Electronic Supplementary Material (ESI) for Journal of Materials Chemistry C

Supporting Information

Soft Lithography of Graphene Sheets via Surface Energy Modification

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Synthesis and Transfer of Graphene Sheets: Graphene sheets were synthesized on 100-μm thick Cu foil (from Alfa Aesar) by thermal chemical vapor deposition (CVD). Copper foil was inserted in a quartz tube and heated to 1000°C under ambient pressure with flowing H_2 and Ar. After flowing reaction gas mixtures (CH_4 : H_2 : Ar = 30 : 10 : 1000 sccm) for ~ 20 min, the sample was cooled to room temperature.[S1] Large-scale, single layer graphene was spin-coated on the Cu foil by PMMA, and the Cu foil was removed by the copper etchant (dilute $FeCl_3$ solution in deionized water). After rinsing several times with deionized water to remove residual Cu etchant impurities, the PMMA-coated graphene was transferred to various substrates, such as SiO_2 (300 nm), glass, and PET. PMMA was carefully removed by acetone. The qualities of the CVD-grown graphene were monitored by Raman spectroscopy and optical microscopy. In our experimental conditions, monolayer graphene was dominantly synthesized, which was confirmed by I_{2D}/I_G ratio.

Computational Details of the Work of Adhesion: The work of adhesion of a molecule is calculated by the binding energy of the molecule on a single layer of graphene divided by the surface area. The coverage-dependent binding energy of a molecule is obtained by simulating single molecule adsorption on the graphene layer with variable surface area (from 1x1 to 3x3 unit of graphene, from 5.25 $Å^2$ to 47.27 $Å^2$). The binding energy is calculated by taking the total energy difference between most stable molecular adsorbed configuration and separate energies.

The graphene layer was modeled with a periodically repeated slab of hexagonal carbon lattice separated by a vacuum width of 30 Å. Spin-polarized DFT calculations have previously been performed using the periodic plane-wave approach as implemented in the VASP[S2,S3] within the generalized gradient approximation (GGA) using the Perdew-Burke-Ernzerhof (PBE) exchange-correlation functional.[S4] Frozen-core projector augmented wave pseudopotentials[S5,S6] were used. The plane wave basis set was restricted by a cutoff energy of 400 eV. Total energies are converged to 10^{-6} eV. All structures were fully optimized with the largest residual force is smaller than 0.01 eV/Å. In VASP modeling of polar molecules, the dipole correction[S7] was used because the existence of an electric dipole moment. The correction removes the artificial field arising in the empty space from the use of Periodic Boundary Conditions (PBC) in the solution of the Poisson equation by the Fast Fourier Transform (FFT) method.

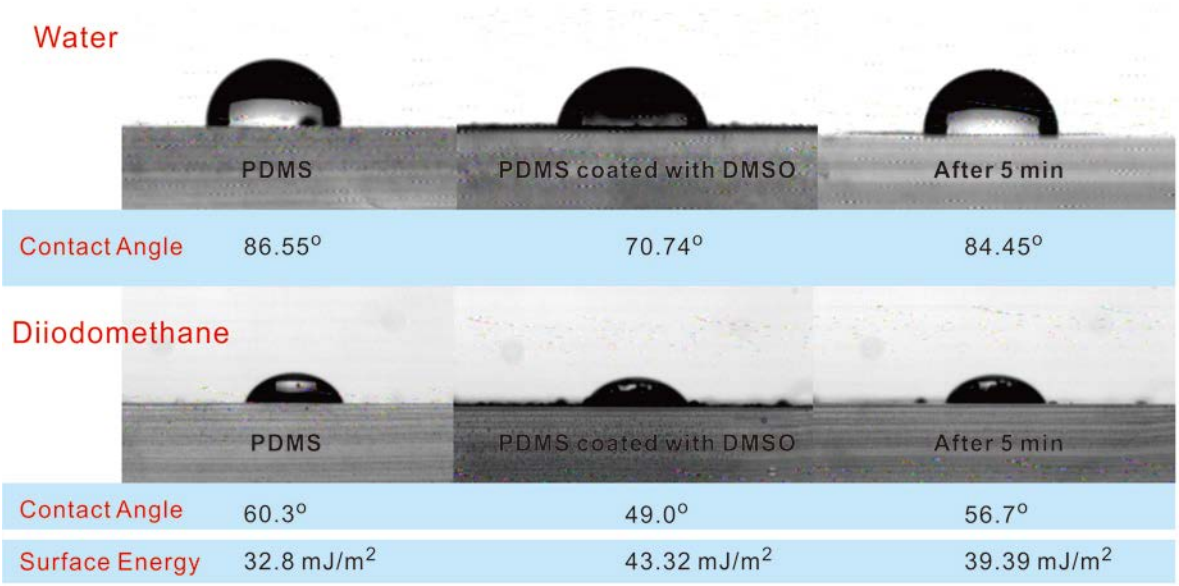

Figure S1 Contact angle and calculated surface energy of PDMS and PDMS coated with DMSO. The surface energy was calculated from Owen-Wendt model based on the contact angle measurement of water and diiodomethane.

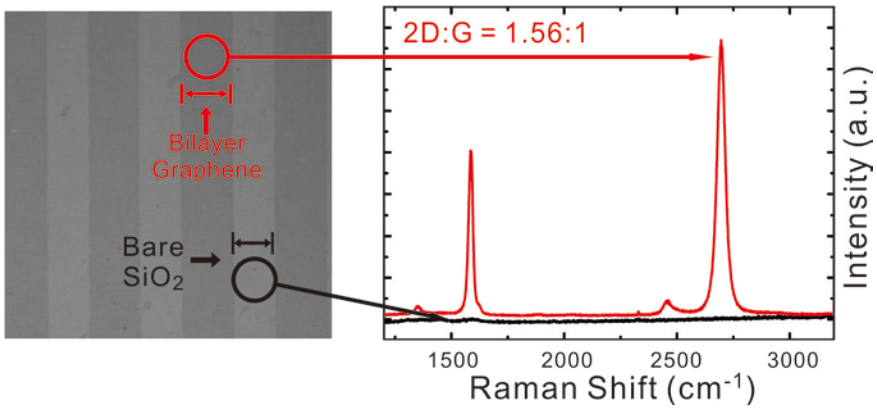

Figure S2 Optical microscope images (left) and Raman spectroscopy (right) of bilayer graphene patterns. The darker area shows a bilayer graphene area and the brighter region is a bare SiO_2 surface. The red and black curves in right graphene show the Raman spectroscopy of bilayer graphene area and bare SiO_2 surface, respectively.

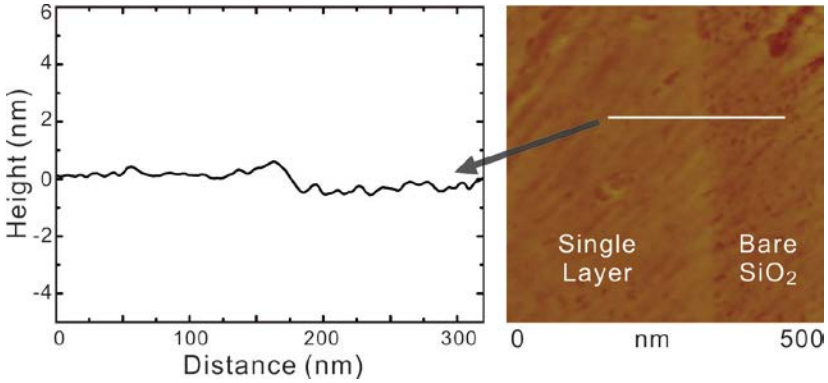

Figure S3 The height profile (left) and AFM image (right) of the single layer graphene sheets after the patterning procedure

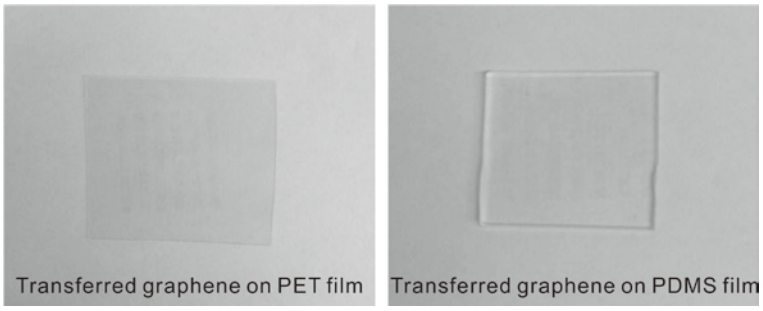

Figure S4 The optical images of the transferred graphene electrodes on the PET (left) and PDMS (right) film.

Graphene and Graphene (E_{GR-GR})	Graphene and DMSO ($E_{GR-DMSO}$)	PDMS and DMSO ($E_{PDMS-DMSO}$)	Graphene and Water ($E_{GR-Water}$)	Graphene and SiO_2 ($E_{GR-SiO2}$)	Graphene and PSMS ($E_{GR-PDMS}$)
~22.4 meV/Å^2	~11.7 meV/Å^2	~7.9 meV/Å^2	~5.6 meV/Å^2	~4.3 meV/Å^2	~2.5 meV/Å^2

Table S1. Interlayer interaction

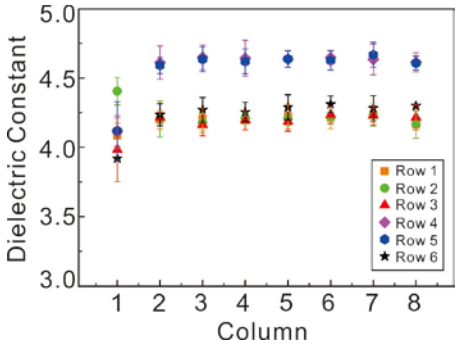

Figure S5 Distribution of dielectric constants of the graphene-insulator-graphene capacitors in each column.

Reference

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