Supporting Information

Application of Tungsten as a Carbon Sink for Synthesis of Large-domain Uniform Monolayer Graphene Free of Bilayers/Multilayers

Wenjing Fang[†], Allen Hsu[†], Yong Cheol Shin[§], Albert Liao[†], Shengxi Huang[†], Yi Song[†], Xi Ling[†], Mildred S. Dresselhaus[†,‖], Tomas Palacios[†] and Jing Kong[†,*]

[†]Department of Electrical Engineering and Computer Sciences, [§]Department of Material Science and Engineering and [‖]Department of Physics, Massachusetts Institute of Technology, Cambridge, Massachusetts 02139, United States

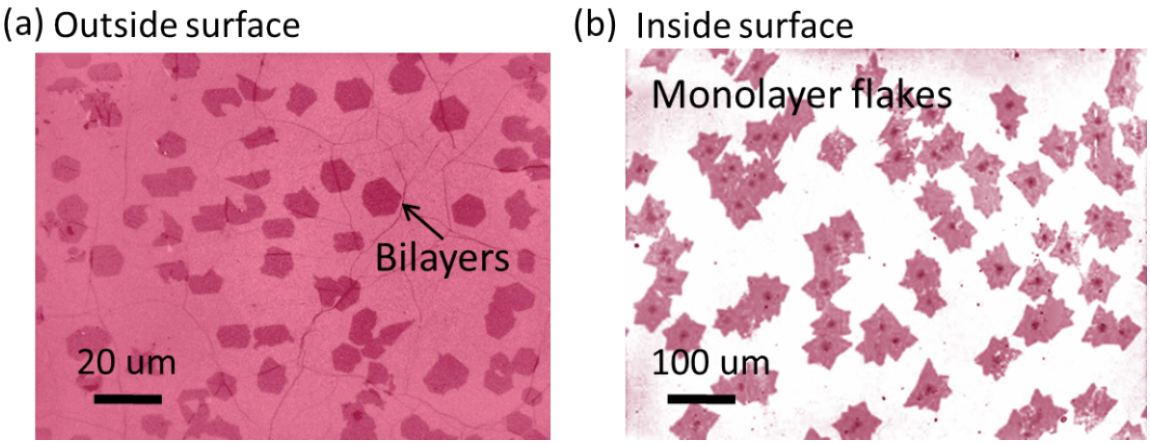

Figure S1. Optical images of transferred graphene grown on the outside surface of the Cu enclosure with W foil inside for 5 hours. (a) On the outside surface, bilayers start to appear after 5 hours under the same growth condition as that in Figure 2. (b) On the inside surface, graphene flakes start to grow.

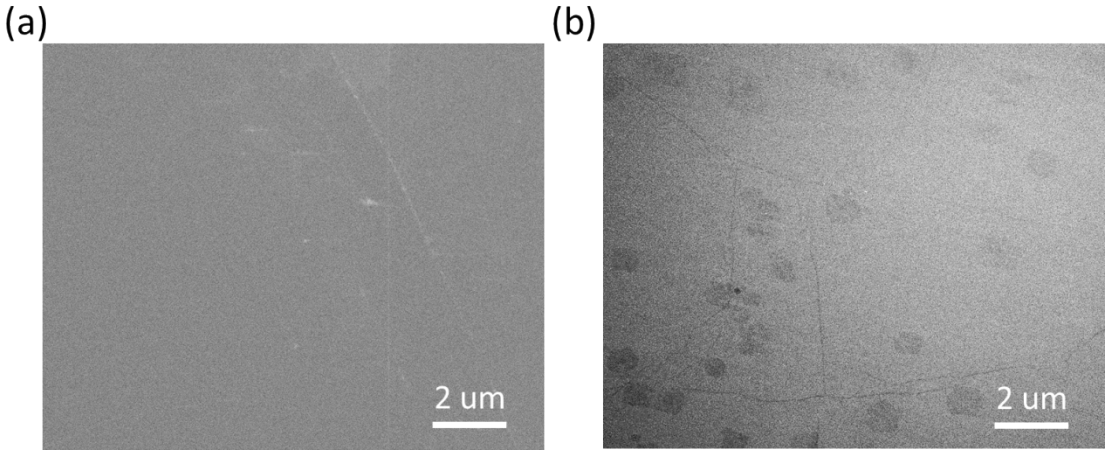

Figure S2. High resolution SEM image. (a) The monolayer graphene is uniform with no bi-/multi-layers on the outside of the Cu enclosure with W enclosed. (b) Small bilayer flakes on the flat Cu foil.

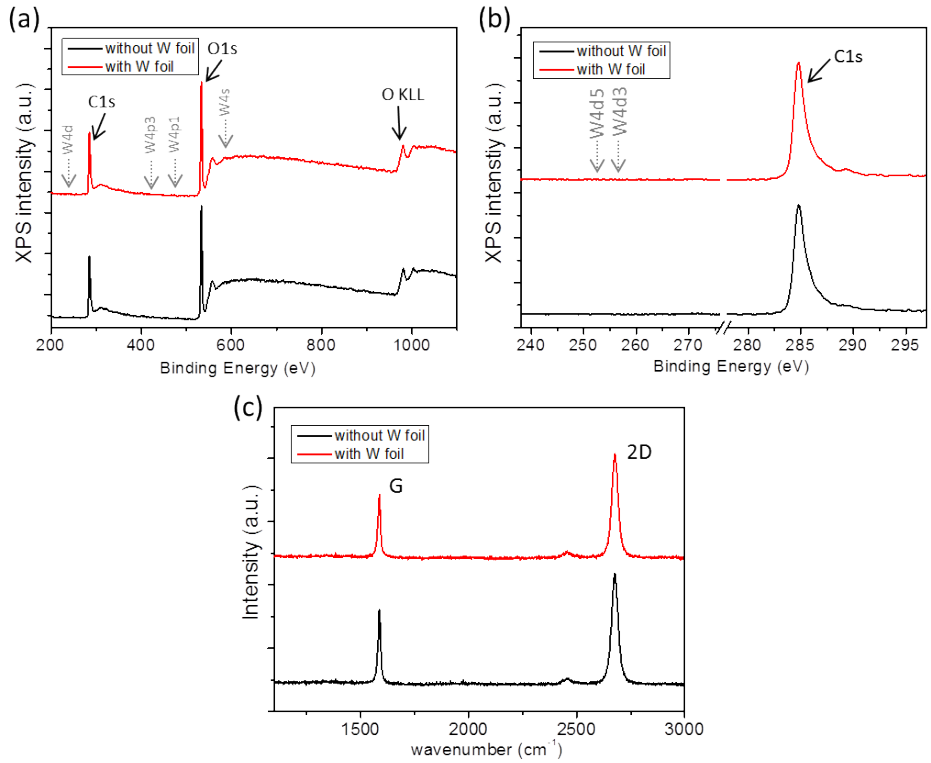

Figure S3. Comparison of graphene grown on Cu enclosure with and without W foil. (a) XPS survey on transferred graphene on SiO_2 substrate. (b) High resolution XPS for $W4d_5$/$W4d_3$ and C1s peaks. (c) Raman spectral from the same transferred samples.

We prepared graphene films using an empty Cu enclosure and a Cu enclosure with a W foil enclosed, respectively. After transferring graphene film onto SiO_2 substrate, we performed XPS and Raman spectroscopy on both samples. XPS survey results (black) shows that there is no W signal on the graphene grown on the empty Cu enclosure, indicating no W contamination in the growth system. For graphene grown on the Cu enclosure with the W foil, the XPS results (red) are the same as the reference sample, which indicates that there is no W impurity even with the W foil enclosed throughout the growth process. The reason that there is no W on the graphene is probably because the melting temperature of W is very high (~3422°C). Meanwhile the growth temperature (1045°C) is close to the melting temperature of Cu (~1,085°C) that Cu can easily get evaporated and coated onto W but not vice versa. Furthermore, the Raman spectral from both samples confirmed that the graphene grown on the Cu enclosure with the W enclosed is the same as the reference samples. There is no discernible D band in both Raman spectral, suggesting a high quality graphene film. Both G bands locate at 1587 cm^{-1} while 2D bands locate at 2674 cm^{-1} (black) and 2675 cm^{-1}(red), indicating a similar doping level. Thus, we conclude that there are no W impurities that caused any defects or extra doping in the graphene films.

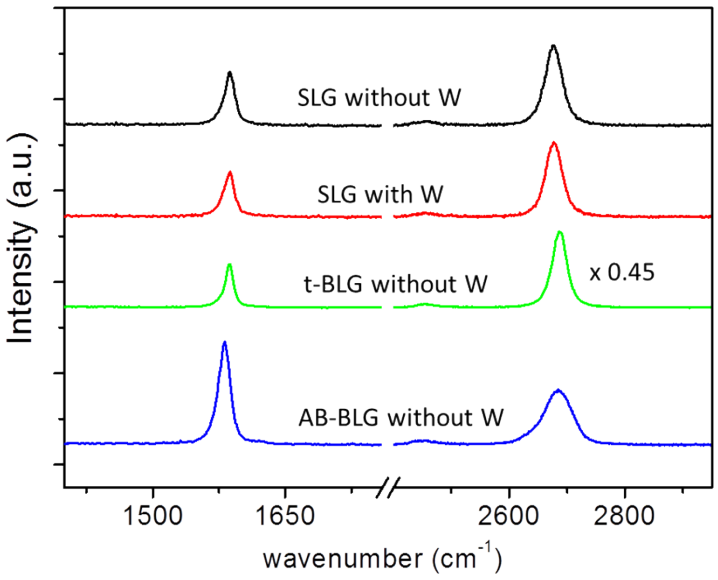

Figure S4. Raman spectra taken from CVD-grown graphene films. Monolayer graphene (SLG) grown on Cu enclosure without W (black) and with W (red). Turbostratic bilayers (t-BLG, green) and AB-stacked bilayers (AB-BLG, blue) grown from Cu enclosure without W.

We further performed Raman spectroscopy on monolayer and bilayer graphene from empty Cu enclosure and monolayer graphene from Cu enclosure with W enclosed. The black and red curves in Figure 4S represent monolayer graphene films with 2D band located at 2674 cm^{-1} and 2676 cm^{-1}. The full width at half maximum (FWHM) for both 2D bands are 33 cm^{-1}, indicating a monolayer.[1, 2] The intensity of the green curve is normalized according to the 2D band. The original intensity of Raman spectra is almost twice that of the monolayer graphene. Moreover, the FWHM is ~ 35 cm^{-1}, suggesting that the films are turbostratic bilayers. For the blue curve, the intensity of G band is almost twice that of monolayer graphene. Furthermore, the FWHM of the 2D band is ~55 cm^{-1}, suggesting AB-stacked bilayers.[1]

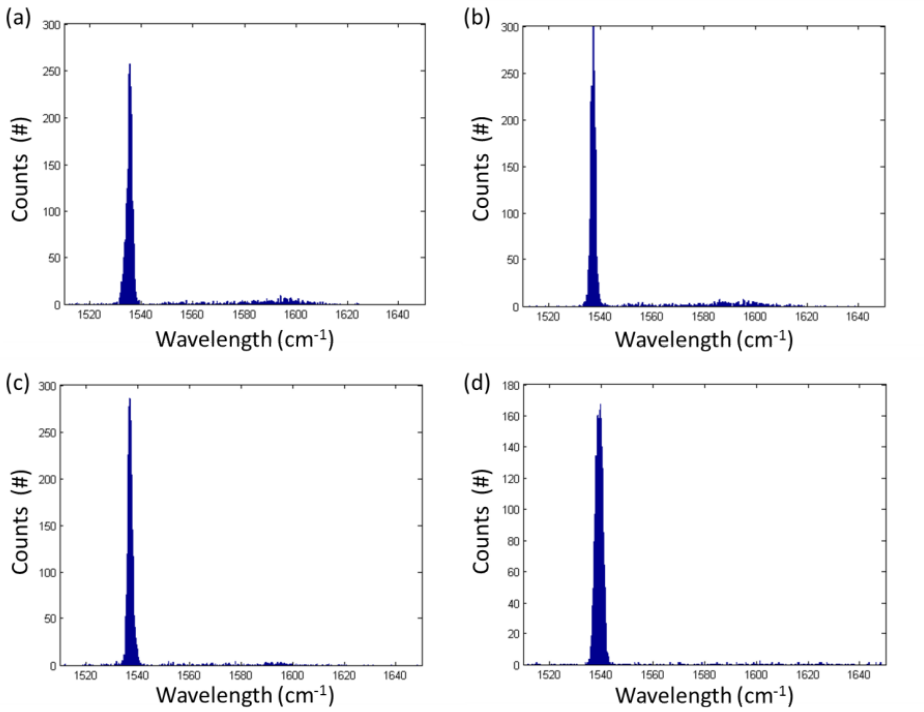

Figure S5. Statistics of G peak position for graphene grown on the outside of the Cu enclosures with W enclosed. Graphene was prepared by introducing $^{13}CH_4$ for the first 15 min, then switched to $^{12}CH_4$ for a total growth time of (a) 30 min, (b)1 hour, (c) 2 hour and (d) 5 hours.

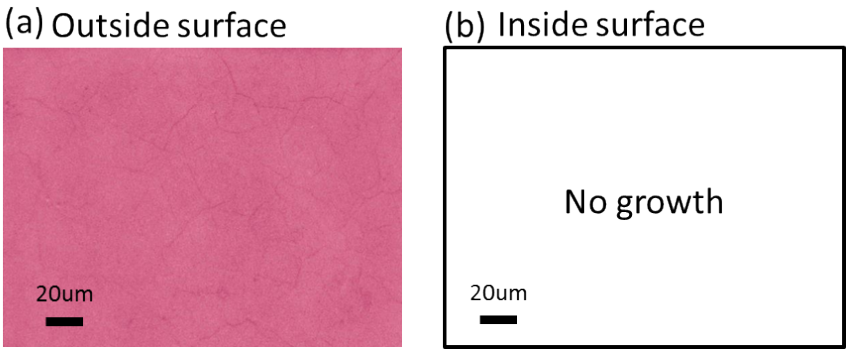

Figure S6. Optical images of transferred graphene after 2 hour growth using Cu-coated W foil. The W foil was placed inside of the Cu enclosure and annealed at 1045^0C under 10 sccm H_2 for 1 hour for Cu coating. Then the W foil was taken out and put inside another fresh Cu enclosure for graphene growth. (a) Uniform monolayer graphene grows on the outside of the Cu enclosure. (b) On the inside, there is no graphene growth.

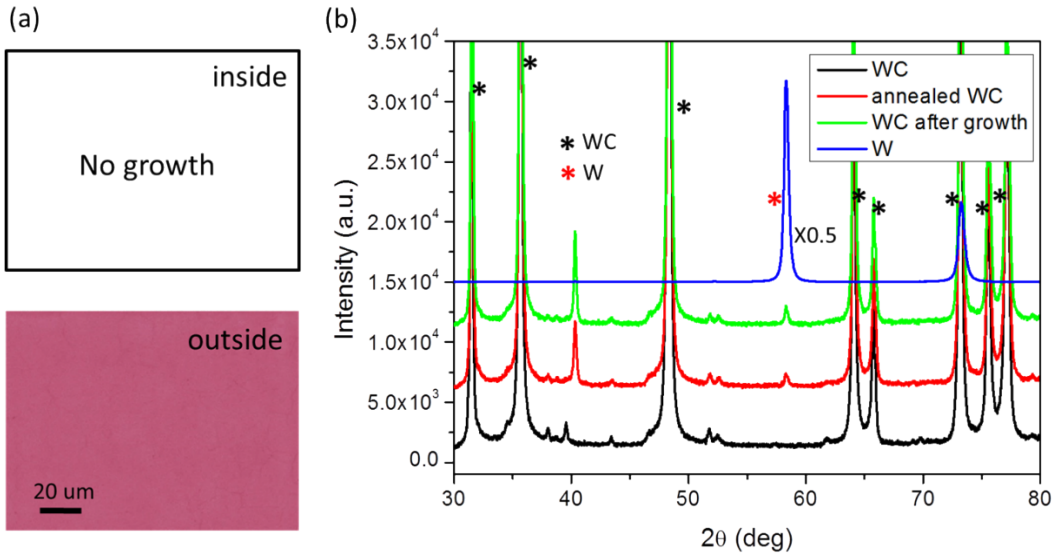

Figure S7, Graphene growth using Cu enclosures with WC for 2 hours. (a) Optical images of graphene grown on the inside and outside surfaces of the Cu enclosure. (b) XRD results on as received tungsten carbide (black), WC after annealing (red), growth (green) and as-received W.

Using WC inside of the Cu enclosure, the results are the same as when we used W foil. From the results of XRD analysis, we observe the peaks of W. It is possible that during the annealing steps, WC gets reduced under the H_2 environment, which acts as the same way as W foils.

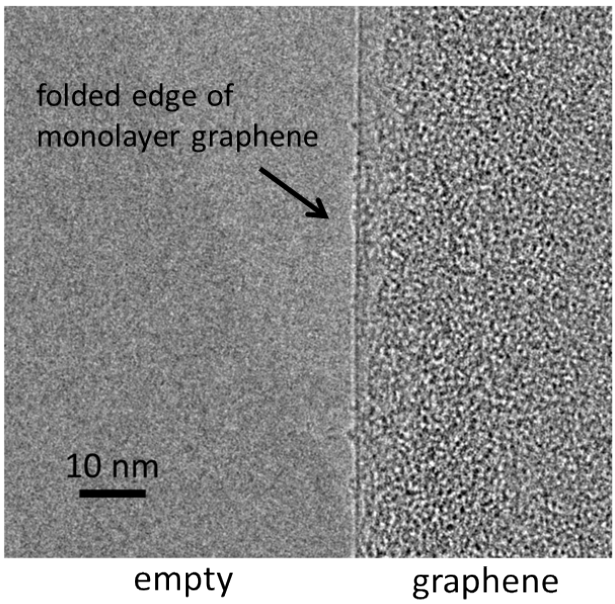

Figure S8. TEM characterization of monolayer graphene. TEM image shows the edge of graphene film consisting of one layer.

1. Ferrari, A. C.; Meyer, J. C.; Scardaci, V.; Casiraghi, C.; Lazzeri, M.; Mauri, F.; Piscanec, S.; Jiang, D.; Novoselov, K. S.; Roth, S.; Geim, A. K. *Phys Rev Lett* **2006,** 97, (18).
2. Calizo, I.; Balandin, A. A.; Bao, W.; Miao, F.; Lau, C. N. *Nano Lett* **2007,** 7, (9), 2645-2649.