ADVANCED FUNCTIONAL MATERIALS

Supporting Information

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Experimental Section

Materials

The PEDOT:PSS aqueous suspension (Clevios PH1000) was purchased from Heraeus Electronic Materials GmbH (Leverkusen, Germany). Capstone FS-30 was purchased from Alfa Chemicals. Agarose powder (molecular biological grade) and glass slides were purchased from Thermo Fisher Scientific. Polydimethylsiloxane (PDMS, Sylgard 184 silicone elastomer kit) was purchased from Dow Corning. Polyethylene terephthalate (PET) sheets were purchased from Policrom Inc. (Bensalem, PA, U.S.A.). Parylene (Parylene C) was from SCS coating.

Temporary tattoo transfer paper, PVA hydrogel carrier (3M 5414), the liquid metal alloy EGaIn, pressure-controlled conductive adhesive (3M 9703), protection tapes (3M Tegaderm roll -16002), and copper tapes were purchased from Amazon. Dulbecco's phosphate-buffered saline (DPBS) was purchased from Gibco.

The electronic kit for the wireless circuit design RFduino-Starter Kit (Simblee^{TM}, DEV-13785) and DAC (BOB-12918 ROHS for gate simulation) was purchased from SparkFun Electronics.

DBSA, glycerol (99.5+ % purity), GOPS, PVA powder, gelatin powder (gelatin from porcine skin), Hexadecyltrimethylammonium bromide (or cetyltrimethylammonium bromide, CTAB), Ag wire (gate electrodes), glucose, glucose oxidase from Aspergillus Niger (G7141-50KU), chitosan (high molecular weight), and acetic acid were purchased from Sigma-Aldrich. Ti and Au were supplied by the Integrated Systems Nanofabrication Cleanroom (ISNC) at CNSI, Pt was supplied by NanoLab at Electrical Engineering at UCLA. The optical shadow masks were purchased from Fineline Imaging, Inc.

PEDOT:PSS processing

The PEDOT:PSS suspension was first agitated for 5 min and then mixed with various additives, including DBSA (0.5 v/v.%), Capstone FS-30 (1 v/v.%) glycerol (5 v/v.%) and GOPS (1 v/v.%). The thickness of the PEDOT:PSS was controlled by varying the spin-coating speed from 500 rpm to 4000 rpm, resulting in film thicknesses ranging from 40 nm to 200 nm. Surfactants DBSA and Capstone were used to increase the film's wettability on glass. Glycerol was used to enhance film conductivity. For the annealing test, films were baked on a hotplate for a time period from 30 seconds to 1800 seconds. In all other cases, the films were baked for 1800 seconds on a hotplate after spin-coating on glass.

Characterization of PEDOT:PSS thin films

The thickness of the PEDOT:PSS was measured with a Vecco Dektak 150 profilometer. The contact angles of PEDOT:PSS mixed with different additives were measured with an AST VCA-3000S Wafer Surface Analysis System (AST Goniometer).

Parylene patterning of PEDOT:PSS films on glass

The glass substrates were pre-cleaned with acetone, IPA and DI water. CTAB solution (0.005 M) was spin-coated at 500 rpm for 30 seconds on the glass substrates prior to Parylene (Parylene C) coating. Next, 2-μm-thick Parylene was deposited. The photoresist SPR 700-1.2 was spin coated at 2000 rpm for 30 seconds (with a thickness around 1.8 μm) on the Parylene layer, and then the samples were exposed to the UV light of a mask aligner (12 mw/cm^2 for 6 seconds, Karl Suss MA6) and developed in the developer MF-26A (Micro Chemicals Ltd.) for 1 min. Next, the unprotected Parylene was etched away using an Oxford 80 Plus - Fluorine RIE (oxygen plasma). After etching, the remaining photoresist was stripped using acetone. After a 10-min UV treatment (Jetlight - UV Cleaner) of the patterned parylene layer, PEDOT:PSS was spin-coated on top of the substrate. After baking the PEDOT:PSS for 2 min, the parylene layer was gently

peeled-off using tweezers, leaving patterned PEDOT:PSS on glass. A hard baking of 100 °C for 30 min was performed to totally remove the water in the PEDOT:PSS films.

Hydrogel and PDMS preparation

PVA powder and DI water were mixed together and stirred at 90°C to acquire a clear 10 wt.% PVA solution. The solution was dried in a chemical hood to obtain the PVA hydrogel. Gelatin powder was mixed with DPBS (16 wt.%) and vortexed for 5 min. The mixture was then put in an 80°C oven overnight to form a clear solution. The obtained solution was poured on a plastic petri dish and naturally cooled to form uniform gelatin hydrogel. Agarose was mixed with DI water (9 wt.%) in a microwave heater for 1 min to form a clear agarose solution. The solution was poured on a plastic petri dish and naturally cooled to form uniform agarose hydrogel. PolyHEMA was synthesized by mixing HEMA monomers, 1 v/v.% EGDMA as a crosslinker, 0.5 v/v.% BP as a heat initiator. The mixture was then cured in an oven at 80 °C for 1 hour. After curing, the samples were immersed in water for 1 day before use.

PDMS (10:1) was mechanically stirred before de-gassing in a vacuum desiccator. CTAB solution (0.005 M) was spin-coated on glass, acting as an antiadhesive layer. Next, the premixed and de-gassed PDMS was spin-coated on the glass substrates. Afterward, the samples were cured at 85 °C for 1 hour in an oven. Finally, the PDMS substrates can be detached easily from the glass slides with tweezers.

For the swelling test of the PEDOT:PSS film, the PEDOT:PSS film was obtained by drop-casting PEDOT:PSS suspension (0.5 v/v.% DBSA) on glass substrate. After baking at 100 °C (overnight), gelatin hydrogel was placed on top of the film, with an area of 10 mm by 10 mm. The weight of the films was monitored by a 5-digit weight balance.

Transfer-printing PEDOT:PSS onto flexible substrates

PEDOT:PSS films (with surfactants) were first spin-coated on glass. Next, hydrogel carriers (PVA, gelatin or agarose) were gently attached on top to obtain a seamless contact. The hydrogel carrier was then slowly peeled-off from the substrates which also led to the peeling-off of the PEDOT:PSS from glass. The delaminated PEDOT:PSS/hydrogel was then laminated on target substrates including plastic, elastomer, and skin. In case of using water-soluble hydrogel PVA as a carrier, dissolving PVA with water (60 °C for 30 min) completed the transfer of PEDOT:PSS films onto target substrates. The resultant films on target soft substrates were conformable and conductive as examined by the Agilent Source Measure Unit.

Mechanical test of peeling-off PEODT:PSS with hydrogels

The peeling test was performed with a mechanical testing system (Model 5943, Instron). Hydrogel substrates were first laminated seamlessly on PEDOT:PSS films on glass. The length of the hydrogel was longer than the glass substrates which eased the connections with the Instron. The hydrogel was stretched at a constant rate of 0.1 mm/s. The load force was recorded simultaneously with the system during stretching.

Fabrication and characterization of skin-attachable OECT on tattoo paper

Metallic electrodes (source, drain, and gate) were deposited and patterned on a tattoo paper directly using a shadow mask. Next, the PEDOT:PSS film was transfer-printed and aligned in between the interdigitated source-drain electrodes (with width of 10 mm, and length of 1 mm). After dissolving the hydrogel carrier, the OECT on tattoo paper was transferred onto target substrates with tattoo-adhesive tapes by exposing the PEDOT:PSS channel directly to the target. Dissolving the tattoo substrates with water completed the transfer process. Chitosan or 3M Tegaderm roll (16002) may be coated on the OECTs to prevent film delamination. The electrical measurements (output, transfer, transient) of OECTs were performed using an Agilent B2902A

source-measure unit (SMU) controlled by LabVIEW software. For standard measurements, 10^{-3} M CTAB in water was used as gate electrolyte and an Ag wire was used as the gate electrode. The liquid metal alloy EGaIn, pressure-controlled conductive adhesive (3M 9703), and copper tape were used to facilitate electrical connections between the OECTs and external wires to the SMU.

Glucose sensing with OECT

30 mg GO_x was mixed with 1 ml PBS solution. 0.5 g chitosan and 0.5 ml acetic acid were mixed together in 100 ml DI water. Then 100 µL GO_x solution was mixed with 100 µL chitosan solution. The resultant mixtures were stored in 4 °C fridge for further use. For glucose sensing, 4 µL GO_x and chitosan mixture was deposited on Pt gate electrodes and dried at room temperature. Glucose solution (100 mM) was prepared by dissolving glucose powder in PBS solution. Lower concentrations were obtained by diluting the 100 mM solution with PBS. The experiments were performed by laminating our skin-attachable OECTs onto a plastic substrate. The sequence of sensing was performed from low to high concentrations.

Institutional Review Board (IRB) approval for human subject testing

The conducted human subject experiments were performed in compliance with the protocols that have been approved by the IRB at the University of California, Los Angeles (IRB#17-000170). All subjects gave written informed consent before participation in the study. For all demonstrations on human skin, a signed consent was obtained from the volunteer.

Figure S1. Optical images of transfer-printed PEDOT:PSS thin films, with a thickness of 100 nm, from glass substrates onto skin: a) after transferring; b) under mechanical compression; and c) under mechanical distortion. The scale bar is 1 cm.

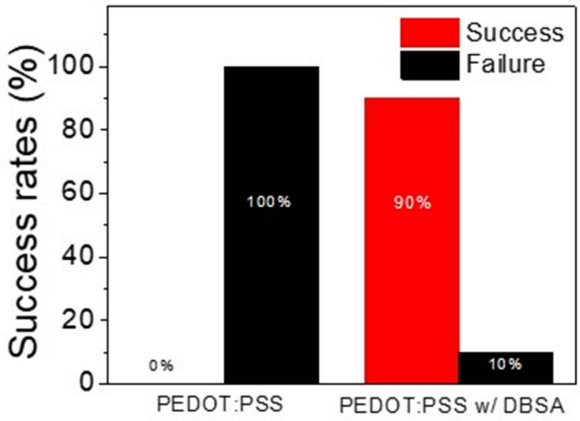

Figure S2. Success rates of transfer-printing pristine PEDOT:PSS films and PEDOT:PSS films doped with DBSA (1 v/v.%). The samples were baked at 120 °C for 30 min. Each measurement was tested for 10 times.

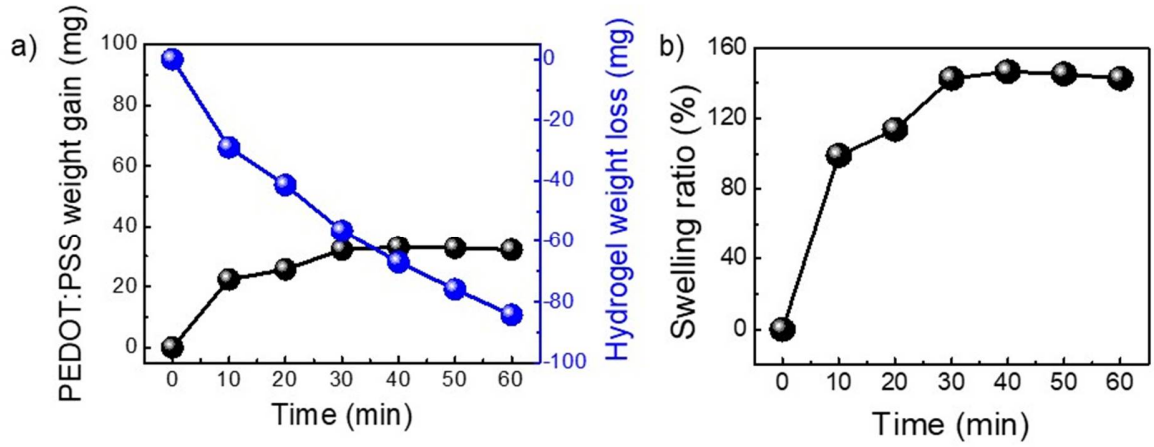

Figure S3. a) Swelling of PEDOT:PSS films upon contacting with hydrogel. The weight of PEDOT:PSS film (10 mm x 10 mm) increased from ~20 mg to 52.1 mg (weight gain of 32.1 mg), suggesting hydrogel significantly swelled the PEDOT:PSS films; b) the calculated swelling ratio of the PEDOT:PSS film. The initial thickness of the PEDOT:PSS film was about 17 μm.

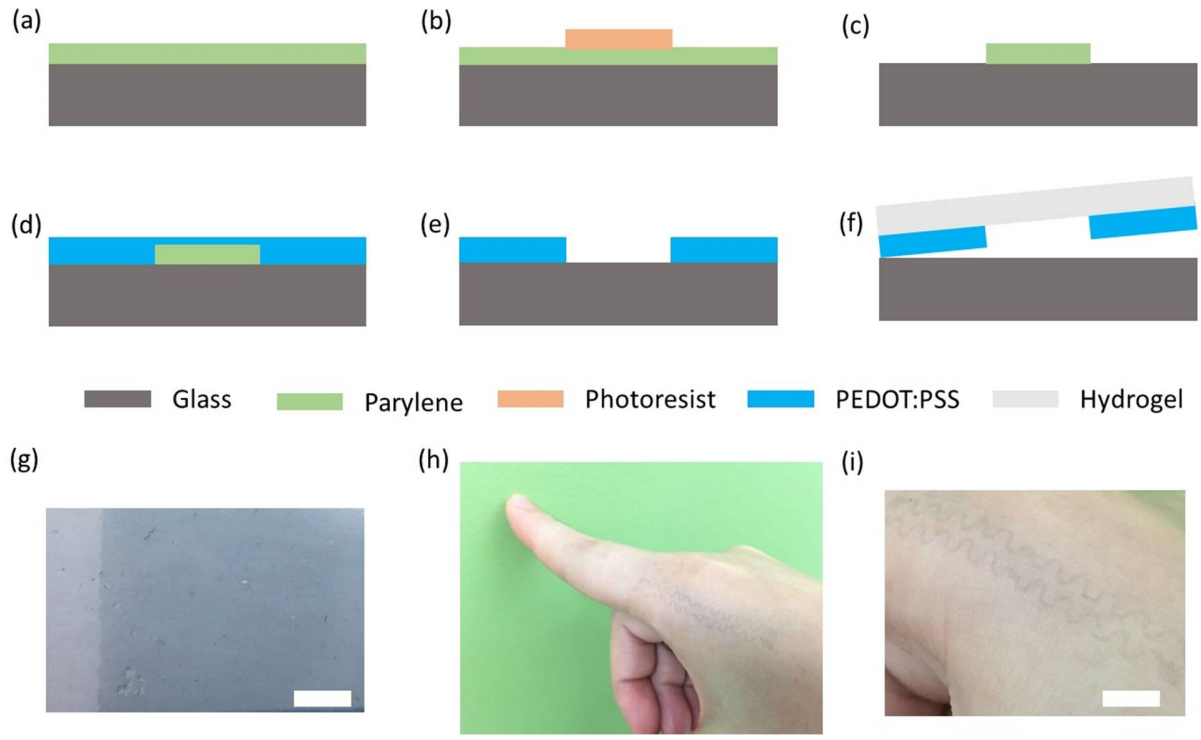

Figure S4. Process flow of transfer-printing micropatterned PEDOT:PSS films onto skin. The PEDOT:PSS thin films were first patterned on glass with a parylene mask: a) chemical vapor deposition (CVD) of 2 µm parylene on glass substrates; b) spin-coating and pattern SPR 700-1.2 photoresist; c) photoresist development and parylene etching with OXFORD RIE; d) spin-coating PEDOT:PSS films (with 0.5 v/v.% DBSA) at 2000 rpm for 30 s; e) parylene layer peeling-off, leaving patterned PEDOT:PSS thin films on glass; f) transfer PEDOT:PSS thin films with hydrogels; g) optical images of transferred PEDOT:PSS films on glass, scale bar is 5 mm.; h) optical images of transfer patterned PEDOT:PSS films with serpentine structures on finger; i) magnification image of PEDOT:PSS patterns on skin. The width of the PEDOT:PSS pattern is 500 µm. The scale bar is 1 cm.

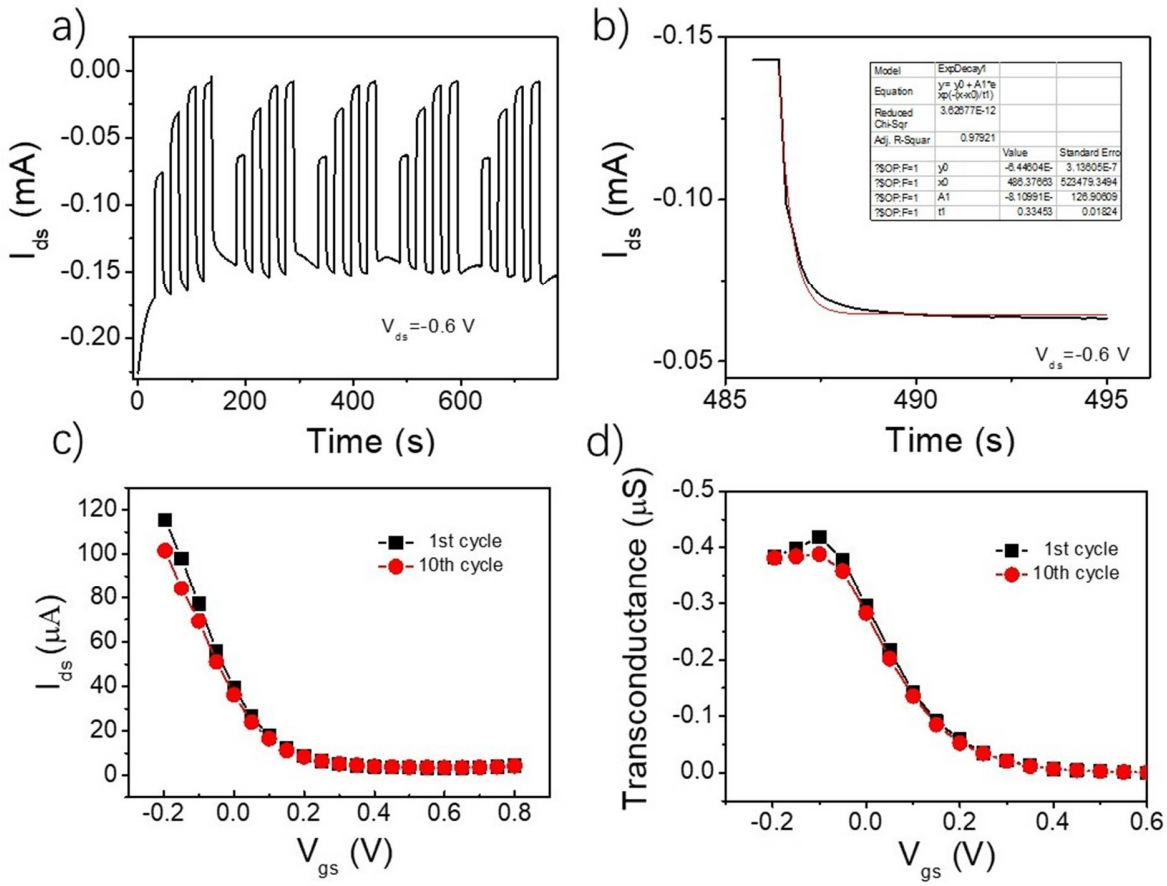

Figure S5. a) Transient behavior of skin-attachable OECTs. The V_{gs} was applied from 0 V to 0.8 V with a step of 0.2 V. The width (W) of the channel is 10 mm, the length (L) of the channel is 1 mm (W/L=10); b) The time constant of the response time is ca. 0.33s (red-line refers to the nonlinear curve fitting result using ExpDecay Equation); c-d) Cyclic stability of transfer and transconductance plots of OECTs.

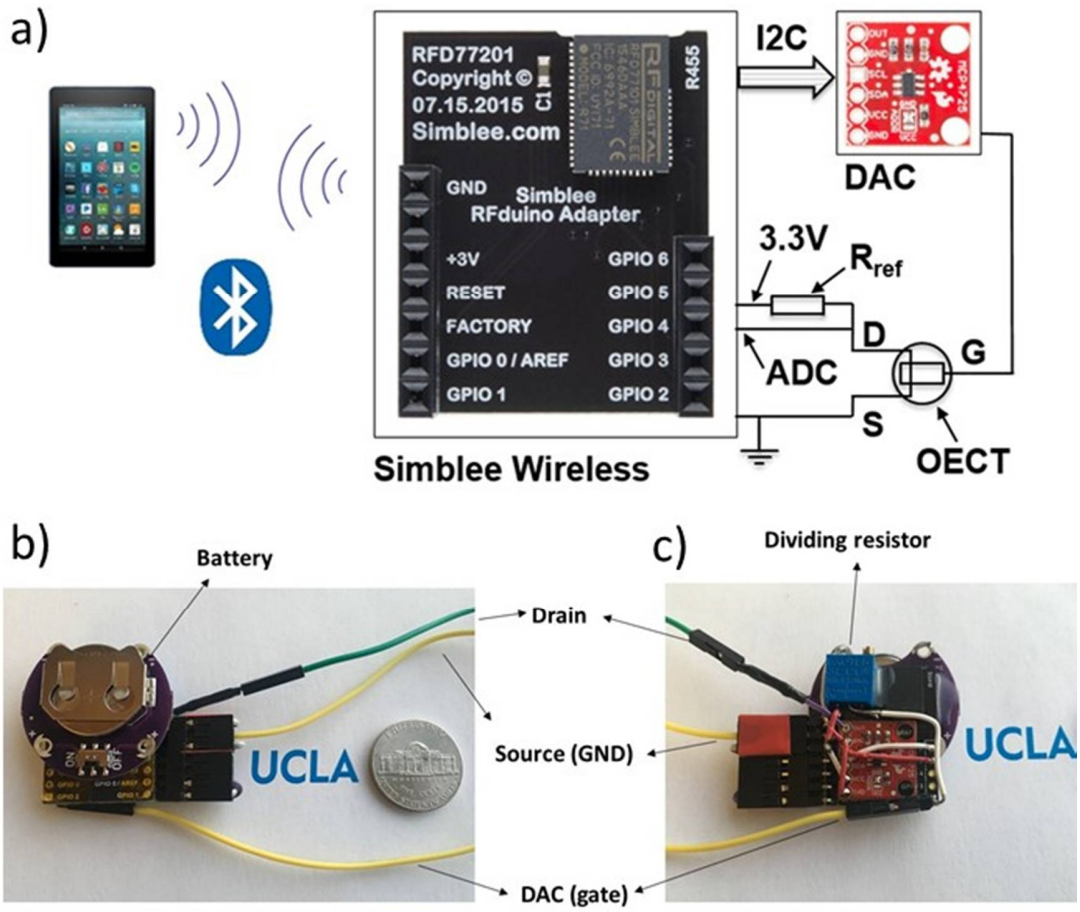

Figure S6. Wireless gating and data readout system for OECT characterization: a) schematic and images of electronic components involved in our minimized portable system. It included a micro-controller unit (Arduino) integrated with radio frequency adapters for Bluetooth communication with mobiles, an analog to digital converter (ADC) for source-drain current monitoring, and a digital to analog converter (DAC) for gate voltage stimulation; b) A button cell was integrated with the system and c) a variable resistor was used to control the voltage drop on the OECTs. The diameter of the coin is 1.9 cm.