Supporting Information

Controllable Atmospheric Pressure Growth of Mono-layer, Bi-layer and Tri-layer Graphene

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Experimental Section

CVD Growth: A 1.5×6.0 cm² strip of Cu foil (25µm thick and 99.8% purity, purchased from Alfa Aesar) was pre-cleaned in ~10% HCl/H₂O solution and inserted into the corundum tube inside a horizontal furnace. After pre-growth heating and annealing in H₂/Ar at 1000°C, the temperature and gas flow rate were adjusted to desired values in 5 minutes. Next, desired layer number of graphene nuclei was formed with a low concentration of dilute methane (0.05% methane in Ar) for 10 minutes, and then the flow rate of methane was increased to enlarge the graphene nuclei size for 15 minutes. The substrate was fully covered with graphene by decreasing the Ar flow rate and prolonging growth time. After growth, the furnace cooled down quickly to room temperature.

Graphene Transfer: The transfer of graphene films onto SiO_2 or quartz substrate was performed by the wet-transfer method as reported elsewhere.[1] In general, one side of the graphene/Cu surface was spin-coated with PMMA (Poly (methyl methacrylate), 950 PMMA A4, MicroChem) and baked at 120°C for 1 min. The other side graphene was removed by O-plasma. 0.1M $(NH_4)_2S_2O_8$ (Sigma Aldrich) solution was used to etch Cu foil overnight. After washed by floated on deionized water for several times, the PMMA/Graphene film was pick up by target substrate, and dried in vacuum. Finally, the PMMA layer was dissolved in hot acetone for 1hour.

Graphene FET: The bi-layer graphene was transferred onto n-Si substrate with 300nm SiO_2 and patterned by O-plasma and e-beam lithography. The source and drain electrodes (Au thickness of 300nm) were deposited using e-beam evaporation. The channel length was 5µm. The carrier mobility was calculated from the I_{DS}-V_{GS} curve as:

$$g_m = \frac{\partial I_D}{\partial V_{bg}} = \frac{W}{L} \mu C_{bg} V_D \Rightarrow \mu = \frac{L \times g_m}{W C_{bg} V_D}$$

Where µ is the carrier mobility, W and L are the FET width and length, respectively, C_{bg} is the gate capacitance per unit area and V_D is the source-drain voltage. As for the dual-gate device, the top-gate dielectrics (aluminum layer of 30 nm thickness) and top gate electrode (Au layer of 50nm) were fabricated using e-beam lithography and e-beam evaporation.[2]

Characterizations: The graphene morphology was characterized with field-emission scanning electron microscopy (SEM, JEOL 6701F, 5kV) on Cu, The structure characterization was performed by Raman spectroscopy (Thermo Scientific DXR, 532 nm laser wavelength) on SiO_2/Si substrate and field-emission transmission electron microscopy (TEM, JEOL 2011F, 200kV). The transmittance spectroscopy was collected by a UV-vis spectrophotometer (Shimadzu UV-2550) on quartz slide. The graphene structure and thickness was studied by atomic force microscopy (AFM, Veeco Multimode).
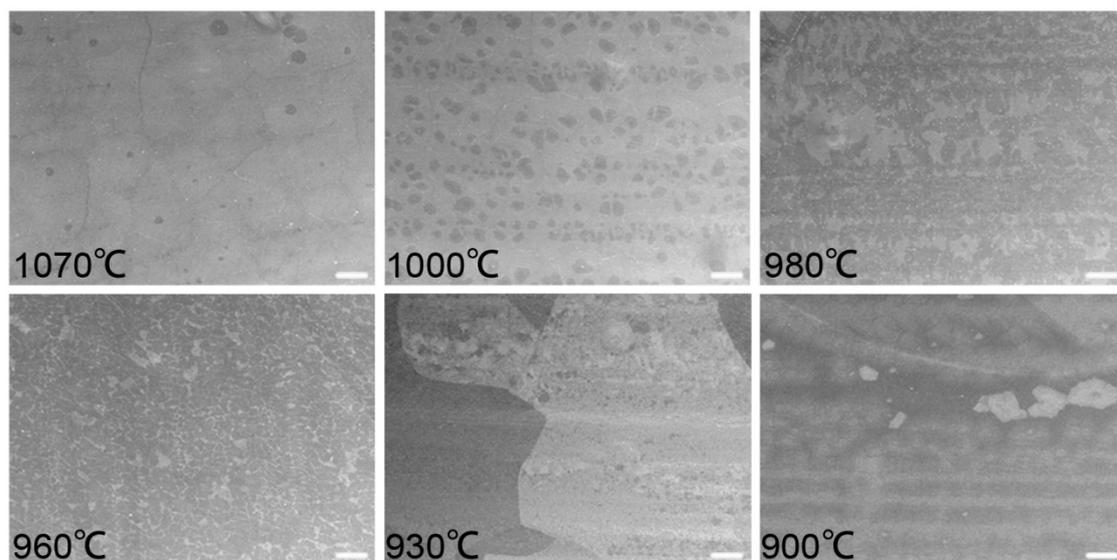
Figure S1. Growth temperature dependent graphene morphology. SEM images of graphene samples obtained under various growth temperature. All of the initial total gas flow rate are 1000 sccm. The growth temperature is labelled in each image. Since the graphene growth rate varies a lot depending on grow temperature, the growth time is not noted here for clarity. The scale bar is 10 μm.

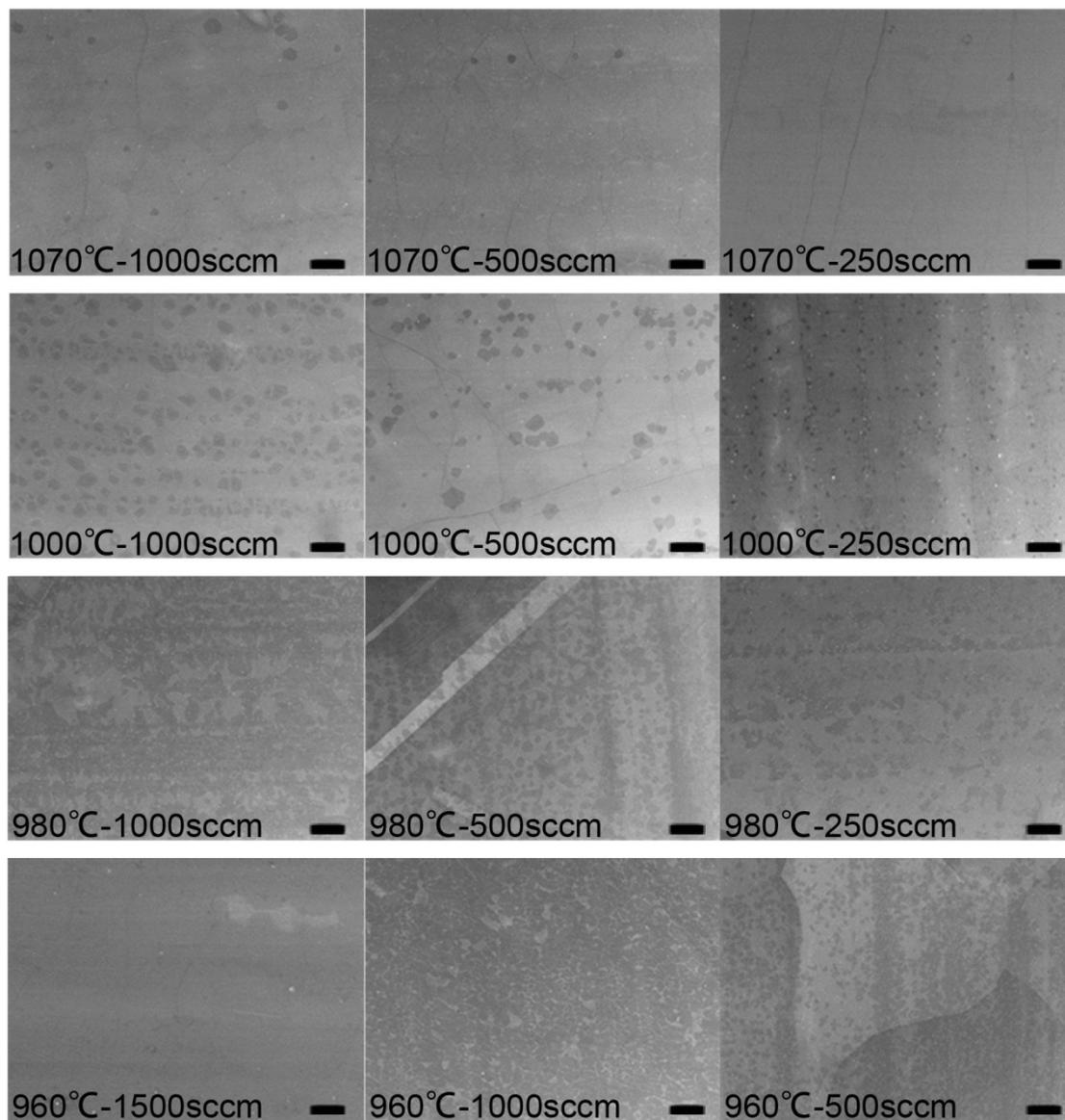Figure S2. Gas flow rate dependent graphene SEM morphology with growth temperature from 960 to 1070 °C. The growth temperature and gas flow rate are labelled in each image. The scale bar is 10 μm.
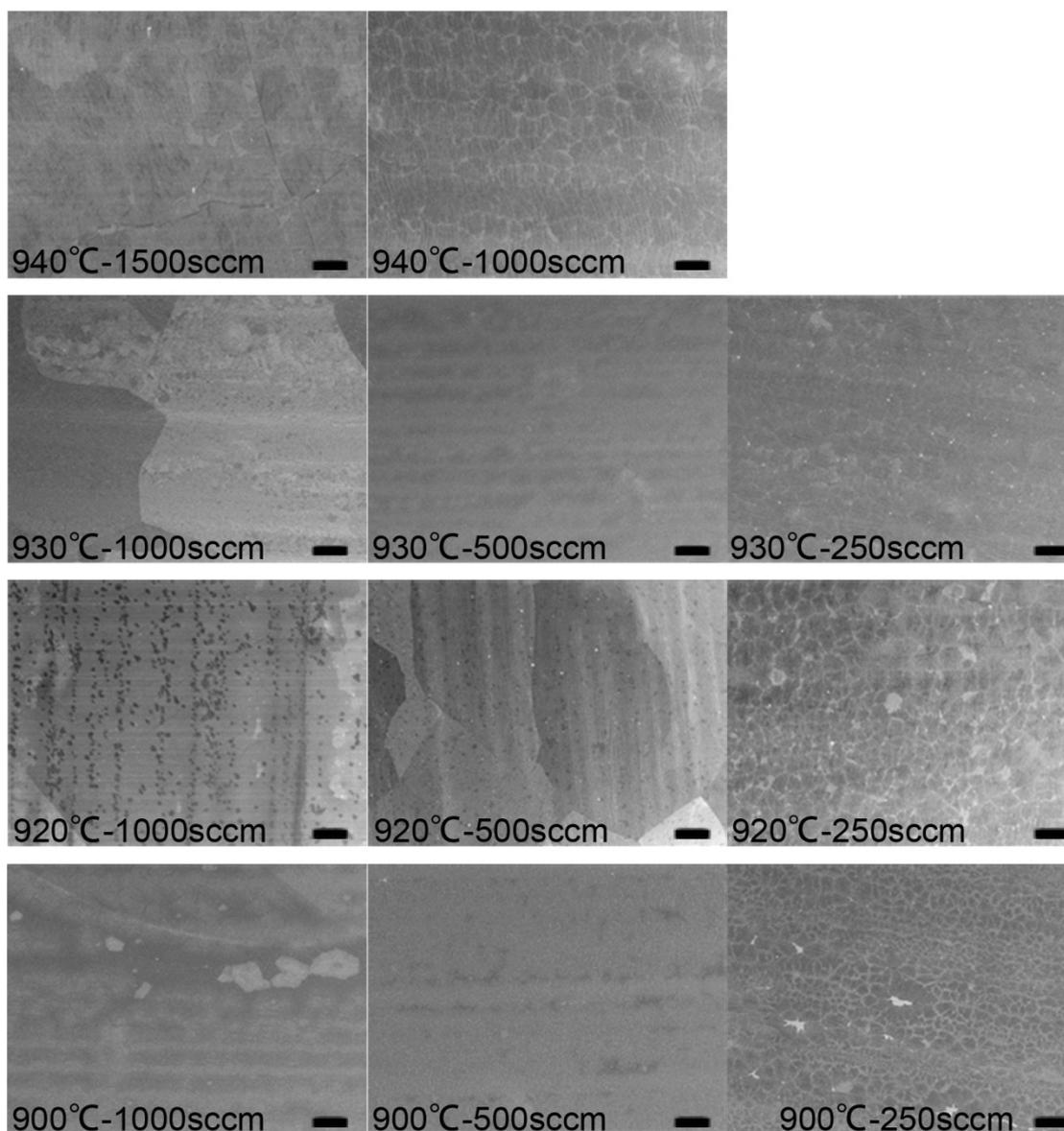
Figure S3. Gas flow rate dependent graphene morphology with growth temperature from 900 to 940 °C. The growth temperature and gas flow rate are labelled in each image. The scale bar is 10 μm.
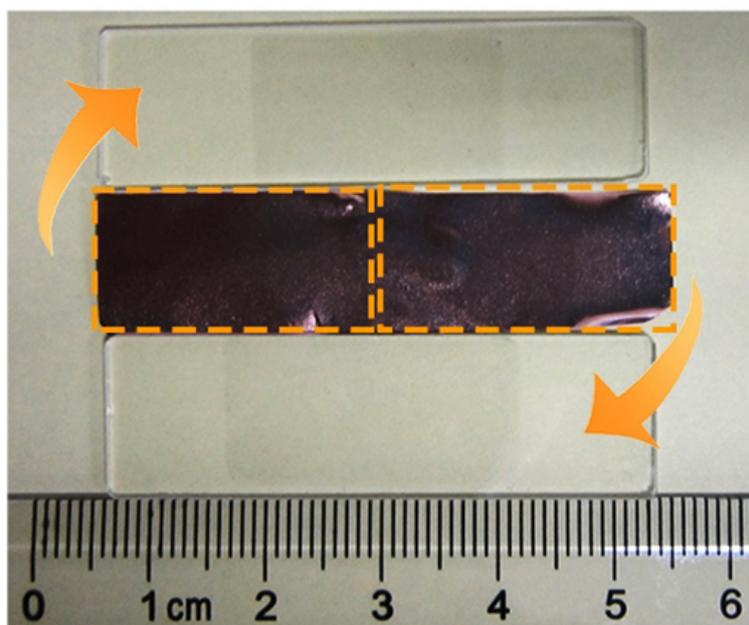
Figure S4. The Graphene/Cu foil of 1.5×6.0 cm^2 was cut into two pieces for the easier transfer process and UV-vis characterization. UV-Vis transmittance of graphene sample was obtained arbitrarily on the graphene sample. About fifteen points were chosen in total for characterization (Figure 1d). SEM images (Figure S5) of graphene samples were obtained every 1 cm from the up-stream Cu edge.
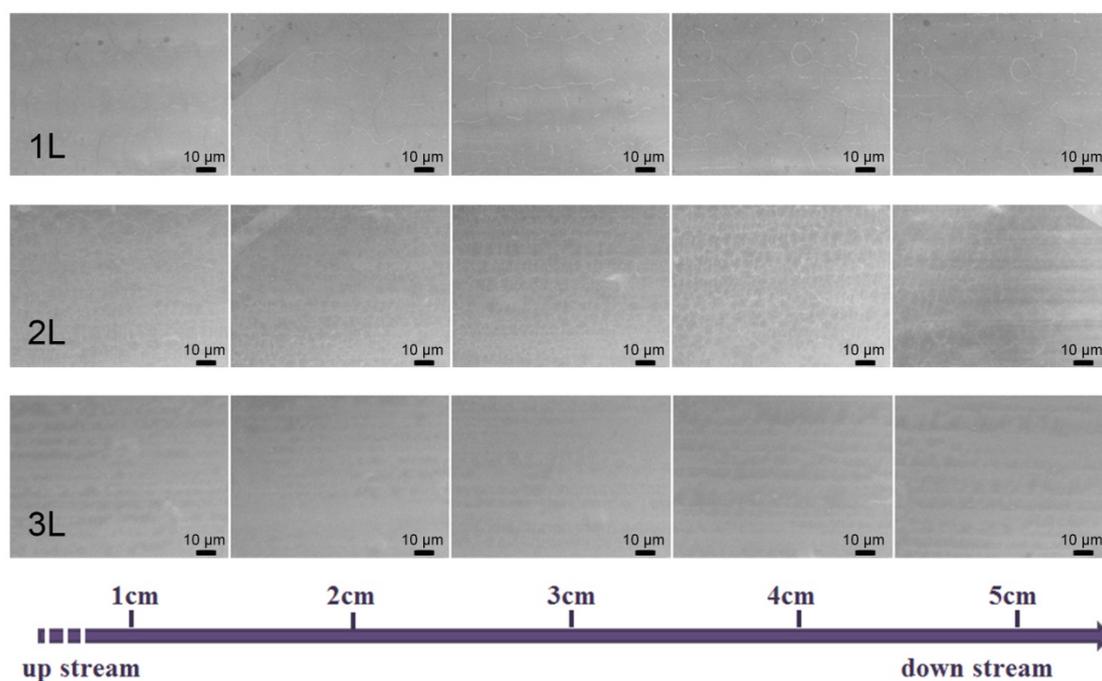

Figure S5. Position dependent graphene morphology on Cu strip. A 1.5×6.0 cm^2 strip of Cu foil was used to study the relationship between Cu position and graphene morphology. SEM images of graphene samples were obtained every 1 cm from the up-stream Cu edge. There is no obvious dependence between graphene layers and the distance from the up-stream of gas flow. Each zone shows uniform graphene films. The scale bar is 10 μm.
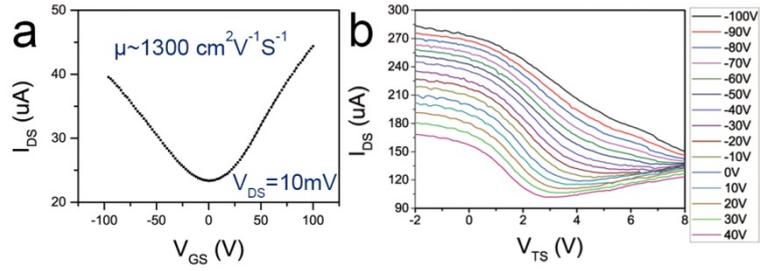
Figure S6. Electrical measurement of as-prepared bi-layer graphene. (a) The I_{DS}-V_{GS} curve of the as-prepared bi-layer graphene backed-gated FET device. (b) The dual-gate FET Graphene electrical conductance as a function of top gate voltage V_{TS} at different bottom gate voltages from -100V to 40V (shown in the right insert).

[1] I. N. Kholmanov, C. W. Magnuson, A. E. Aliev, H. F. Li, B. Zhang, J. W. Suk, L. L. Zhang, E. Peng, S. H. Mousavi, A. B. Khanikaev, R. Piner, G. Shvets, R. S. Ruoff, *Nano Letters* **2012**, *12*, 5679.

[2] K. Yan, H. L. Peng, Y. Zhou, H. Li, Z. F. Liu, *Nano Letters* **2011**, *11*, 1106.