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Supporting Information

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Single-Material OECT-Based Flexible Complementary Circuits Featuring Polyaniline in Both Conducting Channels

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Figure S1. A) AFM images of bare electrodes, and electrodes with an electroactive PANI film on the surface, 50 x 50 μ m (B) and 2 x 2 μ m (C). D-E-F) 3D rendered AFM topography of the same areas. AFM image of the edge of PANI film (G) and its relative thickness profile (H).



Figure S2. Three cycles were recorded in ionic liquid at a scan rate of 50 mV.s⁻¹. The counter electrode was a Pt plate and the reference was Ag/Ag^+ electrode. The black arrow and grey box highlight hysteresis as we sweep the potential back from positive to negative, in correlation with the hysteresis observed in the transfer curves. The yellow regions indicate the edges of the finite electrochemical window of PANI that does not exceed 1 V.



Figure S3. A) Drain current I_{DS} vs gate voltage V_{GS} at drain-source voltage $V_{DS} = -0.1$ V. Black lines represent the leakage current. B) I_{DS} vs V_{DS} for V_G from -0.8 V to -0.3 V (blue) and -0.2 V to +0.5 V (red). Data on the *n*-slope and *p*-slope sides of the conductivity peak are presented in red/blue for both A) and B). C) Voltage-transfer characteristic (VTC) for amplifier circuit made using a pair of PANI-on-IDME OECTs connected by external wiring and operating with ionic liquid electrolyte. D) Corresponding voltage gain of the amplifier.



Figure S4. A and B) Transfer curves of the two PANI-based OECTs used to build the inverter circuit. Regions highlighted in yellow are the "active regions" we use in the circuit. The orange and green part of the curves are the "inactive regions". C) I_{DS} of each single OECT device plotted versus the input voltage V_{in} applied in the complementary circuit. The threshold voltages, $V_{Th\cdot n} = 0.15$ V and $V_{Th\cdot p} = 0.27$ V, were found to be centred about 0.20 V. The scheme in (C) represents the circuit featuring two identical PANI OECTs but with a V_{shift} applied to the *n*-like device. We label the device close to ground as the *n*-like device and the one close to V_{DD} as the *p*-like device. For the devices to operate in a complementary circuit, one device has to switch from 'on' to 'off' (*p*-like) and one device from 'off' to 'on' (*n*-like). However, the devices have the same electronic state initially because they are both made from PANI. To obtain one device operating as *n*-like and the other as *p*-like (regions highlighted in yellow in A and B), we apply a $V_{shift} = -0.4$ V on the *n*-like device in the complementary circuit. At $V_{in} = 0$ V, the *n*-like device is at V = -0.4 V and it is in the 'off' state, whereas the *p*-like device is at V = 0 V and it is in the 'on' state.



Figure S5. Plot of V_{out} (top) and V_{in} (bottom) vs time at $V_{\text{DD}} = +0.2$ V for square-wave input signals at 0.1 Hz for PANI-on-IDME circuit.



Figure S6. Ivium potentiostat (2) and Keithley 6517A electrometer (4) were used for measuring cyclic voltammetry. SMU Keithley 2401 (3) was used for applying V_{DS} and measuring I_{DS} . SMU Keithley 2401 (1) was used to measure the photodiode current. WE: working electrode; RE: reference electrode; CE: counter electrode.



Figure S7. The measurement were performed under LED illumination using as a source LED with $\lambda = 850$ nm. Positive and negative sweeps are presented by solid and dashed lines respectively. The blue lines define the three distinct regions of potential: Region I (-0.8 to -0.1 V), Region II (-0.1 to +0.7 V), and Region III (+0.7 to +1.0 V).



Figure S8. Different views of the polymers and simulation cell considered in the simulations are provided. The dominant interactions between the PANI and gold surface are hydrogen bonds of the Au^{...}H-X type, as it is shown by the equilibrium geometry that renders lowest energy.



Figure S9. A) The leucoemeraldine base is found to be insulator due to a wide band gap of 2.15 eV and position of the energy Fermi level just above the valence band edge. B) The emeraldine base is likely to be conductor under small electric bias due to the appearance of unoccupied electronic states at only ~ 0.1 eV above the valence band edge. C) The pernigraniline base is likely to be insulator under large enough electric bias due to the presence of a band gap of 1.55 eV just below the conduction band edge.



Figure S10. Plot of V_{out} (top) and V_{in} (bottom) vs time at $V_{DD} = +0.2$ V for square-wave input signals at 0.1 Hz for PANI-on-chitosan circuit.

C) A) B) -0.3 0.0 1.5 In (mA) Vous (V) -0.2 Vour (3) 1.0 -0.1 0.5 -0.1 0.0 0.0 -0.2 0.0 -0.8 -0.4 0.4 0.8 -0.2 0.0 0.2 0.4 -0.2 0.0 0.2 0,4 $V_{\rm G}(V)$ V_{in} (V) $V_{in}(V)$

Figure S11. A) Drain current I_{DS} vs gate voltage V_{GS} at drain-source voltage $V_{DS} = -0.1$ V. Black lines represent the leakage current. B) Voltage-transfer characteristic for amplifier circuit made on flexible chitosan film operating with aqueous NaCl electrolyte. C) Corresponding voltage gain of the inverter.