

Supporting Information.

A Highly Practical Route for Large-Area, Single Layer Graphene from Liquid Carbon Sources such as Benzene and Methanol

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1. Graphene Synthesis

(a) Static exposure



(b) Dynamic exposure



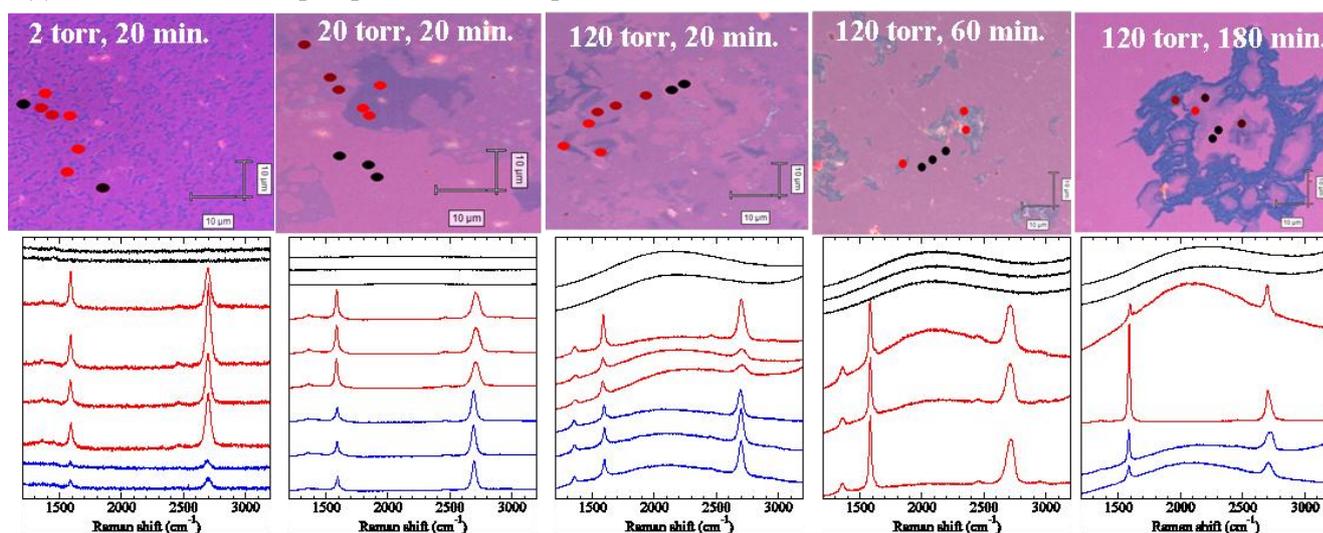
Figure S1. Actual pictures of the safe and simple experimental setup that is used in the synthesis of graphene films on copper foil.

2. Raman Spectra on Methanol-based graphene with pressure and exposure time

Graphene formation from methanol is very different from the other liquid hydrocarbons we studied because amorphous carbon formation was never observed, as demonstrated in Figure S2. At low pressure (2 torr for 20 minute exposure), we observe disconnected, mostly single layer graphene (left top panel in Figure S2a). As we increase the methanol pressures to 20 torr and to saturation at ≈ 120 torr, we do not observe amorphous carbon formation. In fact, the higher vapor pressure yields less carbon deposition on the copper foil. The transferred

samples show less graphene fragments on the SiO₂/Si substrate with increasing methanol vapor pressure as demonstrated from the optical images shown in Figure S2a. At very high pressure (≈ 120 torr) and long exposure times (3 hours), we only obtain a small amount of multilayer graphene.

(a) Static methanol vapor pressure and exposure time



(b) Methanol vapor flow (25 millitorr and 20 min.)

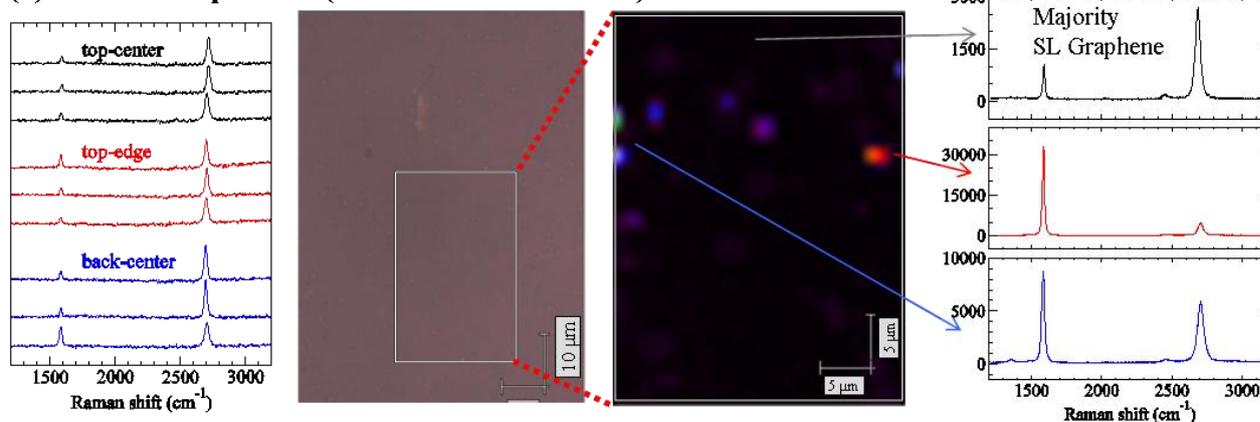


Figure S2. (a) Graphene formation from static exposure to methanol vapor as a function of pressure and time. Note that the amount of graphene formed decreases with increasing exposure time due to methanol inhibiting carbon from adhering to the Cu surface. For 180 minute exposures, we observe only a small amount of multilayer graphene flakes. The Raman spectra from the highlighted spots are shown below. (b) Graphene formation from dynamic methanol vapor exposure. Raman spectra from different parts of the copper foil indicate uniform coverage (left). The middle panel contains an optical image of a transferred sample with spatially-resolved Raman mapping based on G band-intensity, indicating mostly single layer graphene.

3. Optical Transmittance Measurements

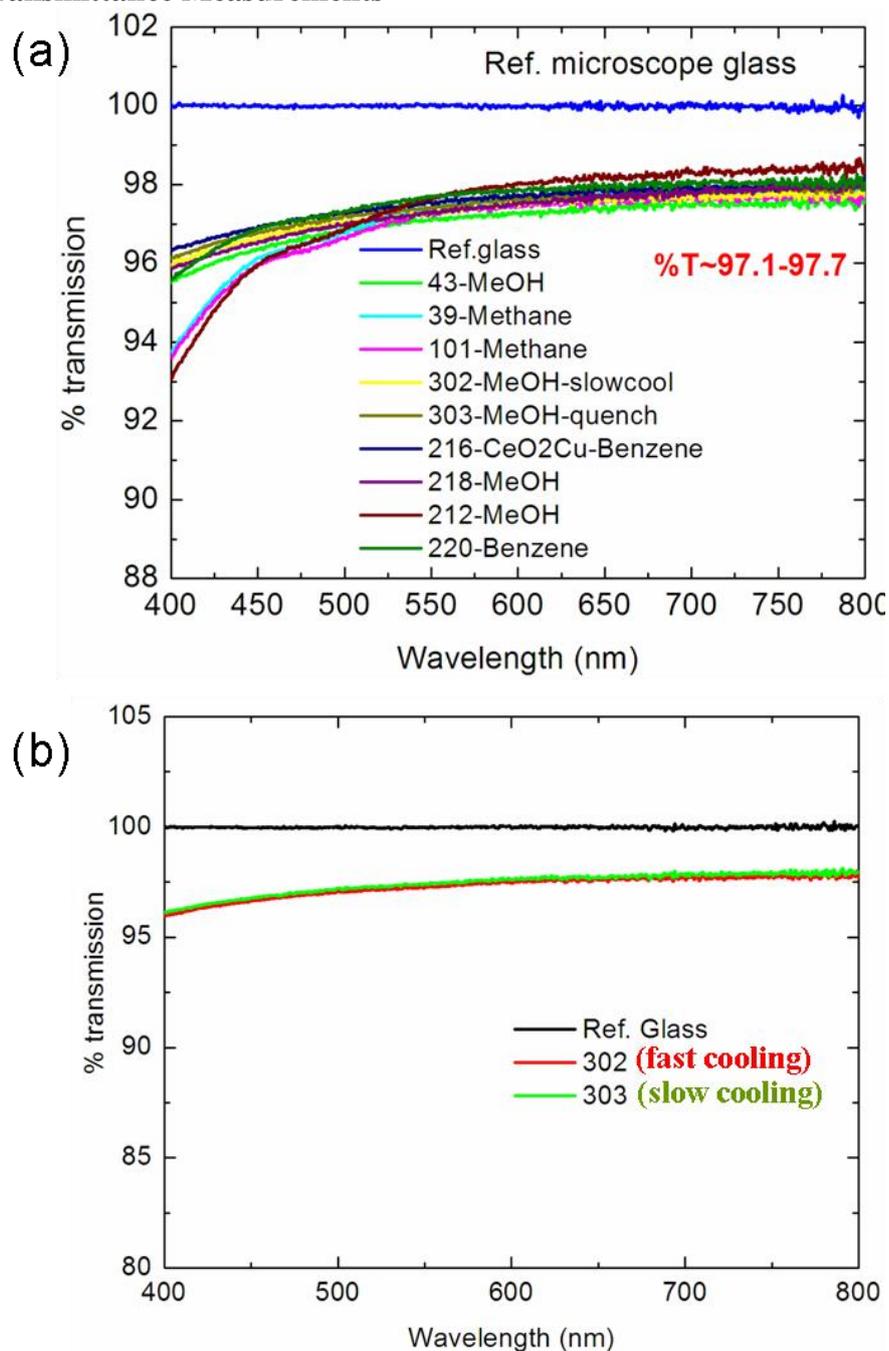
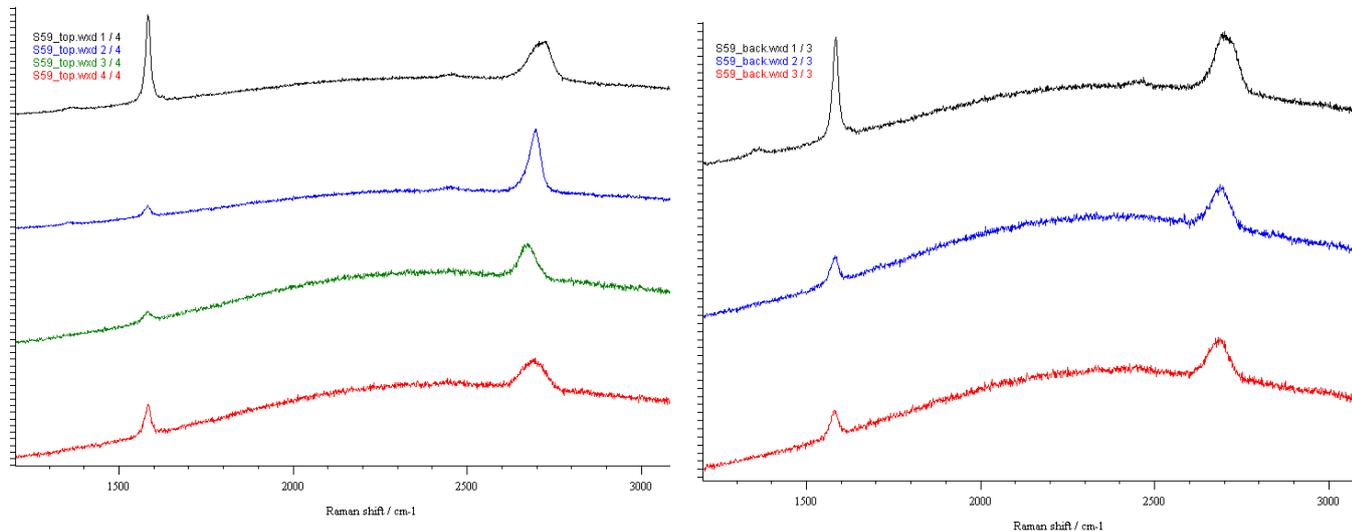


Figure S3. (a) Optical transmittance of large-area graphene films ($\sim 2 \text{ cm}^2$) from a variety of samples. All indicate a single layer graphene value. (b) Optical transmittance of large area graphene films ($\sim 2 \text{ cm}^2$) as a function of cooling. The two curves are almost identical, within the error bars, and therefore no effect of cooling rate on the number of graphene layers produced on copper surface was observed.

4. Raman Spectra on as-grown graphene/copper samples from different liquid carbon sources

4.1 ETHANOL (EtOH)

a) Sample #59: EtOH, 25 millitorr, 20 min, 1000° C



(b) Sample #107 MeOH/EtOH (1:1), 20 min., saturation pressure (~120 torr), 1000° C.

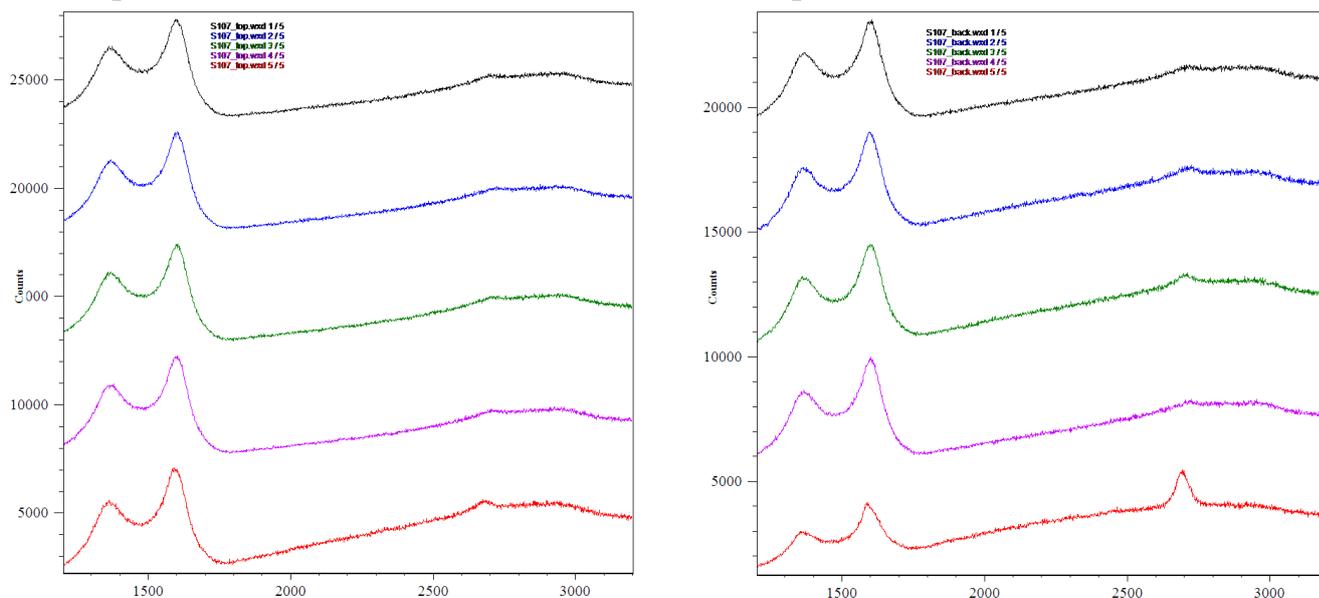


Figure S4. Raman spectra of as-grown copper/graphene samples using EtOH (a) and 1:1 mixture of MeOH/EtOH (b). The left and right panels show spectra from different locations on the top and bottom surfaces, respectively. We were unable to obtain from ethanol. The spectra shown in (a) are typical for graphene films with both single and multilayer graphene domains. The mixture of EtOH and MeOH at high pressure exposures has a spectrum consistent with amorphous carbon.

4.2 HEXANE

Sample #60: Hexane, 5 mtorr for 45 min.

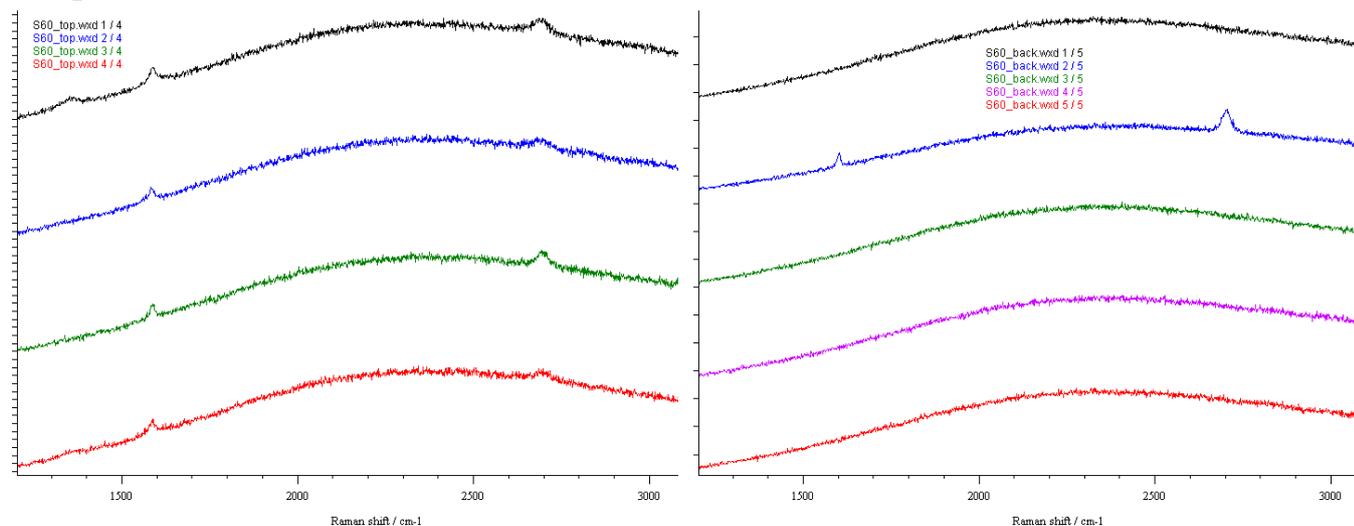
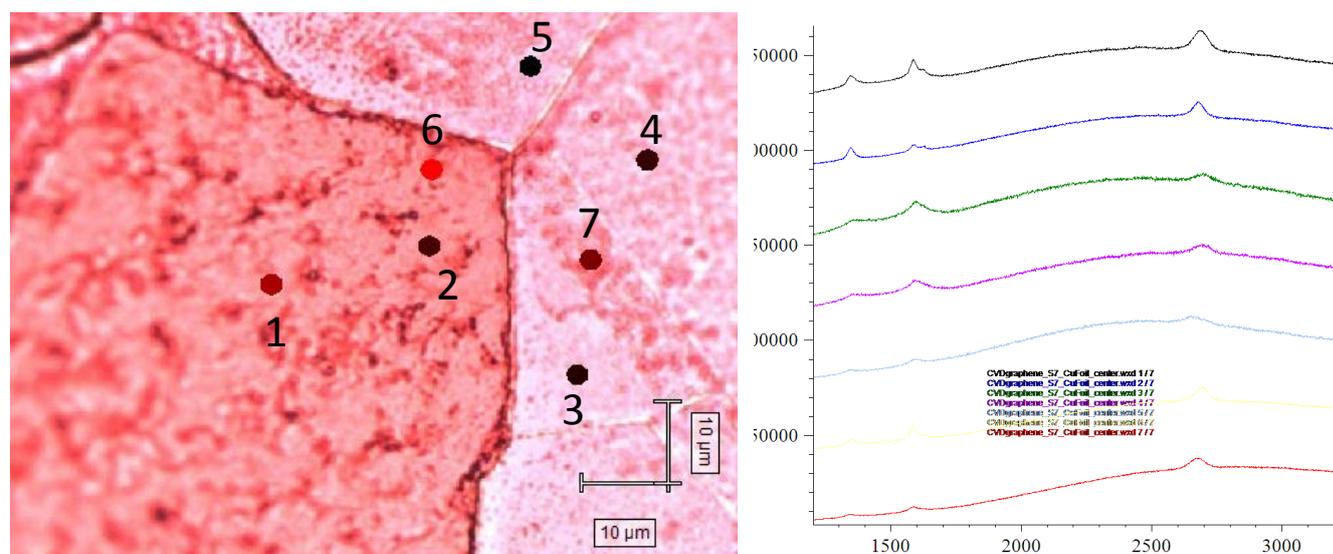


Figure S5. Raman spectra of as-grown copper/graphene samples using hexane. The left and right panels show spectra from different locations on the top and bottom surfaces, respectively. The Raman peaks suggest partial coverage of multilayer graphene.

4.3 TOULENE

(a) Toluene with high flow rate (1 ml/30 min), 100 mtorr, 10 min



(b) Toluene with low flow rate (0.1 ml/30 min, P=10 mtorr)

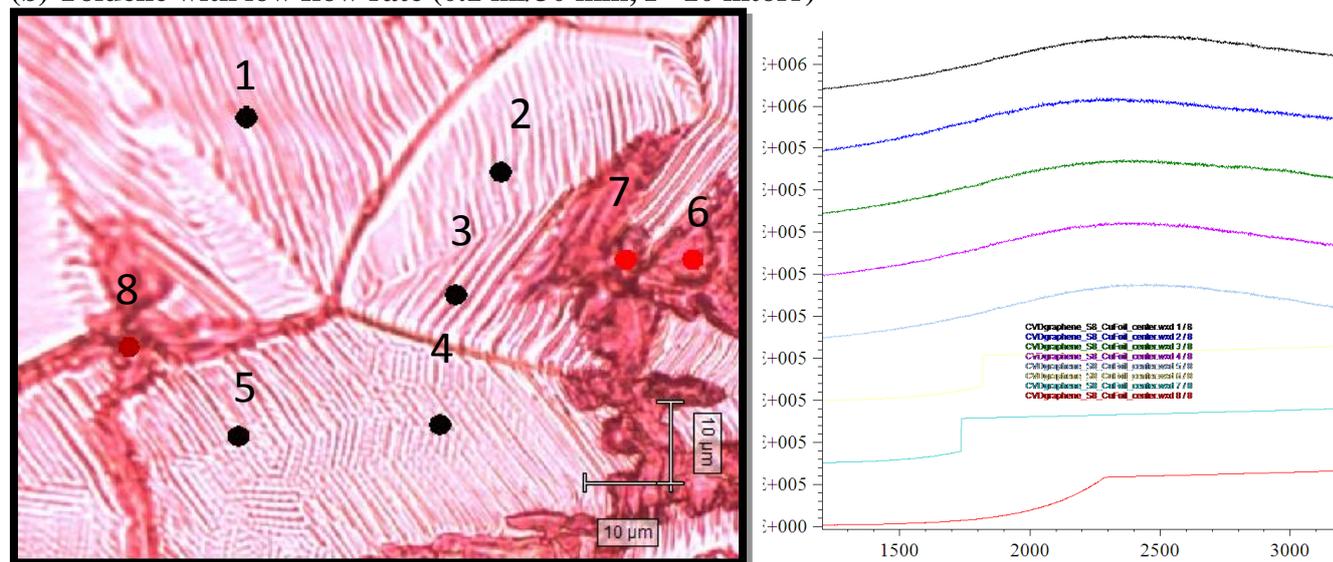
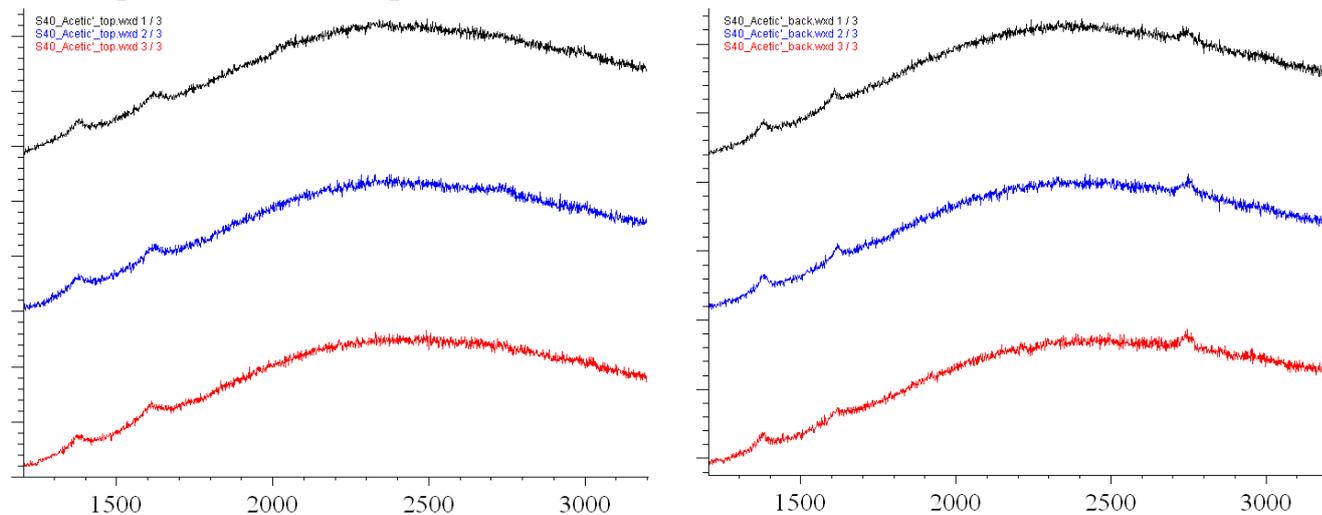


Figure S6. Optical images and Raman spectra of as-grown copper/graphene samples using toluene with high (a) and low flow rate (b). While high-flow rate yields multilayer graphene with large defect peaks, we did not observe graphene formation with a low-flow rate.

4.4 ACETONE

(a) Sample #40: Acetone exposure at 900° C for 20 min., P=32 mtorr.



(b) Sample #42: Acetone, 1000 °C, 15 min. exposure.

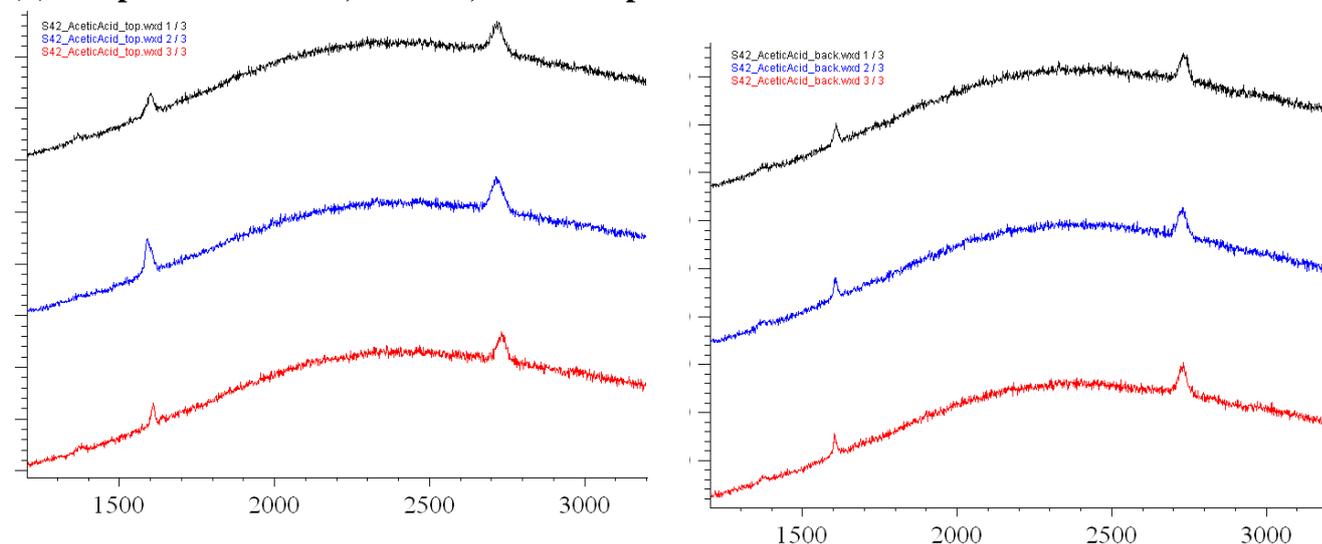


Figure S7. Raman spectra of as-grown copper/graphene samples using acetone at two different temperatures. We obtained multilayer graphene over the entire copper surface at 1000° C, while we only observed partial coverage at 900° C.

4.5 ACETIC ACID

(a) Sample #118: Acetic Acid, 30 mtorr, 1000°C, 20 min.

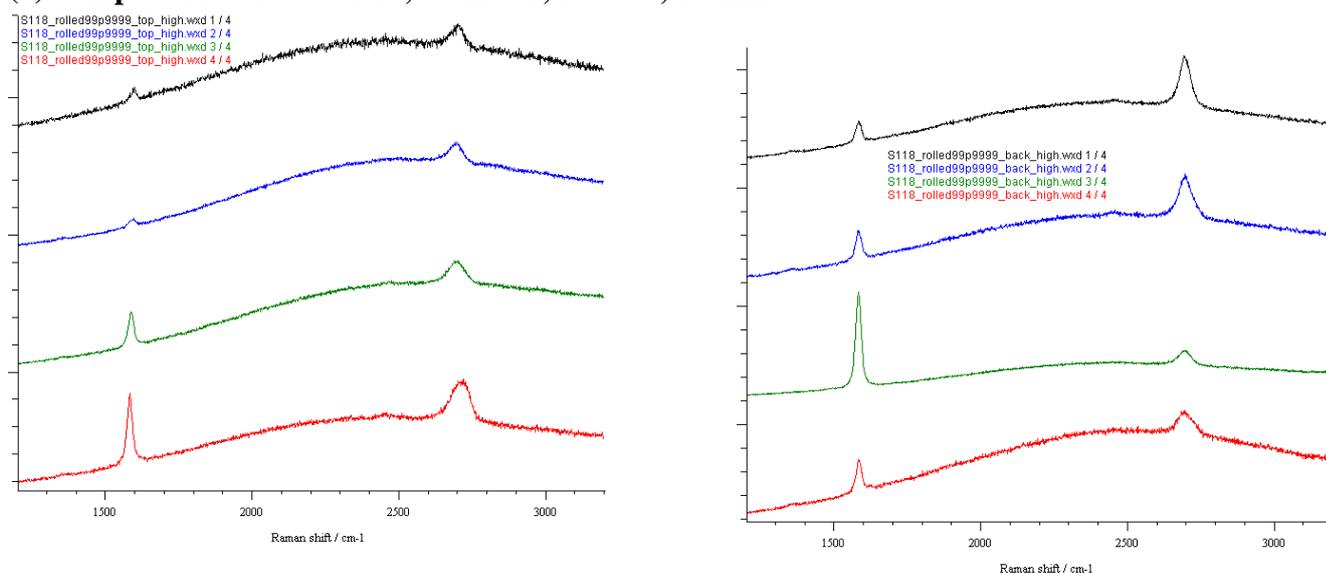


Figure S8. Raman spectra of as-grown copper/graphene samples using acetic acid. The Raman spectra indicate good coverage of multilayer graphene on both sides of the foil.