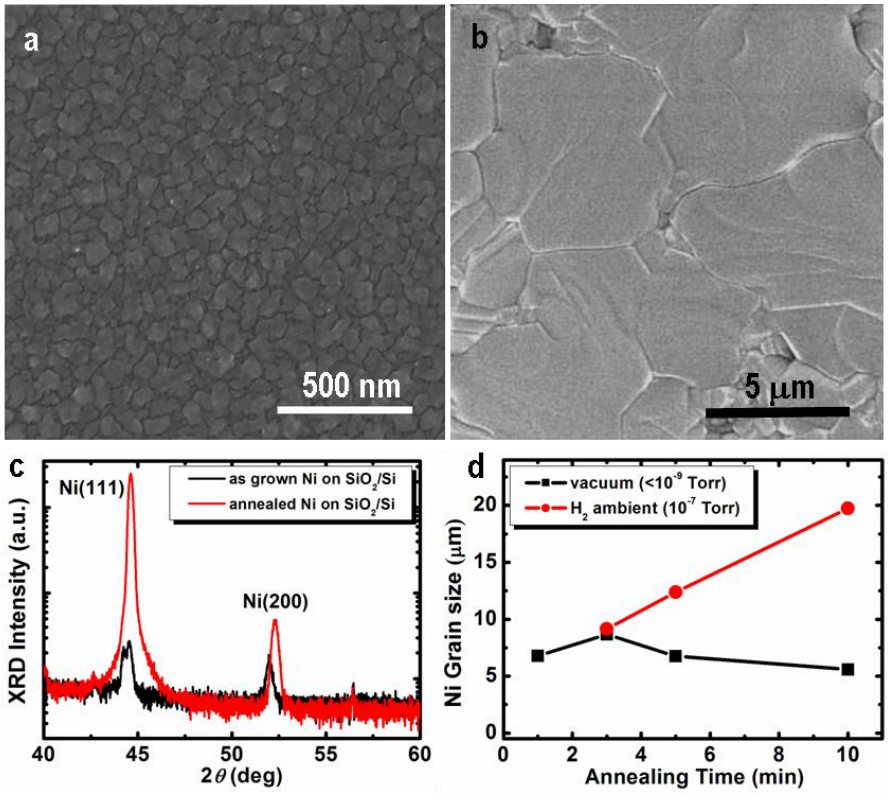
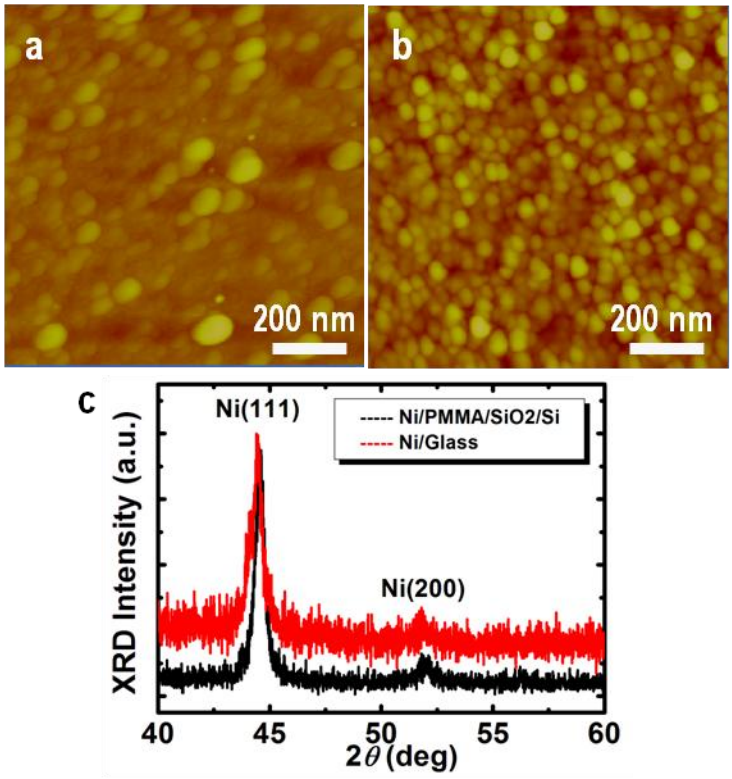
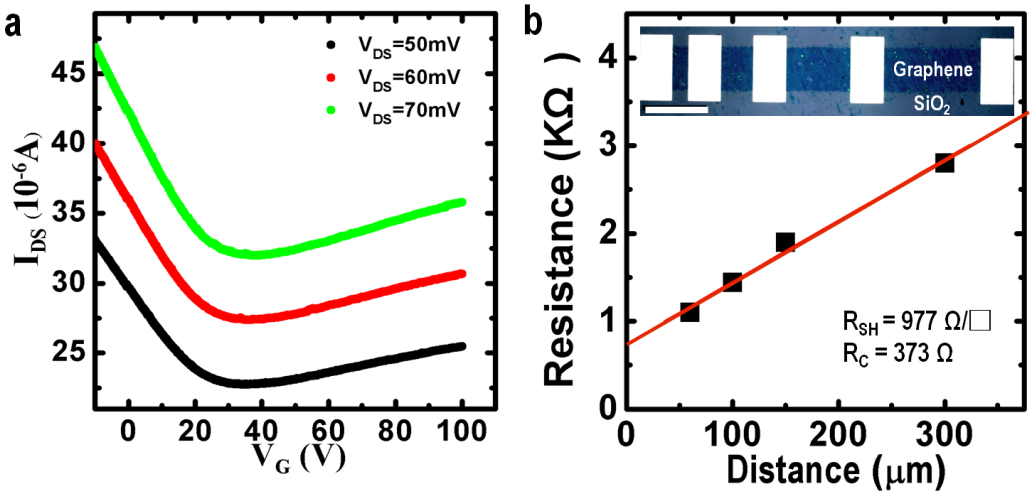
Supplementary Figures



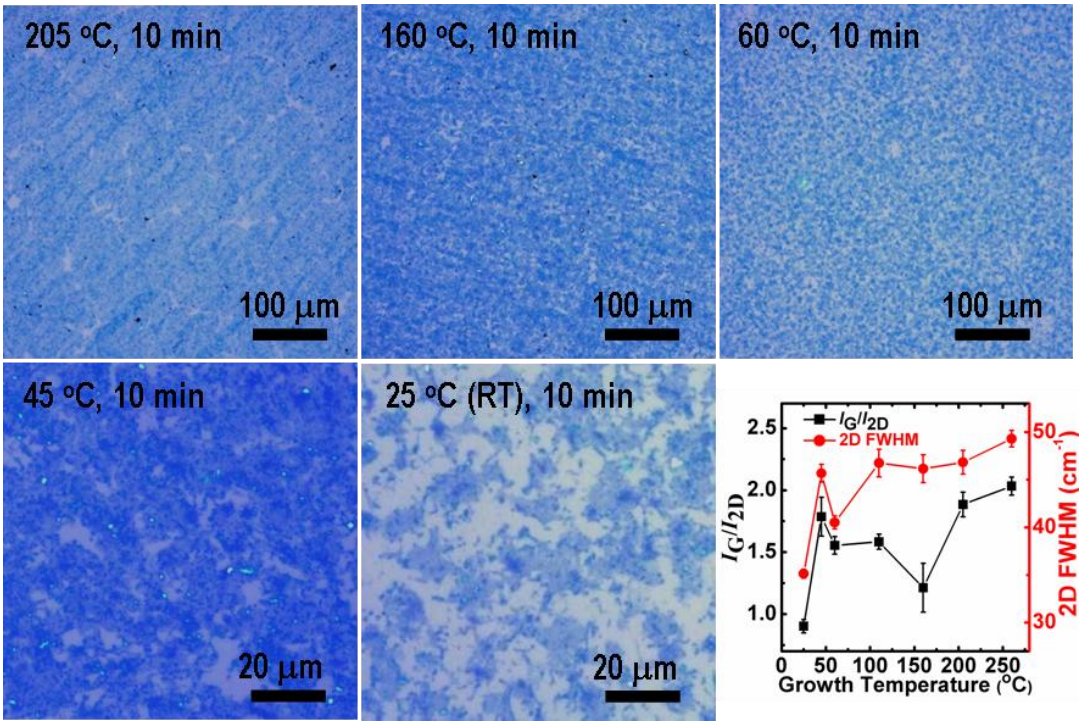
Supplementary Figure S1. Effect of annealing conditions on the microstructure of 100-nm-thick poly-Ni films on SiO_2(300 nm)/Si substrates. **a,** SEM image of poly-Ni film deposited at 400 ^{o}C. **b,** SEM image of poly-Ni film annealed at 1,000 ^{o}C for 10 min in a H_2 ambient. **c,** Semi-log plot of XRD data of poly-Ni films before and after annealing. **d,** Effect of time and ambient on grain size of poly-Ni films annealed at 1,000 ^{o}C.



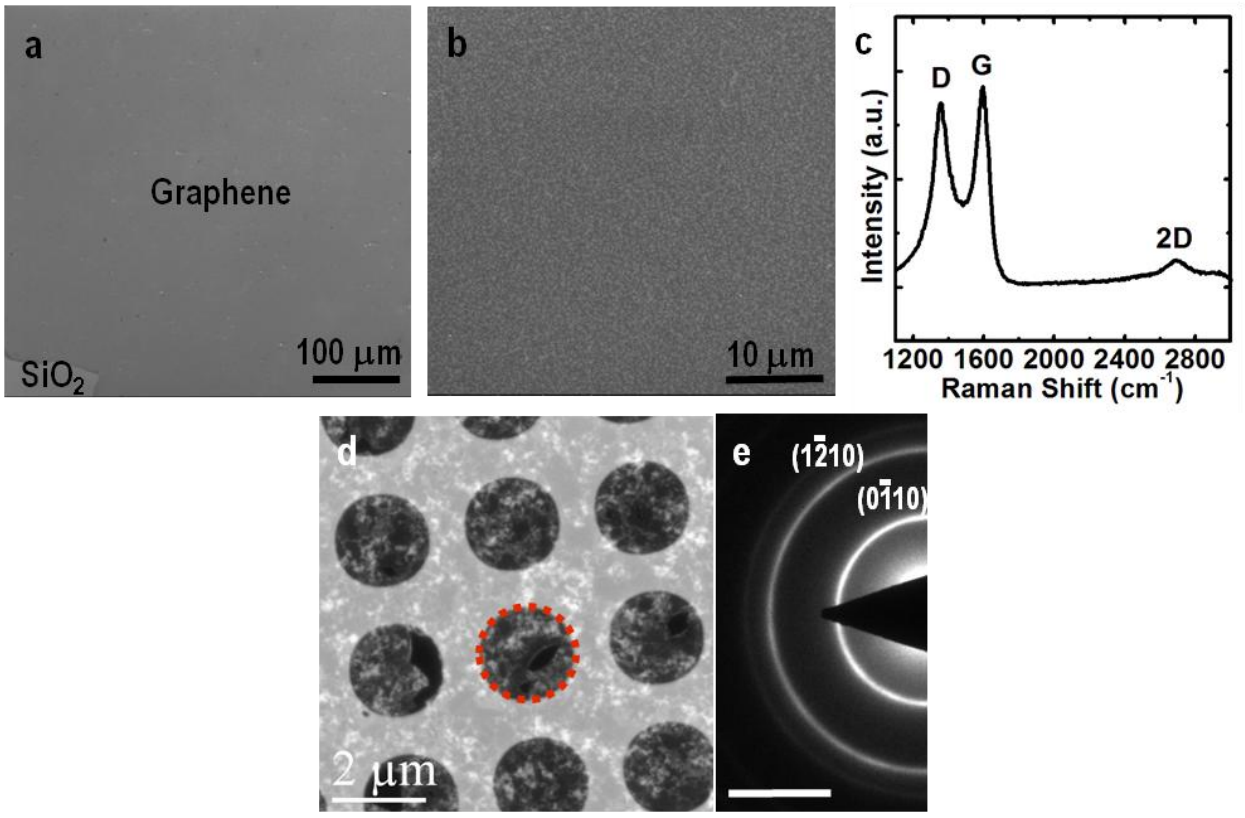
Supplementary Figure S2. Morphology and crystallinity of Ni thin films deposited at room-temperature. AFM images (1 μm × 1 μm) of 100-nm-thick poly-Ni films deposited at room temperature on **a,** 1.5-μm-thick spin-coated PMMA/SiO_2(300 nm)/Si and **b**, glass substrates. The average grain sizes of Ni on PMMA and glass are 50.8 nm and 43.7 nm, respectively. **c,** XRD data of the as-grown 100-nm-thick poly-Ni films on these substrates.



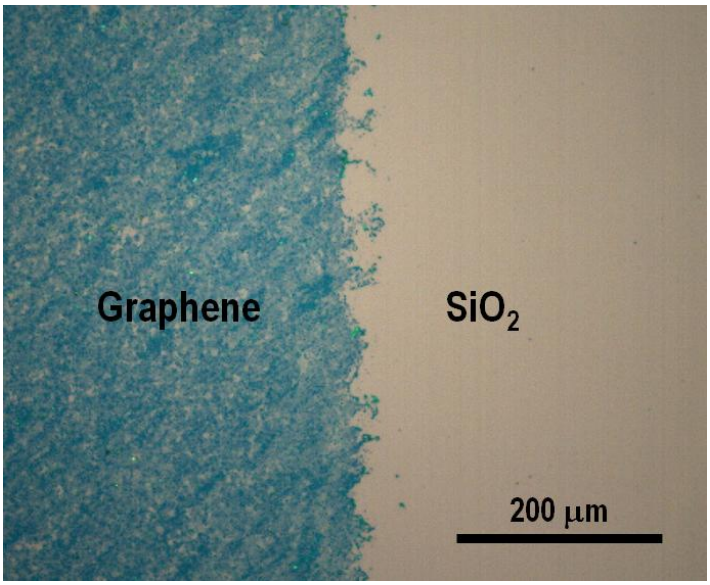
Supplementary Figure S3. Representative **a,** room-temperature I_{DS}-V_G curves from a DAS-grown graphene FET atop 300 nm SiO_2 with highly doped p^{++} Si back gate, depending on V_{DS}. Mobilities of ~660-670 cm^2/Vs at room temperature are achieved. **b,** The plot shows total resistance of the TLM structure as a function of distance. The distances between the contacts on the TLM structure are 60, 100, 150, and 300 μm, respectively. From the slope, we extract a sheet resistance value of ~1,000 Ω/sq. The inset shows an optical microscopy image of patterned graphene layer (140 μm × 1.1 mm) on SiO_2 with Cr(10nm)/Au(60nm) bilayer as Ohmic contacts. The scale bar in the inset is 200 μm.



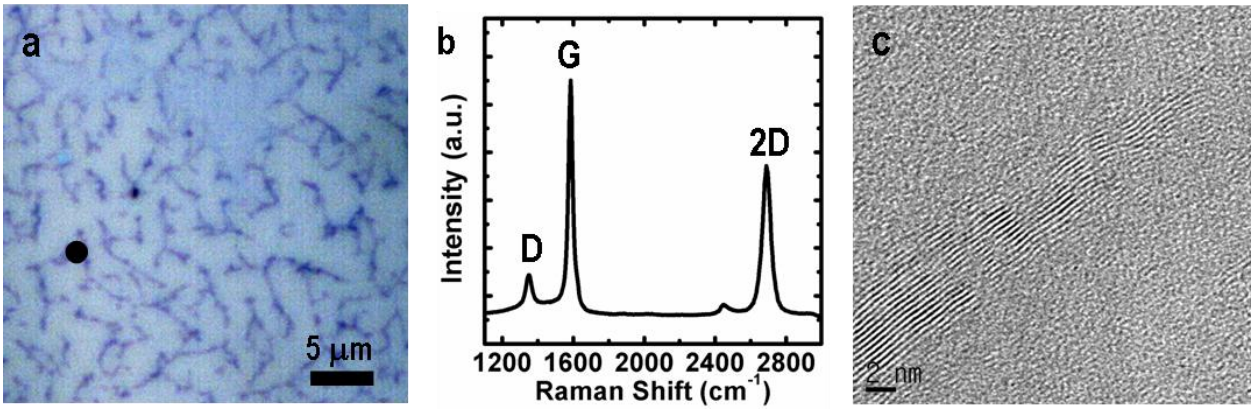
Supplementary Figure S4. Optical microscopy images and Raman spectroscopic characterizations of G-to-2D intensity ratio (I_G/I_{2D}) and FWHM of 2D bands of few-layer graphene films grown for 10 min on SiO_2(300 nm)/Si substrates, as a function of growth temperature *T*. Continuous graphene layers over large areas can be obtained at all $T \geq 160$ °C.



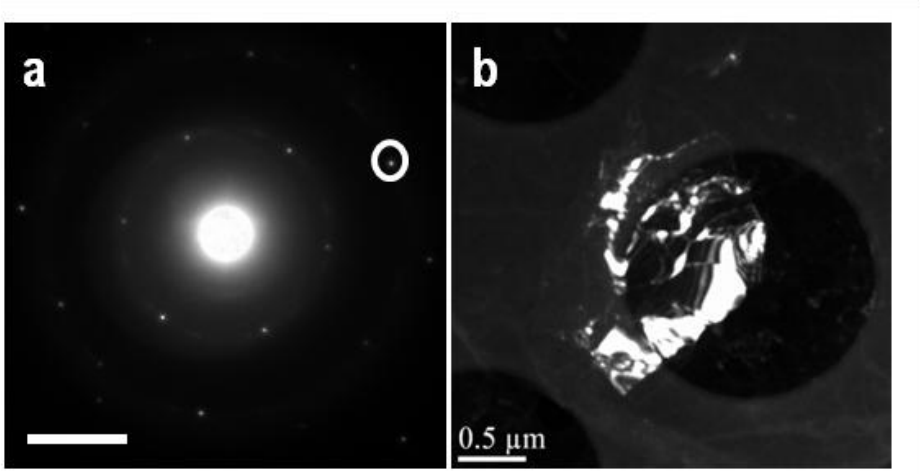
Supplementary Figure S5. Nanocrystalline graphene films grown at $T \geq 460$ ^{o}C on SiO_2/Si substrates. Representative **a,** SEM image of graphene grown at $T = 465$ ^{o}C for 5 min on SiO_2/Si substrate. **b,** Higher magnification SEM image of **a**, showing the presence of high density of grains with lateral sizes of several tens of nanometers. The layer is completely free of graphene ridges. **c,** Typical Raman spectra of graphene grown at $T = 465$ ^{o}C for 5 min on SiO_2. **d,** Typical low magnification plan-view TEM image of graphene grown at $T = 465$ ^{o}C for 5 min on SiO_2 then transferred onto a TEM support grid. **e,** Selected area diffraction pattern from red dotted circle in **d**, which displays continuous ring patterns. The two diffraction rings correspond to graphene crystal planes (0$\underline{1}$10) (interplanar spacing of 0.213 nm) and (1$\underline{2}$10) (interplanar spacing of 0.123 nm), consistent with earlier report (ref. 39). Scale bar is 5 1/nm.



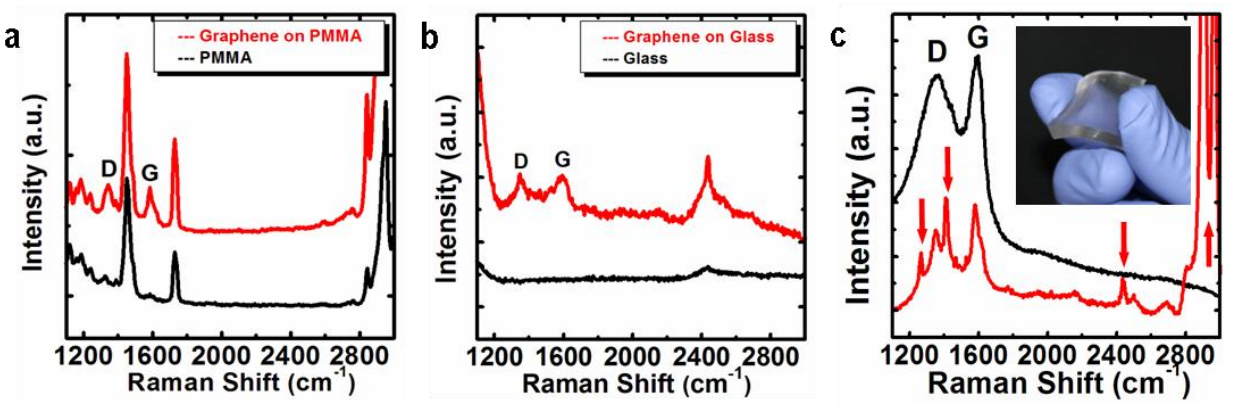
Supplementary Figure S6. Optical microscopy image showing the effect of hydrogen during annealing of poly-Ni films on graphene grown by DAS process. During thermal annealing process of poly-Ni film on SiO_2/Si substrates in a H_2 ambient (P ~ 10^{-6} Torr), half of the sample is covered by a molybdenum-made sample holder (left) and remaining part of the sample is directly exposed to H_2 flow (right). After DAP process at 45 ^oC for 10 min, graphene did not grow in regions directly exposed to H_2 flow, possibly due to Ni catalyzed methanation of the carbon atoms (refs. 24, 40). This phenomenon was observed in all samples, irrespective of the DAS process conditions.



Supplementary Figure S7. Formation of graphene ridges on a SiO_2(300 nm)/Si substrate. **a,** Optical microscopy image of graphene-free surface with traces of graphene ridges in the sample grown at T = 160 ^{o}C for 2 min on SiO_2. The morphology of graphene ridges reflects the grain boundaries in poly-Ni films. **b,** Typical Raman spectra acquired from a ridge highlighted by a black dot in **a**, showing the D, G, and 2D peaks at peak positions expected for graphene. **c,** Cross-sectional TEM image of a ridge structure formed in a sample grown at T = 160 ^{o}C for 10 min on SiO_2, showing an average interlayer spacing of 3.3-3.4 Å similar to that of multilayer graphene.



Supplementary Figure S8. Characterization of graphene grown at T = 160 ^{o}C on SiO_2 using dark-field TEM. **a,** Selective area diffraction pattern taken from graphene film using 300 nm diameter aperture, showing a single hexagonal diffraction pattern. This reveals that the grain size is larger than 300 nm. Scale bar is 10 1/nm. **b,** a dark-field image obtained using a g(1$\underline{2}$10) spot (white circle in **a**) shows the real-space shape of these grains. The observed contrast variations in the image are due to wrinkles and tears in graphene film formed during transfer process.



Supplementary Figure S9. Raman spectra of as-synthesized graphene films on **a,** PMMA and **b,** glass substrates. **c,** Raman spectra of graphene film on PDMS before (lower red curve) and after (upper black curve) transfer onto SiO_2(300 nm)/Si substrate. The red arrows in Raman spectra are from local vibration modes of PDMS substrate. The inset shows a photograph of graphene film grown on 4-mm-thick PDMS. All samples were grown at T = 60 ^{o}C for 10 min. The Raman spectra of as-synthesized films were much noisier than those for graphene on SiO_2 due to the strong interaction between graphene and substrate (ref. 41).

Supplementary Methods

Supplementary Method 1. Plan-view transmission electron microscopy (TEM) analysis of graphene films grown on SiO_2/Si substrates

We used a wet-transfer TEM sampling method for plan-view imaging of graphene films grown on SiO_2 to minimize damage and/or contamination of the films. As a first step to transfer the graphene films, Au Quantifoil TEM grids were placed on the graphene covered SiO_2/Si substrates. To ensure efficient contact between the Quantifoil and graphene/substrate couples, isopropyl alcohol (IPA) dropped on top of the grids and the couples were heated on a hot plate at 120 ^{o}C for 10-20 min to evaporate any remaining IPA. Then, the couples were dipped in a BOE solution to etch the SiO_2 layer until the TEM grids floated free off of the substrate. Finally, the Quantifoil-graphene couples were rinsed sequentially with DI water and IPA and dried in air.

Dark-field TEM imaging was performed at 200 keV on a JEOL JEM-2100F TEM. An objective aperture was placed over one of the diffracted beams, and the dark-field image was acquired using an exposure time of 2-5 sec. A Gatan Multiscan CCD camera was used for the acquisition of images with pixel size of 1024 × 1024.

In the TEM, wrinkles or tears in the graphene films were routinely observed. We would, however, like to note that this transfer method can be further optimized to improve the yield of wrinkle- and tear- free graphene layers upon transfer from the growth substrate to the TEM grid.

Supplementary Method 2. Transfer of graphene films from plastics on arbitrary substrates for evaluation.

We develop a wet-transfer process for the graphene film grown on PMMA. In general, ~1-2 μm-thick PMMA films were spin-coated on SiO_2(300 nm)/Si substrates. Then, the graphene films

were grown on PMMA using DAS process. In case of graphene/PMMA film (or the PMMA film), the samples were dipped in 10% diluted HF solution to etch SiO_2 layer and the floating graphene/PMMA film (or the PMMA film) transferred onto arbitrary substrates. For transferring only the graphene film, we made a transfer couple by attaching graphene/PMMA/SiO_2/Si sample to another SiO_2(300 nm)/Si substrate and placed the couple in acetone. The PMMA can be etched away using acetone, leaving behind graphene film on another SiO_2/Si substrate.

To transfer graphene film grown on PDMS, we made a transfer couple by attaching graphene/PDMS sample to SiO_2(300 nm)/Si substrate and then pressed gently so as to apply a constant pressure. After detaching the couple, transferred graphene films can be observed on SiO_2(300 nm)/Si substrate. In this dry-transfer process, the transfer yield is strongly dependent on the rigidity of PDMS.

We would like to note that these transfer methods can be further optimized to improve the transfer yield of graphene layers from the growth substrate to other medium. In our experiments, we found that the strong interaction between the graphene films and the underlying substrates resulted in low transfer efficiency.

Supplementary References

39. Meyer, J. C. *et al*., The structure of suspended graphene sheets. *Nature* **446**, 60-63 (2007).

40. Tomita, A. and Tamai, Y., Hydrogenation of carbons catalyzed by transition metals. *J. Catal*. **27**, 293-300 (1972).

41. Calizo, I. *et al*., The effect of substrates on the Raman spectrum of graphene: Graphene-on-sapphire and graphene-on-glass. *Appl. Phys. Lett*. **91**, 201904 (2007).