

Supporting Information

Title: Selective Growth of Graphene in Layer-by-Layer via Chemical Vapor Deposition

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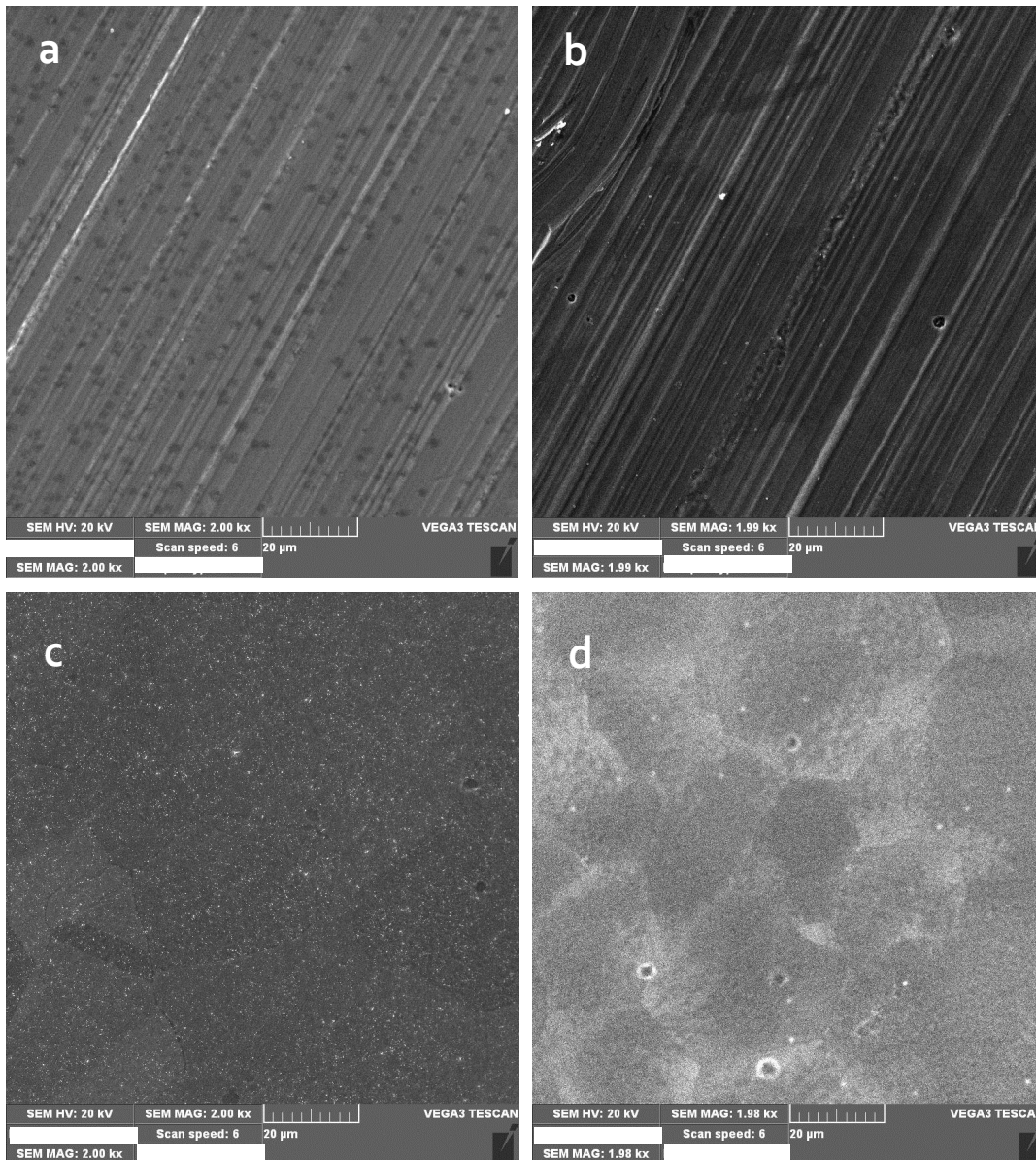


Figure S1. SEM images of the as-grown graphene films using unpolished Cu foil and polished one. (a) After first CVD using an unpolished Cu foil. (b) After second CVD using an unpolished Cu foil. (c) After first CVD using a polished Cu foil. (d) After second CVD using a polished Cu foil. The first CVD was performed for 10 min. A thin Cu (~40 nm) was deposited, and the second CVD was performed for 30 min.

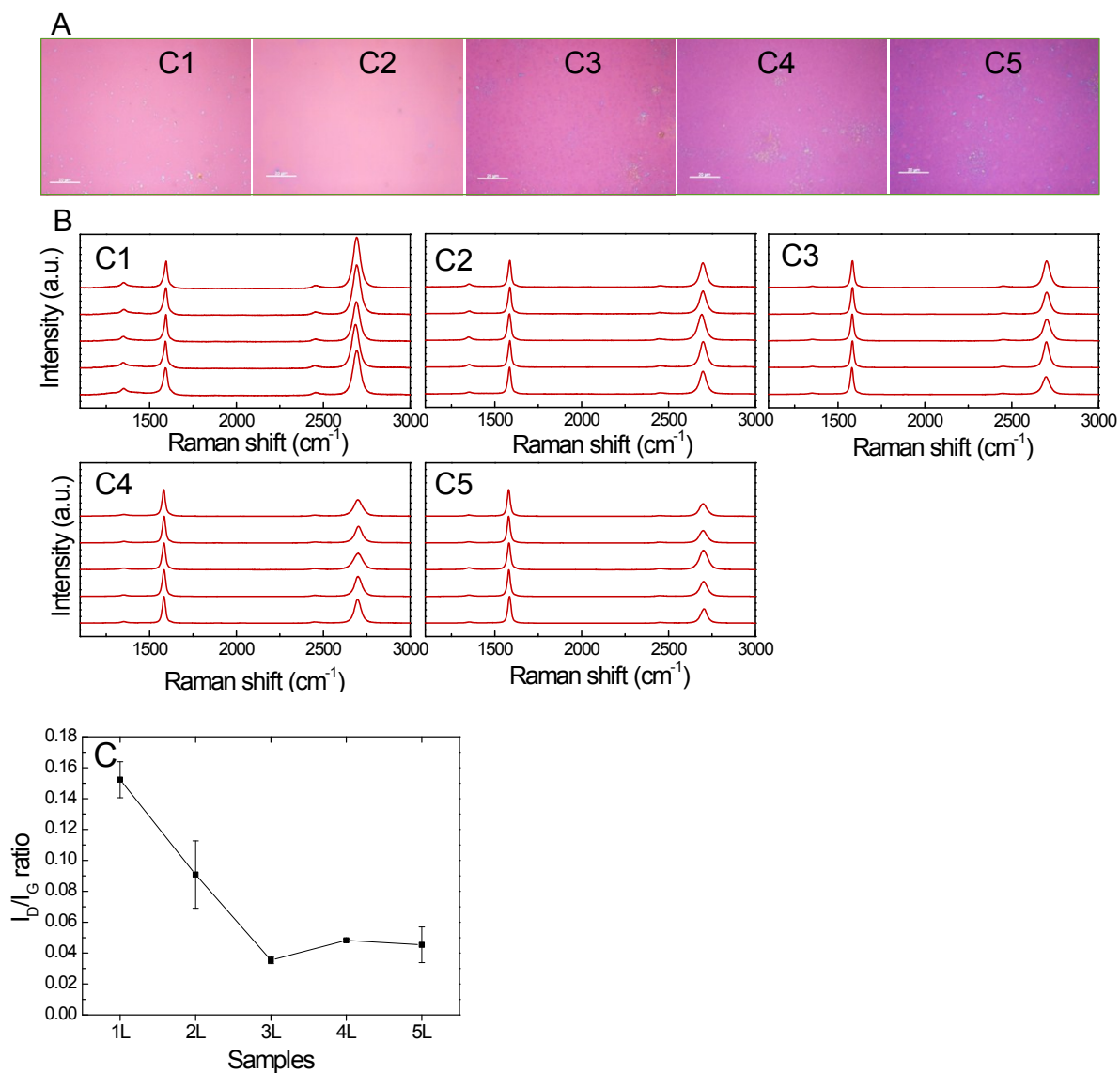


Figure S2. (A) Optical image of transferred graphene films (C1,C2,C3,C4,C5) onto a SiO₂/Si substrate (scale bar, 20 μm). (B) Raman spectra (Renishaw Co., wavelength 514 nm) taken from five random points of each graphene sample (C1-C5). (C) Extracted I_D/I_G ratios (average and standard deviation values) of transferred graphene films (C1,C2,C3,C4,C5).

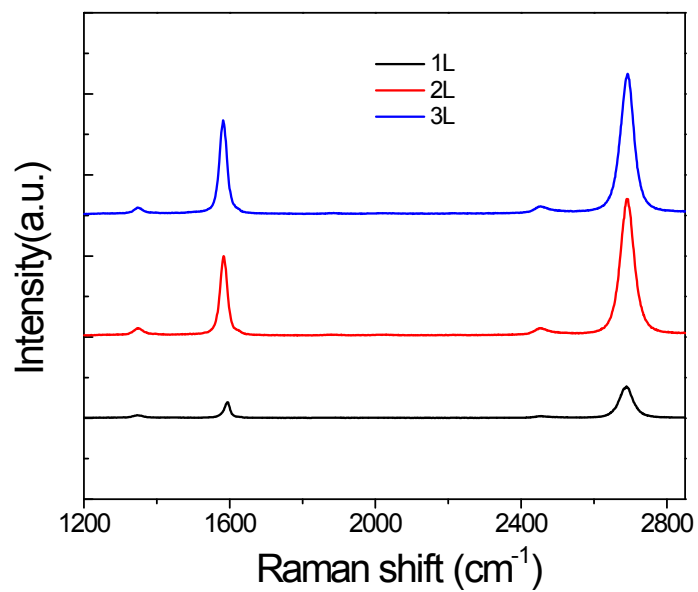


Figure S3. Raman spectra (wavelength 514 nm) of 1L graphene and non-Bernal-stacked 2L, 3L graphene. The 2L and 3L graphene films were made by layer-by-layer transfer onto a SiO₂/Si substrate using the PMMA method.

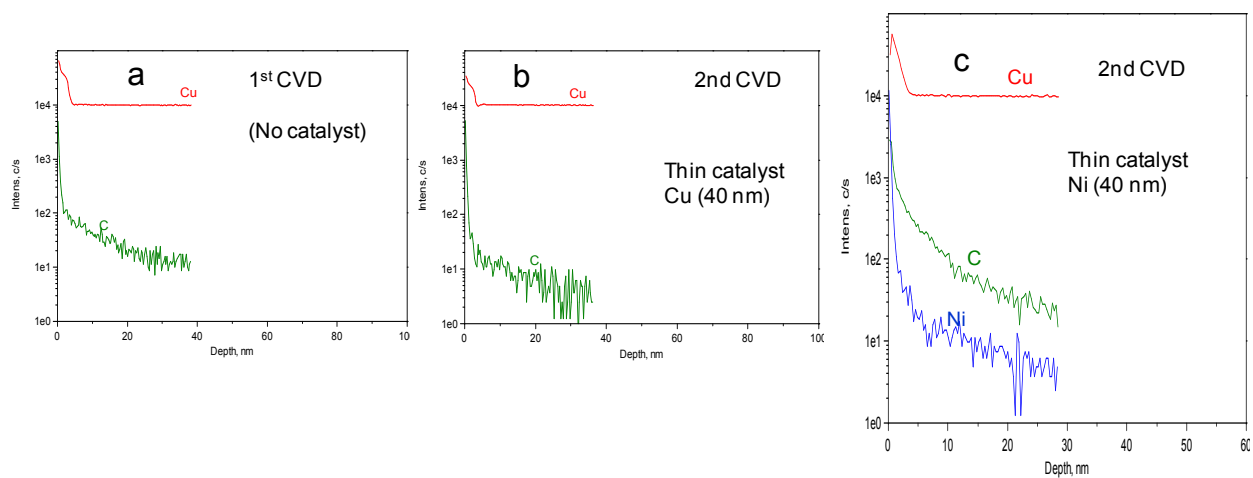


Figure S4. SIMS depth profile of Cu and carbon in Cu foil (a) after first CVD, (b) after second CVD (a thin 40 nm Cu catalyst was applied), and (c) SIMS depth profile of Cu and carbon and Ni after second CVD (a thin 40 nm Ni catalyst was applied).

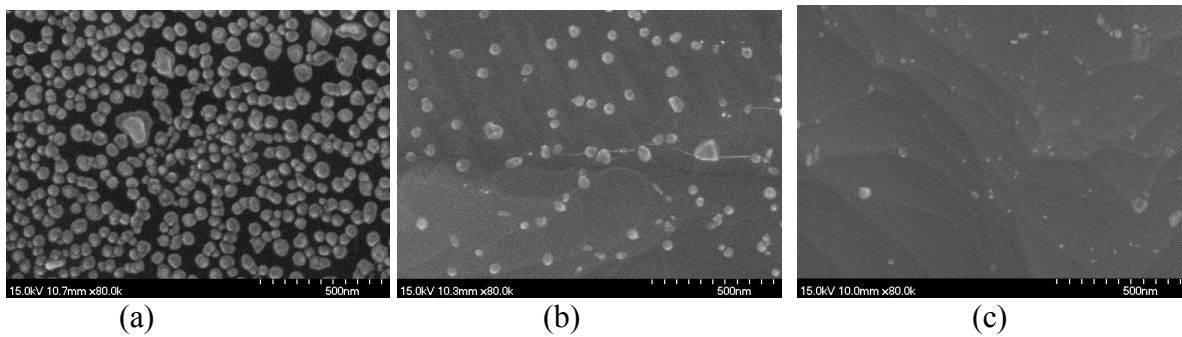


Figure S5. SEM images of thin Cu layer after second CVD. (a) after 30s, (b) 1 m, (c) 1m 30s.

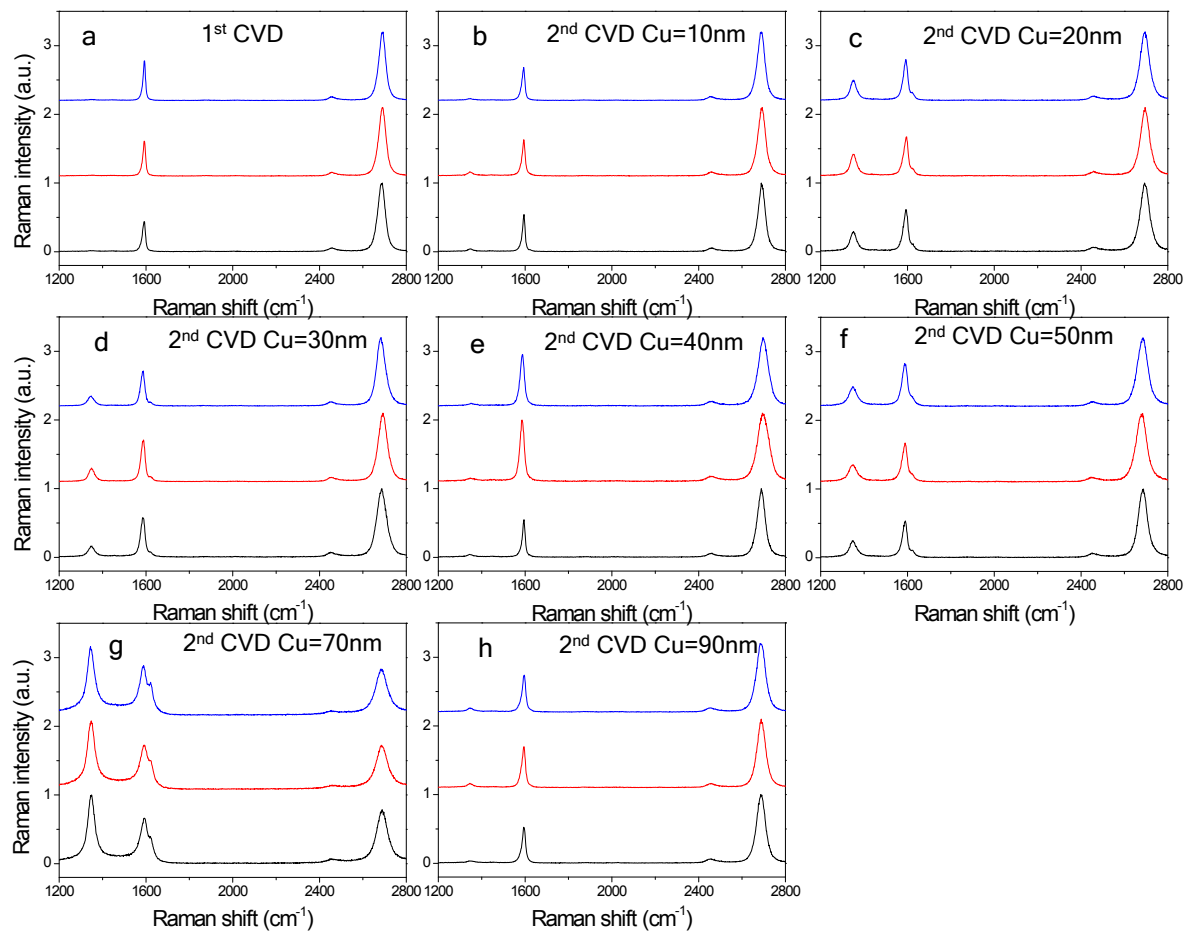


Figure S6. Raman spectra of graphene films grown using different top Cu film thickness. The methane and hydrogen flow was set to 10 and 200 sccm, respectively.

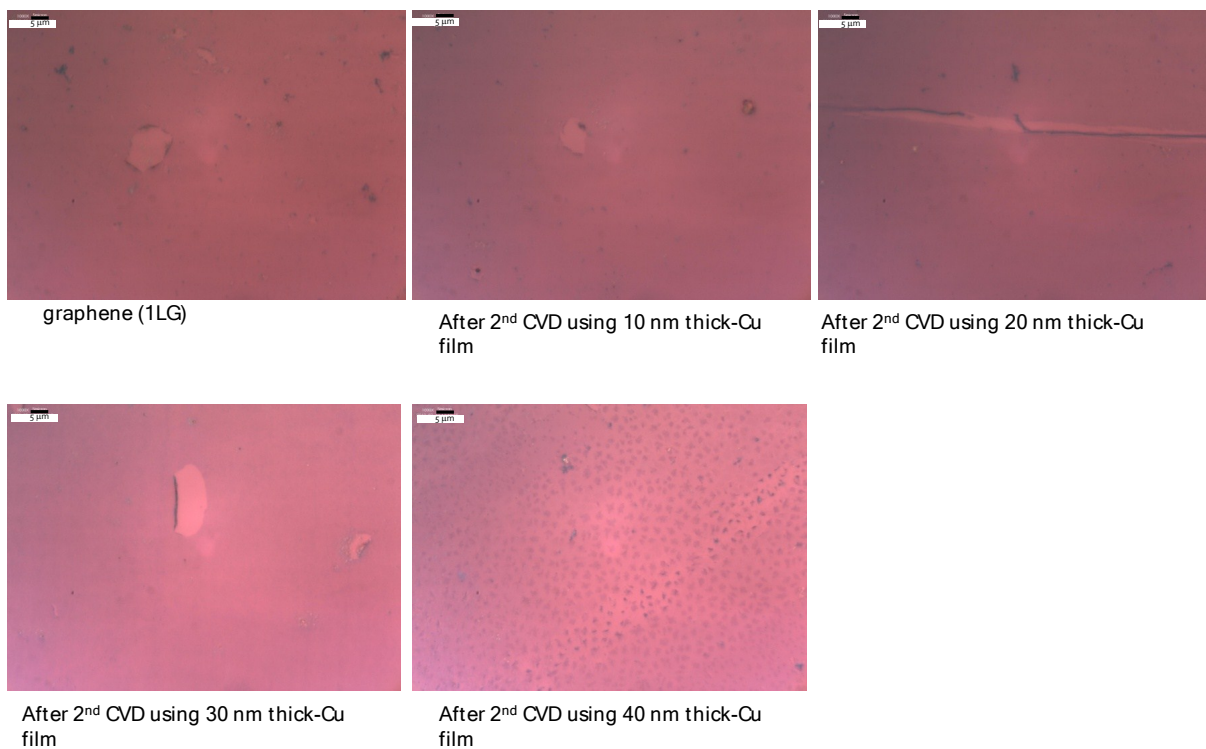


Figure S7. Optical images of the transferred graphene films grown using different Cu film thickness. For 40 nm-Cu case, 2L with isolated grains are spotted. The methane and hydrogen flow for the second CVD was set to 10 and 200 sccm, respectively.

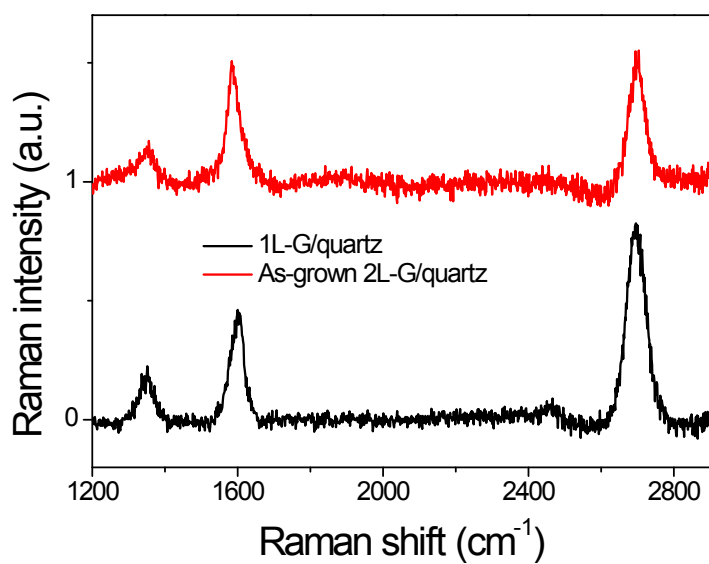


Figure S8. Raman spectra of graphene films on a quartz wafer. The 1L graphene was transferred on a quartz wafer (black line). And second CVD was performed (red one) after ~ 40 nm Cu film was deposited. The I_{2D}/I_G ratio decreased to ~ 1 after the second CVD, supporting that the layer thickness was increased.