Supporting Information

Simple and rapid cleaning of graphenes with a 'bubble-free' electrochemical treatment

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Fig. S1 AFM topographic image (left, 5 μ m × 5 μ m), work function distribution (middle) taken simultaneously, and transport characteristics (right, at V_{DS} =100 mV) of SLGs treated by the following means: an additional treatment of SLG using an HCl-based cleaning solution (3.5%, 20 min, room temperature) after Cu etching with FeCl₃ (a), repeated and prolonged rinsing SLG in ultrapure DI water for more than 2 h (b), further rinsing SLG using acetone (20 min) after prolonged rinsing after PMMA removal and prolonged rinsing (c), and thermal annealing (300°C) for more than 2 h under a low vacuum (~1.3 × 10⁻² Pa) (d). The insets in the middle panels show the corresponding KPFM surface potential maps. Among these SLGs, only the thermally annealed SLG showed a reduced work function (~4.8 eV) and the appearance of the Dirac point, while the other SLGs exhibited high work functions (5.16~5.05 eV) without any signature of the Dirac point.



Fig. S2 Transport properties of a liquid-gate G-FET with the as-prepared SLG channel at $V_{DS} = 100 \text{ mV}$ for successive sweeping of the gate bias in the reverse direction from +0.8 to 0 V, then to -0.8 V (backward), and back to +0.8 V (forward). The liquid-gate G-FET presents an asymmetric I_{DS} - V_G curve for the first backward sweeping gate bias, showing a large shift of the Dirac point ($V_{G,Dirac} \sim 0.60 \text{ V/V}_{Ag/AgCl}$). Abrupt changes in the current curve after passing $V_G = 0 \text{ V}$ for the first backward sweeping gate bias appear to be due to the EC-cleaning effect in the negative-voltage region.

Device fabrication process

To fabricate organic light-emitting devices (OLEDs) with the SLGs studied here, the light-emitting polymer Super Yellow (SY, catalogue No. PDY-132, Merck)⁶⁴, was used as received. The OLEDs in this study were fabricated as follows. First, a transferred SLG on a glass substrate was used as the transparent anode. On the SLG anode, a solution of SY dissolved in toluene was directly spin-coated to form an emissive layer (EML, 70 nm thick). Next, Cs_2CO_3 (2 nm) / Al cathode (50 nm) layers were formed on the SY EML by thermal deposition (0.1 nm s⁻¹) at a base pressure of less than 2.7×10^{-4} Pa. The OLED fabricated thus had a device configuration of [SLG anode / SY EML / Cs_2CO_3 / Al cathode], and the active area of the fabricated device was 8 mm². All fabricated OLEDs were then tested under ambient conditions without encapsulation. A luminescence and colour meter (Chroma Meter CS-200, Konica Minolta Sensing, Inc.) and a source meter (Keithley 2400) were used to measure the electroluminescent characteristics.

To fabricate organic photovoltaic (OPV) cells with the studied SLGs, PV materials of the low-bandgap electrondonating polymer poly[[9-(1-octylnonyl)-9H-carbazole-2,7-diyl]-2,5-thiophenediyl-2,1,3-benzothiadiazole-4,7-diyl-2,5thiophenediyl] (PCDTBT, Lumtec, HOMO level: 5.5 eV)^{65,66} and the electron-accepting material [6,6]-phenyl C₇₁ butyric acid methyl ester (PCBM₇₀, Lumtec) were used as received. The OPV cells investigated were fabricated as follows. First, SLG transferred on glass was used as the transparent anode. The SLG anode was then pre-coated with a poly(3,4ethylenedioxythiophene):poly(styrene sulfonate) (PEDOT:PSS, CLEVIOSTM 4083, H. C. Starck Inc., 30 nm) layer by spincoating and a molybdenum oxide (MoO₃, Aldrich, thickness: 12.5 nm) layer by thermal evaporation (0.05 nm s⁻¹) at a base pressure of less than 2.7×10^{-4} Pa to form stacked hole-collecting layers (HCLs). A blended solution of PCDTBT (0.456 wt%) and PCBM₇₀ (1.824 wt%)^{65,66} in monochlorobenzene was spin-coated onto the stacked HCLs of PEDOT:PSS/MoO₃ to fabricate a bulkheterojunction (BHJ) PV layer. The PCDTBT:PCBM₇₀ BHJ PV layer was approximately 85 nm thick. Details of the preparation of the PCDTBT:PCBM₇₀ layer are similar to those described in earlier experiments.⁶⁶ Next, LiF (0.5 nm) / Al cathode (50 nm) layers were formed on the PCDTBT:PCBM₇₀ PV layer by thermal deposition (0.05 nm s⁻¹) at a base pressure of less than 2.7×10^{-4} Pa. The structure of the OPV cells was therefore [SLG anode/ PEDOT:PSS / MoO₃ / PCDTBT:PCBM₇₀ / LiF / Al cathode]. The active area of the fabricated device was 8 mm². The PV performance was measured using a source meter (2400, Keithley) and calibrated using a reference cell (BS-520, Bunkoh-Keiki) under illumination of 100 mW cm⁻² produced by an AM 1.5G light source (96000 Solar Simulator, Newport). For comparative purposes, we also fabricated a reference OPV cell using a cleaned ITO anode with a structure of [ITO / PEDOT:PSS (40 nm) / PCDTBT:PCBM₇₀ / LiF / Al]. Apart from the differences in the anodes and HCLs described, the reference OPV cell was fabricated using a method identical to that used for the sample OPV cell with the SLG anode.