

SUPPLEMENTARY INFORMATION

**Contact angle measurement of freestanding square millimeters of single
layer graphene**

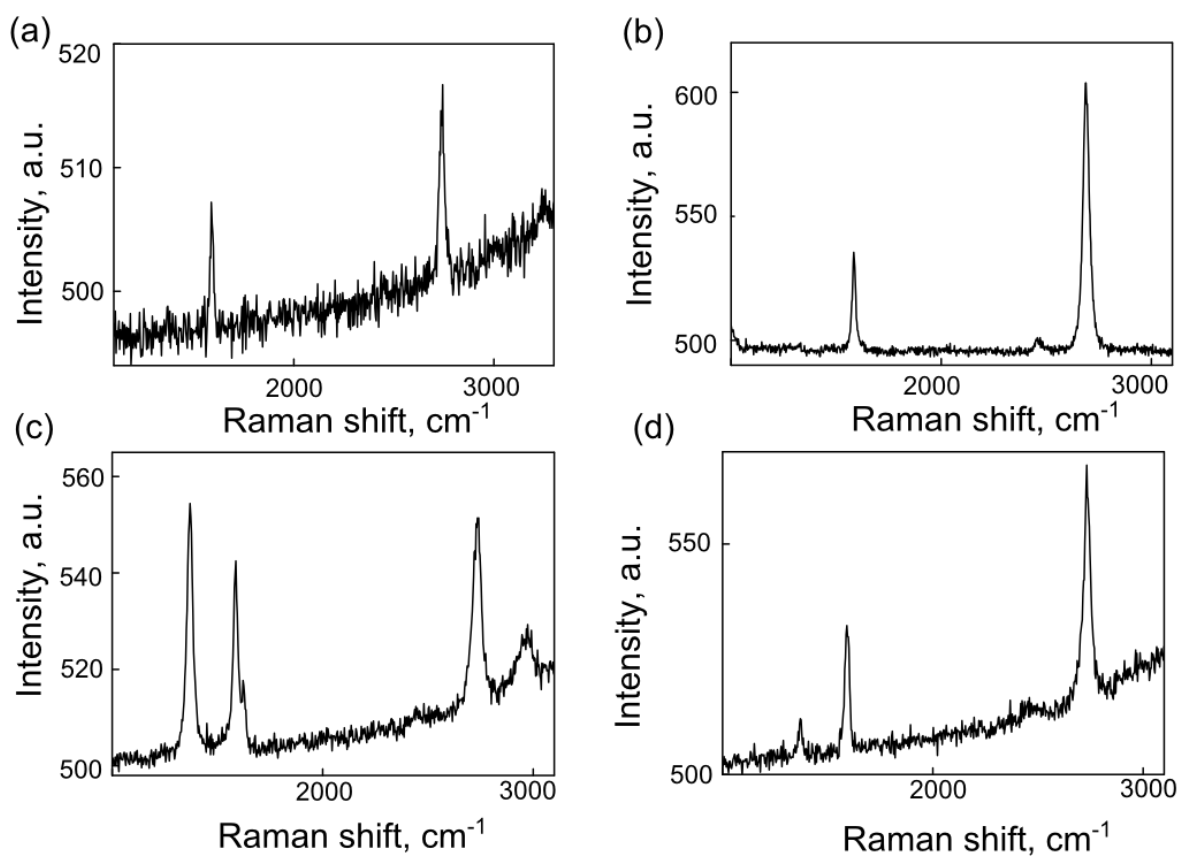
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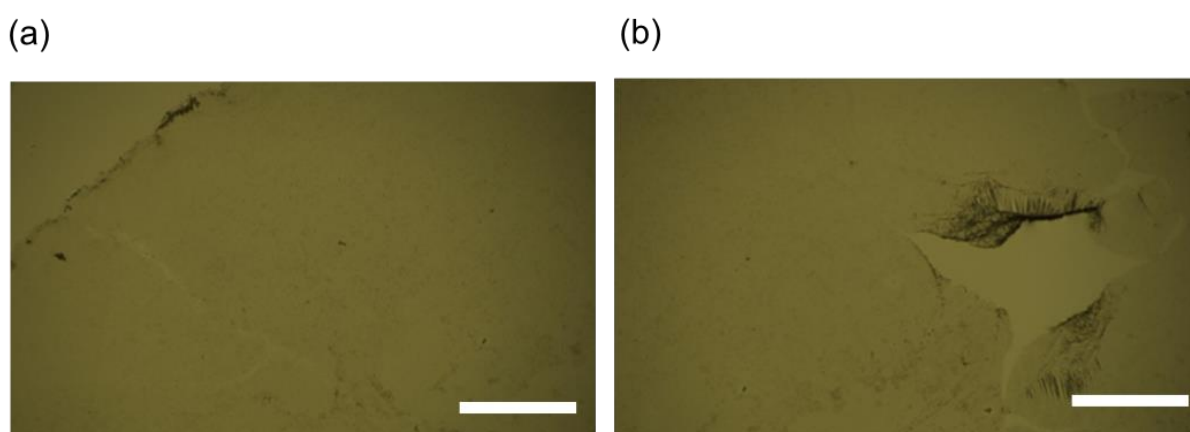
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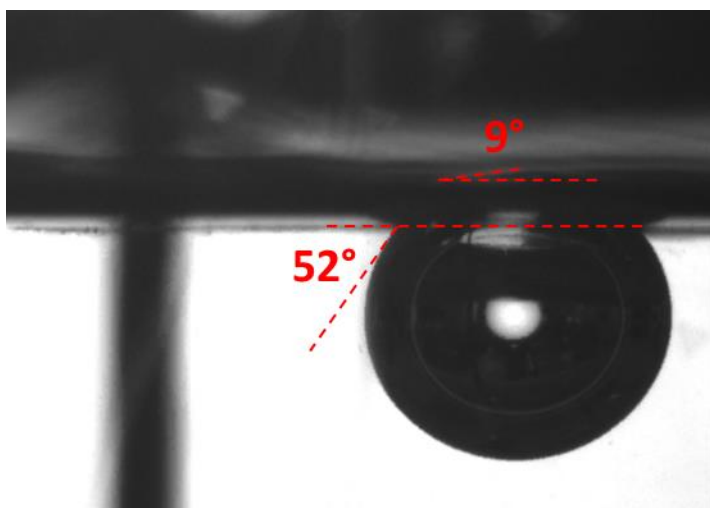
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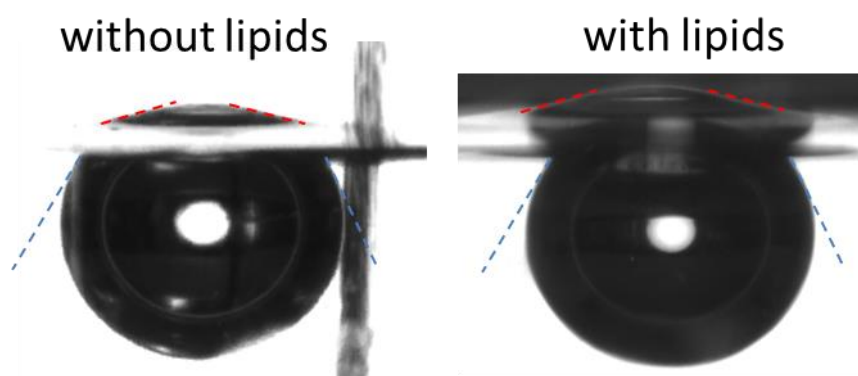
Supplementary Figure 1. Raman spectra of CVD (chemical vapor deposition) graphene before and after plasma modification. (a) Non-treated graphene on Cu after the growth. (b) Non-treated graphene transferred onto a Si/SiO₂ wafer. (c) Graphene on Cu after H₂ plasma treatment. (d) Graphene on Cu after O₂ plasma treatment.



Supplementary Figure 2. Optical images of graphene floating on the surface of water. (a) Before captive bubble measurement. The scale bar represents 500 μm . (b) After captive bubble measurement. The scale bar represents 500 μm .



Supplementary Figure 3. Contact angle of graphene on the surface of 0.1 M aqueous solution of FeCl_3 . Presence of ions in concentrations below 0.3 M does not affect the measured contact angle, the contact angles of graphene in different etchant solutions are equal to the contact angle of graphene in pure water.



Supplementary Figure 4. Captive bubble on graphene with (right) and without (left) a DPPC (1,2-dipalmitoyl-sn-glycero-3-phosphocholine) lipid scaffold. Lipids do not affect the contact angle value of $42^\circ \pm 3^\circ$.

Supplementary Note 1

Calculation of surface free energy

Surface energies of bare PMMA (poly(methyl methacrylate)) and PMMA-coated graphene samples were calculated according the Owens-Wendt model¹. Based on the contact angle measurements with

liquids of different polarities, the Owens-Wendt equation allows for the determination of total surface energy of a solid and its polar and dispersive components:

$$\gamma_{lv}(1 + \cos \theta) = 2(\sqrt{\gamma_s^d \gamma_l^d} + \sqrt{\gamma_s^p \gamma_l^p})$$

The polar and dispersive components of liquids γ_L^P and γ_L^D were determined by measuring contact angles (sessile drop method) and applying the Owens/Wendt Theory for PTFE (teflon), which is a solid with known polar and dispersive components of the surface tension ($\gamma_s^P=0 \text{ mN m}^{-1}$, $\gamma_s^D=18 \text{ mN m}^{-1}$). The determined surface tensions and their components of all used liquids are listed in Table 1.

Then contact angles of bare and PMMA-coated graphene with the liquids listed in Table 1 were

measured. The results were plotted as $\frac{\gamma_L(\cos\theta+1)}{2\sqrt{\gamma_L^D}}$ versus $\frac{\sqrt{\gamma_L^P}}{\sqrt{\gamma_L^D}}$ for each substrate and the dependences

were fitted linearly. The slope of the plot equals $\sqrt{\gamma_S^P}$ and the intercept equals $\sqrt{\gamma_S^D}$. The squares of the latter two equal γ_S^P and γ_S^D respectively. The resulting surface tensions and their polar and dispersive components are presented in Supplementary Table 2 below and charted in Figure 1c of the main text.

Supplementary Table 1. Calculated surface tensions, polar and dispersive components of tested liquids

| Liquid | γ_L^P , mJ m ⁻² | γ_L^D , mJ m ⁻² | γ_L^{total} , mJ m ⁻² |
|----------------------|-----------------------------------|-----------------------------------|--|
| Water | 51 | 21.8 | 72.8 ± 2.4 |
| Ethylene glycol | 19.2 | 28.8 | 48.0 ± 1.9 |
| 10% Ethanol in water | 36.1 | 23.9 | 60.0 ± 2.2 |
| Diiodomethane | 0 | 50.8 | 50.8 ± 2.3 |
| 1-Methylnaphthalene | 0 | 42.0 | 42.0 ± 1.1 |

Supplementary Table 2. Calculated surface tensions, polar and dispersive components of PMMA, freshly PMMA-coated graphene and PMMA-coated graphene aged for six days

| Surface energy/Sample | PMMA | Graphene on PMMA | Graphene on PMMA after 6 days |
|--|----------|------------------|-------------------------------|
| γ_s^P , mN m ⁻¹ | 9.8±1.6 | 10.8±1.5 | 9.1±1.4 |
| γ_s^D , mN m ⁻¹ | 41.9±1.3 | 39.1±2.1 | 30.6±2.4 |
| γ_D^{total} , mN m ⁻¹ | 51.6±2.3 | 47.9±1.6 | 39.7±1.5 |

Supplementary References

- Owens, D. K. & Wendt, R. C. Estimation of the surface free energy of polymers. *J. Appl. Polym. Sci.* **13**, 1741–1747 (1969).