

Supplementary Information for

***In-situ* observations of gas phase dynamics during graphene growth using  
solid-state carbon sources**

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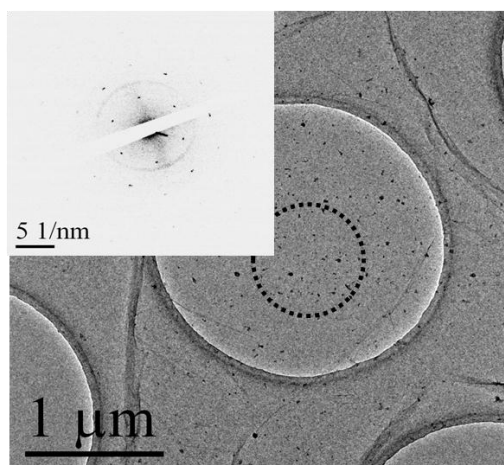
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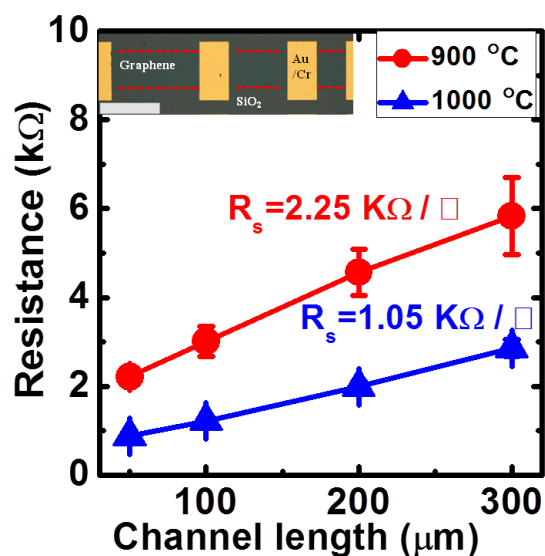
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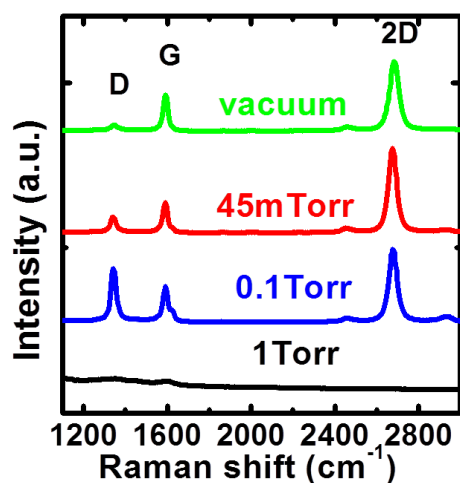
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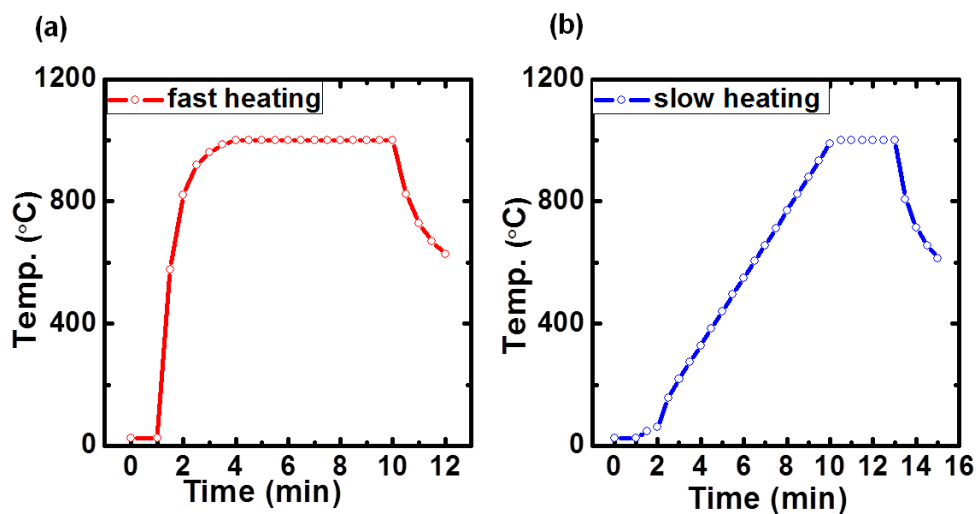
**Figure S1.** Representative low-magnification plan-view TEM image of graphene film grown at 1000 °C for 3 min then transferred to TEM support hole. Inset is selected area diffraction pattern obtained from a dotted circle region.



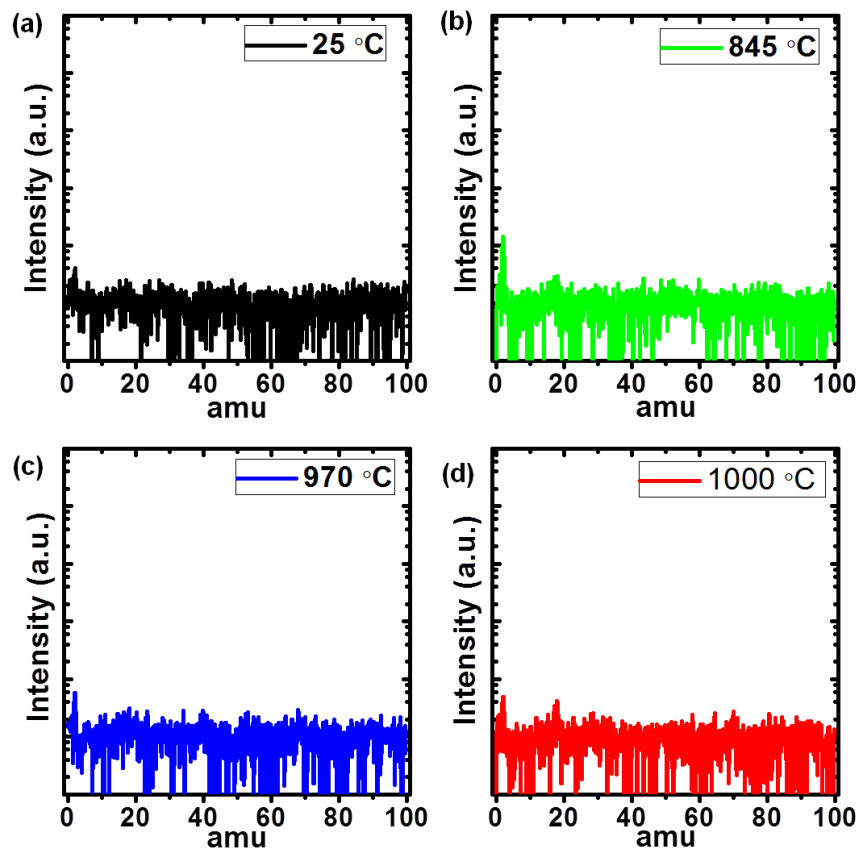
**Figure S2.** Sheet resistance measurement using TLM method. The plot shows total resistance of the TLM structure as a function of distance. The distance between the contacts on the TLM structure are 60, 100, 150, and 300 μm, respectively. The inset shows an optical microscopy image of patterned graphene layer on SiO<sub>2</sub> with Au (60nm)/ Cr (10nm). The scale bar in the inset is 200 μm.



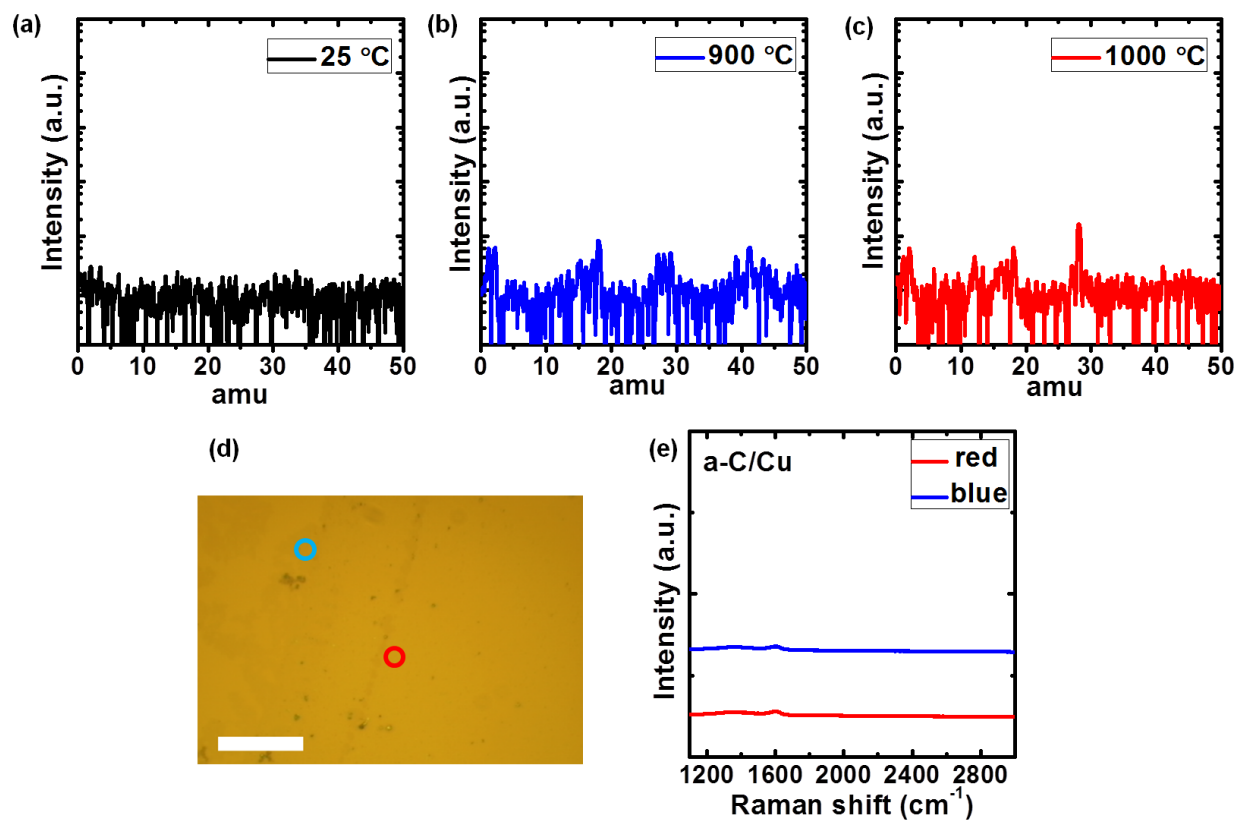
**Figure S3.** Typical Raman spectra of resulting layers on Cu foils after heating the 10 wt. % PMMA/Cu foil at 1000 °C for 3 min under a different total pressure in a process chamber. In case of the vacuum, the total pressure in the process chamber was  $\sim 1 \times 10^{-3}$  Torr. With increasing the total pressure, intensity of the D band increases, suggesting that the amount of efficient carbon sources may not be enough. If the total pressure was larger than 1 Torr, the Raman spectrum (black line) do not exhibit any carbon-related characteristics over a range of 1000-3000  $\text{cm}^{-1}$ , indicating that no graphitic layers were produced.



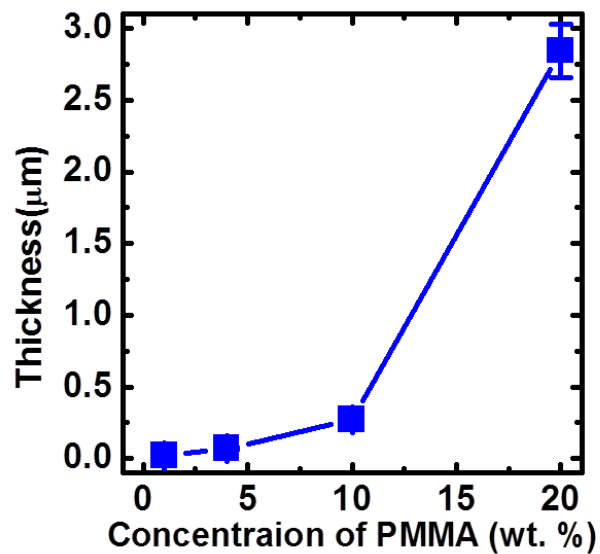
**Figure S4.** Temperature profiles as a function of time during heating. (a) At the fast heating temperature profile, heating started at 1 min and the temperature was reached around 1000 °C at 4 min. (b) At the slow heating temperature profile, heating started at 1 min and the temperature was reached around 1000 °C at 11 min.



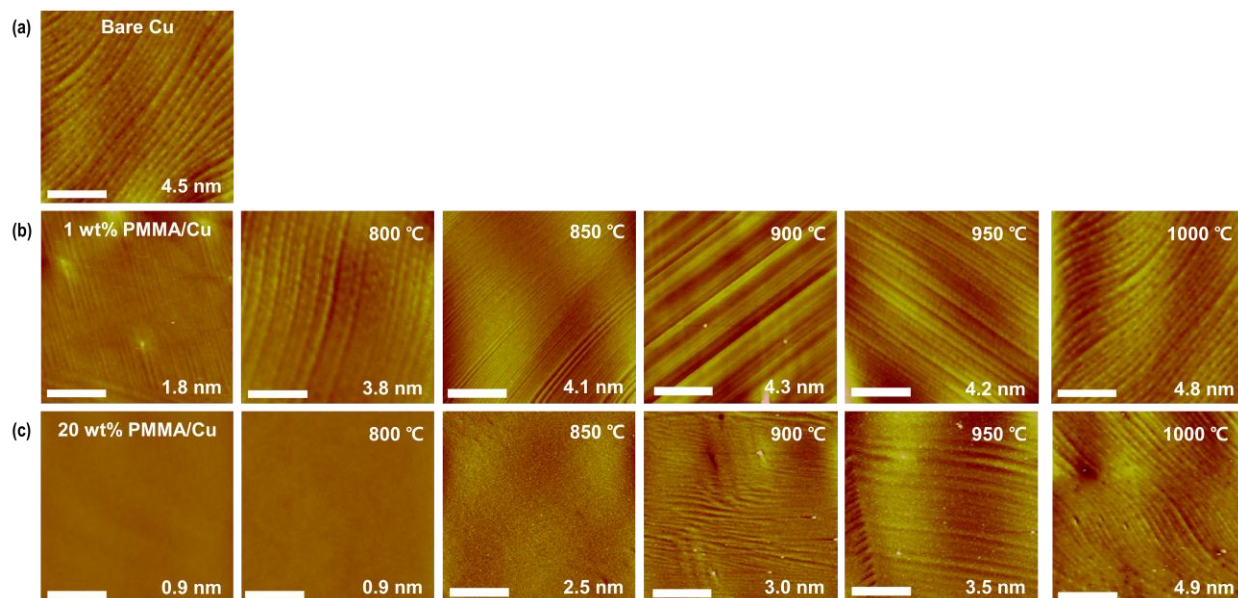
**Figure S5.** *In-situ* quadrupole mass spectra while ramping temperature from 25 °C to 1000 °C in vacuum ( $\sim 10^{-9}$  Torr), with a bare Cu foil. The horizontal axis represents atomic mass unit and the vertical axis is the logarithm of ion current.



**Figure S6.** (a-c) *In-situ* quadrupole mass spectrum while ramping temperature from 25 °C to 1000 °C in vacuum ( $\sim 10^{-9}$  Torr), with a 20 nm-thick amorphous carbon (a-C) film deposited on a piece of Cu foil. X-axis represents atomic mass unit and Y-axis is the logarithm of ion current. (d-e) Representative optical microscopy image and Raman spectrum of a 20 nm thick amorphous carbon-coated Cu foil after annealing at 1000 °C for 3 min under vacuum by using RTA then transferred into the SiO<sub>2</sub> substrate.



**Figure S7.** With a different concentration of PMMA in toluene, the thickness of the PMMA film deposited on SiO<sub>2</sub>/Si by spin coating at 5000 rpm for 1 min. The thickness of PMMA films were determined by a surface profiler.



**Figure S8.** (a) An AFM image of a bare Cu foil. AFM images of (b) 1 wt.% and (c) 20 wt.% of PMMA-coated Cu foils and those after heating between 800 and 1000 °C (from left to right). All scale bars are 3 μm. Root-mean-square (rms) roughness from each scan is labeled.