Supplementary Figures



S. Figure 1 | Laser-cut stencil characterization. (a) The 2D schematic of laser cutting the stencil of electrode vias. The stencil was cut using a commercial CO2 laser cutter (PLS6.140D, Universal System) through the PI tape supported by a stainless-steel sheet. (b)-(e) Images of laser cut stencils at different power and speed (b) A laser power setting of 10% and speed setting of 50% failed to create a distinct boundary between 2 neighboring electrodes. (c) A laser power setting of 10% and a speed setting of 100% created fully etched vias with distinct boundary between electrode tracks. (d) At a laser power setting of 5% maximum and speed setting of 100% maximum, the laser could fully cut through the PI tape but had carbonized residue filling in the stencil, which was hard to remove. (e) At a laser power setting of 2.5% maximum and speed setting of 100% maximum, the laser could not fully cut through the PI tape. The scale bar indicates 2 mm. We chose the settings in (c) for further experiments.

Laser-cut stencil optimization: to achieve the most stable design of the electrode array, the laser cutting power and speed determine whether the adjacent electrodes are isolated from each other without any interconnects. **S. Figure 1a** presents the animated schematic, in which the CO₂ laser cut through the PI stencil supported by a stainless-steel sheet. The optimized power at 10% and speed at 100% experimentally achieve the best stencil of the electrode's center to center distance of 500 μ m (**S.Fig 1b – e**).



S. Figure 2 | **bonding between PCB and ECoG electrode array. (a)** 2D and 3D schematic of the bonding between the graphene ECoG array and interface PCB using conductive adhesive. (b) Photograph illustrating graphene ECoG array and interface PCB at the bonding sites. The scale bar indicates 2 mm.

Bonding between ECoG electrode array and PCB interface: The PCB's metal pads are normally used for soldering. Silver epoxy (8331 Silver Epoxy Adhesive, MG Chemical) was applied individually on each metal pad and subsequently bonded to the graphene electrode **(S.Fig. 2a,b)**.



S. Figure 3 | **Bending test. (a)** The 3D and 2D schematic of the bending test setup. 3 identical electrodes built by the same fabrication procedure of the graphene ECoG electrode array were conformed to the custom supporting structure with a curvature. The electrode length is 15.7 mm. The electrode width is 100 μ m. (b) Photograph of the measurement setup.

Bending test: to evaluate the electrode performance under static bending stress, 9 straight electrodes built using the same ECoG stencil printing technique were placed on the support structures with different radii of curvature. The impedance was measured using the interface board (RHD2000, Intan Technologies Inc.) (**S.Fig 3a, b**).

а



i. A pair of silver contacts were deposited on PI film using e-beam evaporator.



b

ii. 4 mm by 20 mm Cyrene/Graphene traces were deposited on the silver contacts for further electrical characterization tests

S. Figure 4 | **Sample preparation for electrical characterization: (a)** A pair of silver contacts were deposited on PI film using e-beam evaporator through a lithography mask. (b) 4 mm x 20 mm graphene trace was patterned on the silver contact for electrical characterization tests.

Sample preparation for electrical characterization: To fabricate the Ag contacts on the PI film, a dark mask was first printed on a transparent PET (Polyethylene terephthalate) substrate (**S.Fig 4. a**) using a regular desktop printer. The AZ5214 photoresist was spin-coated at 4000 rpm for 30 s on the PI film and baked at 110°C for 90 s on a hot plate. It was then exposed to the dark mask with UV light using a mask aligner for 30 s. It was then developed in the AZ developer and after developing, a clear transparent pattern on the PI film was visible. Then the e-beam evaporator was used to deposit the Ag, which deposited a uniform coating, which later went through a lift-off process with acetone solvent to achieve the desired pattern of Ag contact. ~ 0.5 mL of three graphene inks were drop cast and annealed at 350°C for 90 minutes to create the samples.



S. Figure 5 | **Custom-built spin coater: (a)** Photograph of a custom-built microprocessor-controlled spin coater. **(b)** The spin coater has one spinning disk with 2 substrate holder clamps that hold the substrate for spin coating. **(c)** The major components of the spin coater include a microcontroller, rotary encoder, liquid crystal display (LCD) interface, power switch, power supply, motor, and chamber. **(d)** Circuit layout. ESC in the schematic abbreviates electronic speed controller. **(e)** The relationship between spinning speed and PDMS thickness was obtained using the custom-built spin-coater compared to results obtained using a commercial spinner [1].

Custom-built spin coater: the spin coater was designed to be powered by a 12-volt power supply and run with an Arduino microcontroller (**S.Fig 5. d**). The use of an Arduino is convenient due to its ability to be easily programmed and tested. The motor can operate in a specific rpm window of about 2000 to 8000 RPMs; an electronic speed controller (ESC) can supply a pulse-width modulation signal from the Arduino microcontroller to control the motor (**S.Fig. 5d**). The device is mainly controlled by a rotary encoder (**S.Fig. 5c**). A standard 16x2 LCD was used to display adjustable options and a timer and RPM counter when the spin-coater was working (**S.Fig. 5c**). To allow easy cleaning, acrylic sheets for the enclosure and a removable cake pan were selected as the spinning chamber (**S.Fig. 5a**). The sample can be placed on the spinning disk and clamped by a pair of substrate holders (**S.Fig. 5b**)



S. Figure 6 | **Raman spectrum:** Raman spectrum of the three graphene films at room temperature, showing distinct Raman D, G, and 2D-band peaks, where the Id/Ig ratio was lower for the film originated formed from Cyrene-exfoliated graphene ink.

Graphene sample preparation for Raman Spectroscopy: 0.5 mL of the prepared graphene ink was extracted from the vial using a disposable pipette and released onto a Si/SiO₂ substrate, followed by spin coating at 1000 rpm for 20 sec using a custom-built microcontroller-based spinner (S.Fig. 5). To expose single/multilayer graphene flakes and minimize stack-up of multiple flakes, the sample was then annealed at 350 °C for 90 minutes. A 532 nm wavelength laser was used to perform Raman Spectroscopy measurements. The laser power was set to an initial setting of 0.10 mW and increased gradually to 16 mW. The measurement acquisition time was fixed at 0.1 s to minimize any local heating that could be caused by the laser [1].



S. Figure 7 | **Graphene electrode surface profiling: (a)** C/T+EC+Gr electrode. **(d)** DMF+EC+Gr electrode. **(c)** Cyrene+Gr electrode.

We used a surface profiler (K1-P16, KLA-Tencor) to measure the Graphene electrode surface profile.

Supplementary Reference:

[1] F. K. Balagaddé et al., "Microbiology: Long-term monitoring of bacteria undergoing programmed population control in a microchemostat," Science, vol. 309, no. 5731, 2005, doi: 10.1126/science.1109173.

[2] H. Zobeiri et al., "Hot carrier transfer and phonon transport in suspended nm WS2 films," *Acta Materialia*, vol. 175, 2019, doi: 10.1016/j.actamat.2019.06.011.