continuously-recorded, high-pass filtered (1 Hz) EMG signals across multiple channels.

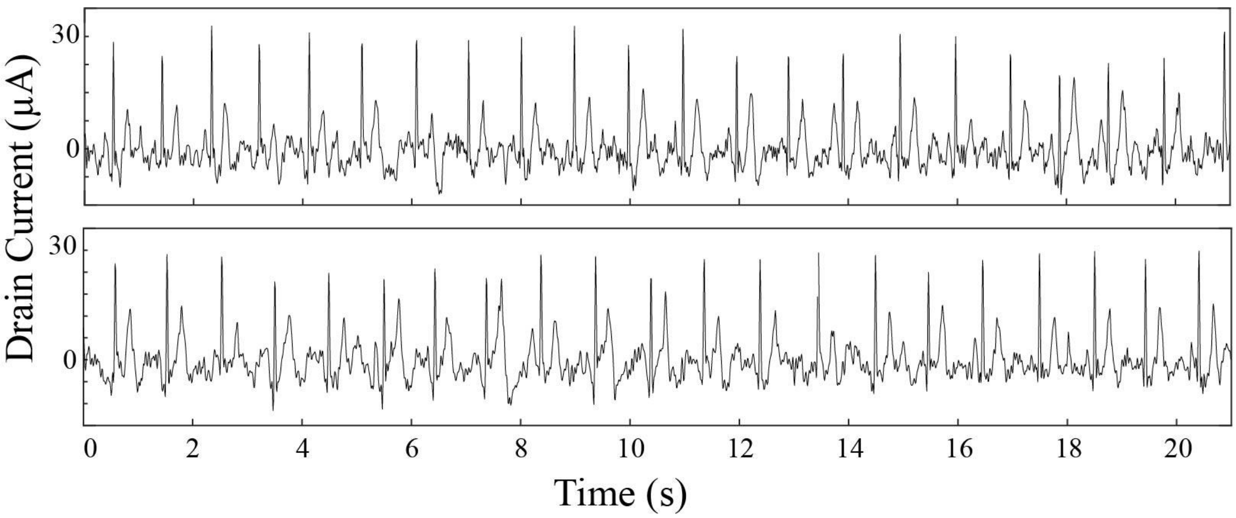


Supplementary Figure 12: MCP enabled EMG recordings with high signal-to-noise ratio and gain, as well as low noise.

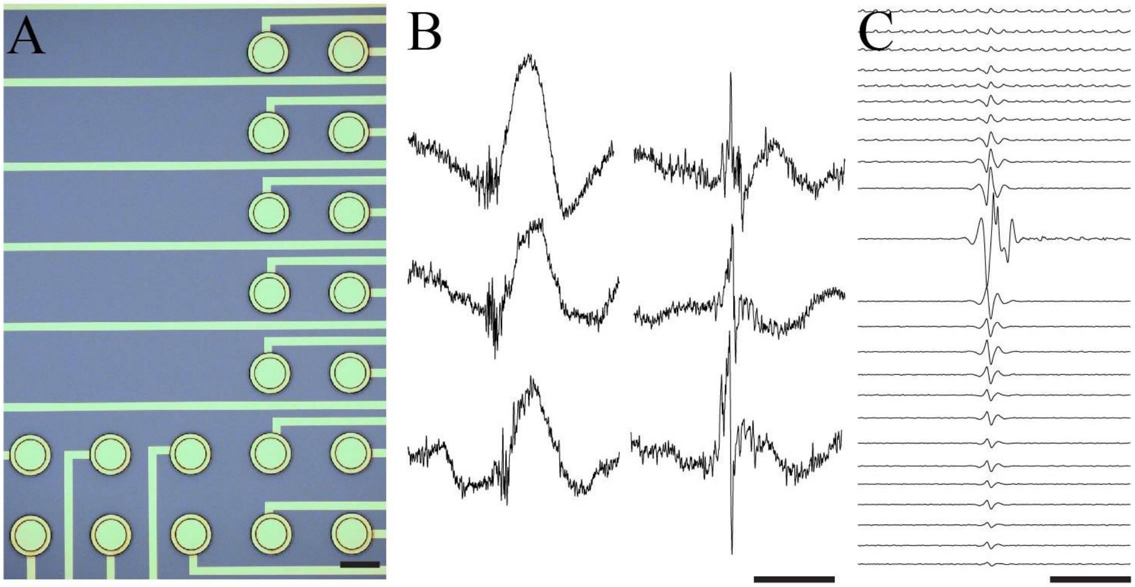
A. Unfiltered (0.1-7500Hz) EMG trace.

B. High-pass filtered (250 Hz) EMG trace. SNR = signal-to-noise ratio (dB), |A| = signal amplitude. n_{rms} = noise (root mean square).

C. High-pass filtered (100 Hz) EMG trace.



Supplementary Figure 13: ECG recordings acquired by MCP interface and organic electrochemical transistor (L = 250 μm, W = 250 μm, thickness = 0.2 μm).



Supplementary Figure 14: MCP-based biopotential array is able to acquire high spatiotemporal resolution EMG signals.

A. Micrograph of conformable EMG array electrodes. Scale bar 250 µm. Photo Credit: Dion Khodagholy, Columbia University
B. Sample EMG activity of two digits (rows) with consistent response across trials (columns) acquired by conformable MCP-based biopotential array. Scale bar 80 ms.
C. High-pass filtered (200 Hz) time traces of nerve action potential across multiple channels. Scale bar 5 ms.

Legend for movie S1: MCP-based neural probe conforms on the curvilinear surface of the human brain.