

Supporting information for

Hydrogen-assisted fast growth of large graphene grains by
recrystallization of nano-grains

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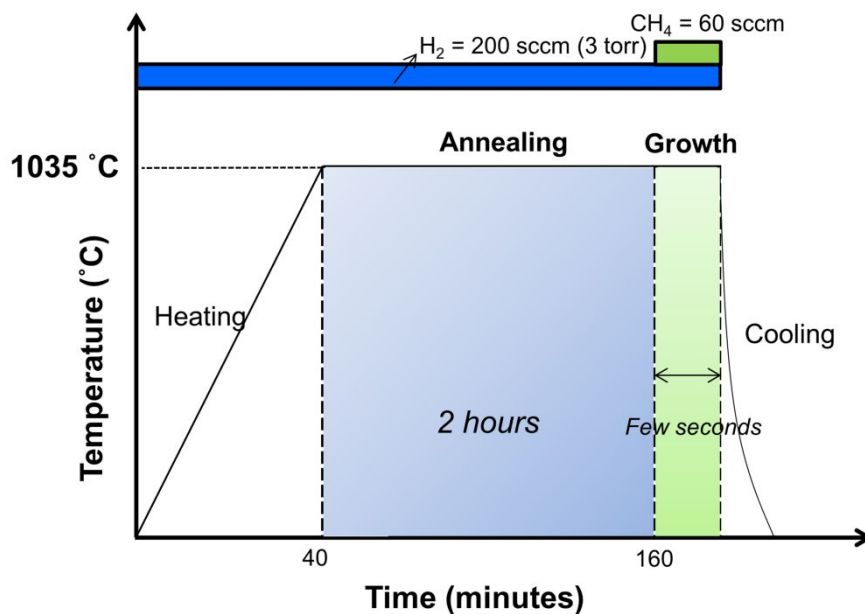


Figure S1. Schematic graph of graphene growth condition which shows fast growth of graphene with high quality. Hydrogen annealing process is proceeded at 1035 °C under ~ 3 torr high-pressure hydrogen (HPH) for 2 hours and subsequent growth is conducted in a few seconds.

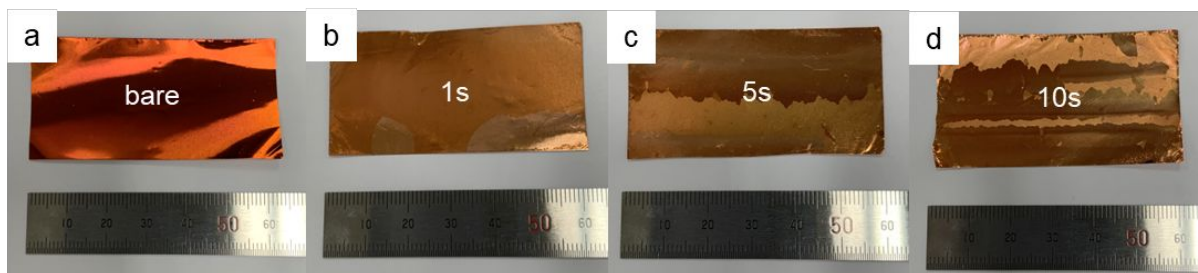


Figure S2. Photographs of 6 cm x 3 cm Cu foils after oxidation experiments at 200°C for 5 min. (a) Bare Cu foil, (b) a second, (c) 5 seconds, and (d) 10 seconds graphene grown Cu foils. Graphene-covered foils show passivation of Cu oxidation compared to bare foil. All foils with graphene-grown exhibit almost full coverage of 6 cm x 3 cm area.

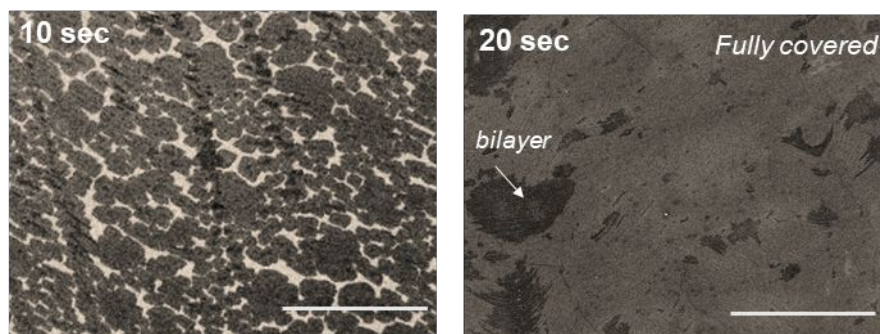


Figure S3. SEM images of graphene grown for 10 secs and 20 secs under LPH conditions. 20 secs grown specimen shows fully covered graphene on Cu foil. Scale bars indicate 10 μm .

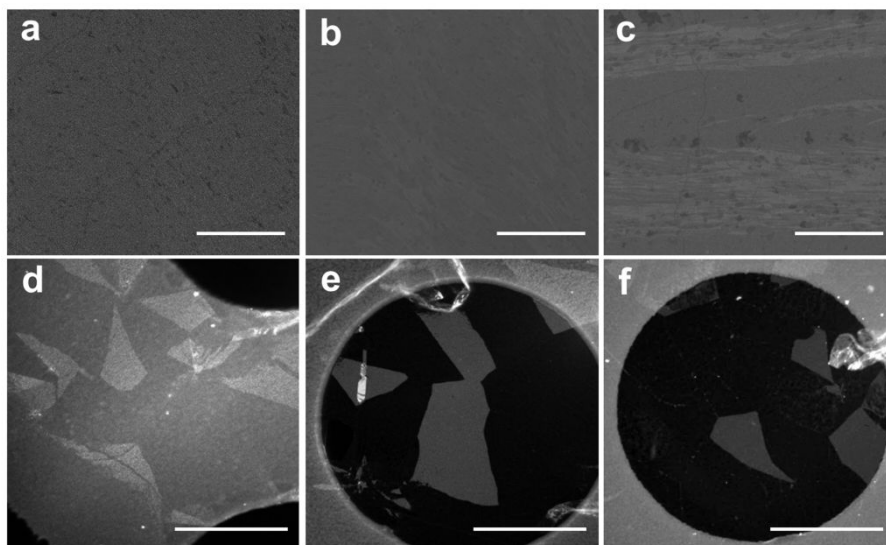


Figure S4. (a-c) SEM images of (a) 10 secs, (b) 3 min, and (c) 1 hour grown graphene on 1 torr hydrogen annealed (IPH) Cu foil for 2 hours. Scale bars in (a-c) indicate 10 μm . Dark field-TEM (DF-TEM) images of graphene grown for (d) 10 secs, (e) 3 min, and (f) 1 hour, respectively. Scale bars in (d-f) indicate 1 μm .

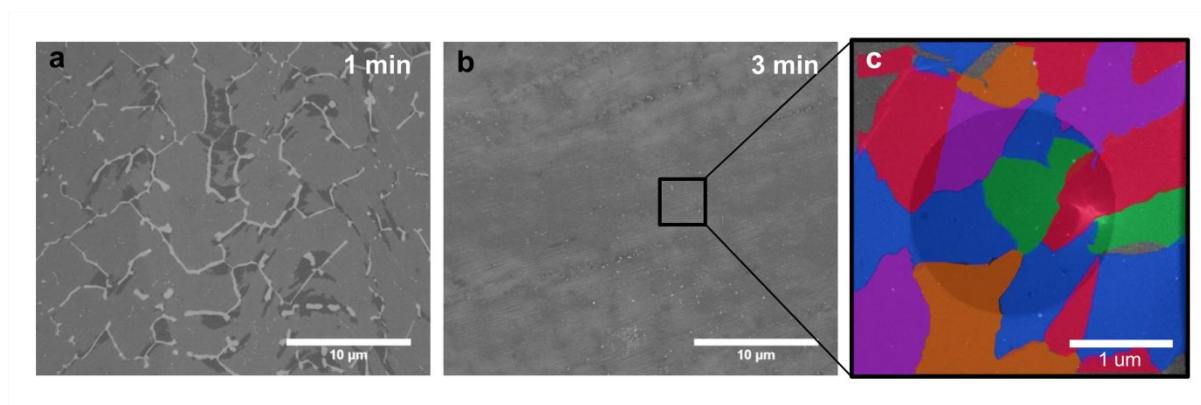


Figure S5. (a-b) SEM images of (a) 1 min and (b) 3 min grown graphene on 3 torr Ar annealed Cu foil. (c) False-colored DF-TEM image in 3 min grown graphene.

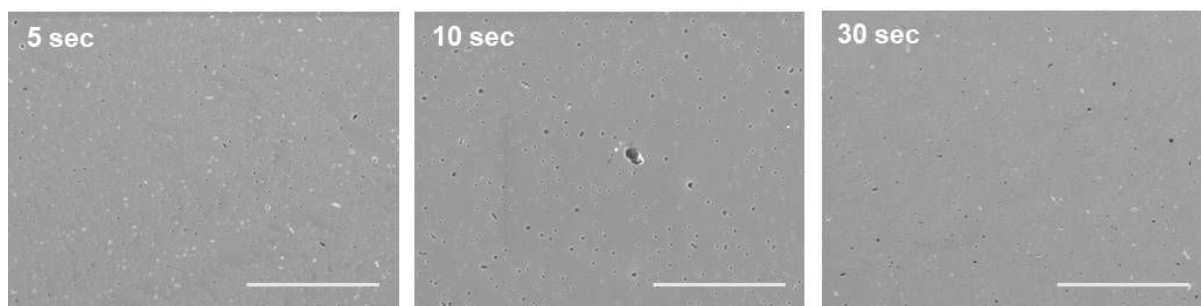


Figure S6. SEM images of graphene grown for different time under 760 torr hydrogen annealed conditions. No graphene is grown in this condition. Scale bars indicate 10 μm .

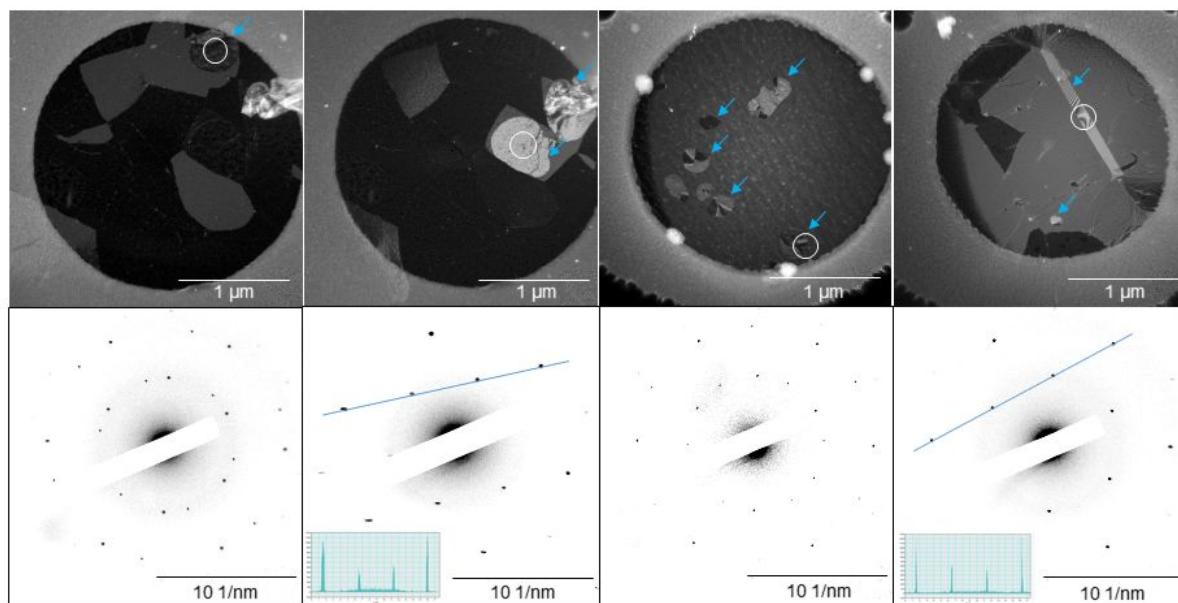


Figure S7. DF-TEM images and selected area electron diffraction patterns (SAED) of bilayer region in graphene grown for 20 secs under LPH and HPH conditions. Two images at left sides are from LPH condition and others are from HPH condition. SAED patterns of individual images are acquired at white circled regions in DF-TEM images and reveal bilayer graphene formation of twisted or bernal-stacked. Blue arrows are all indicate bilayer islands.

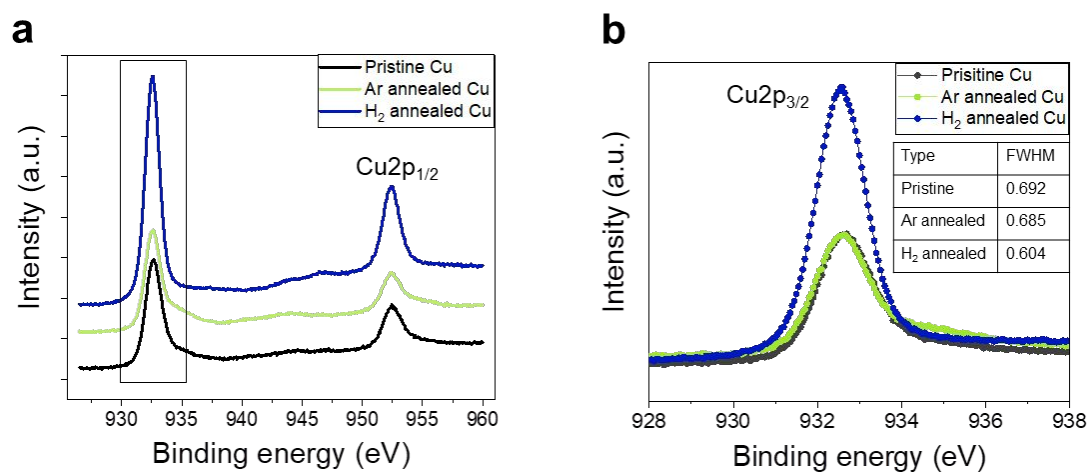


Figure S8. XPS analysis of annealed Cu foils at different annealing condition. (a) Cu 2p spectra and (b) enlarged Cu2p_{3/2} spectra of XPS. Hydrogen annealing is conducted at HPH condition. Inset shows the full width at half maximum values at each condition.

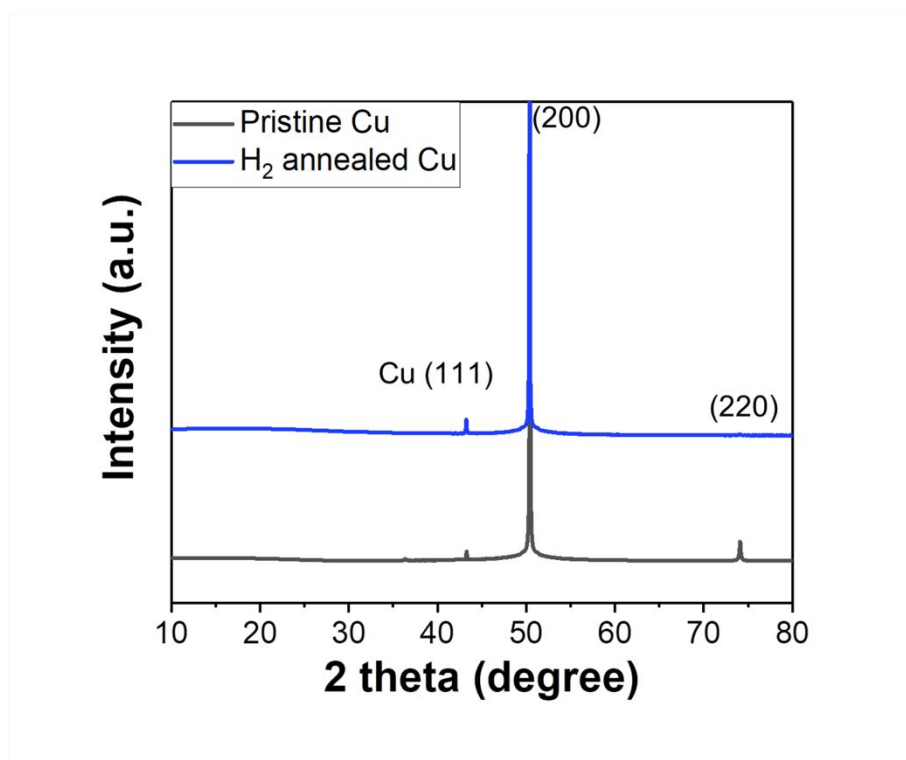


Figure S9. XRD analysis of pristine Cu foil and HPH annealed Cu foil. No special crystallinity difference is observed between pristine Cu foil and hydrogen annealed foil.

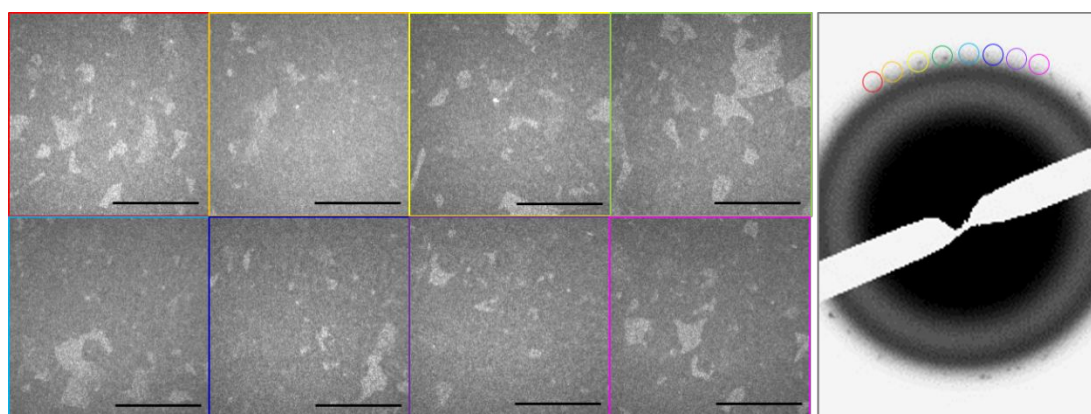


Figure S10. DF-TEM mapping images and corresponding SAED pattern of 1 sec grown graphene at HPH condition. DF-TEM images of different color borders are acquired from diffraction spots marked by corresponding colored circles.

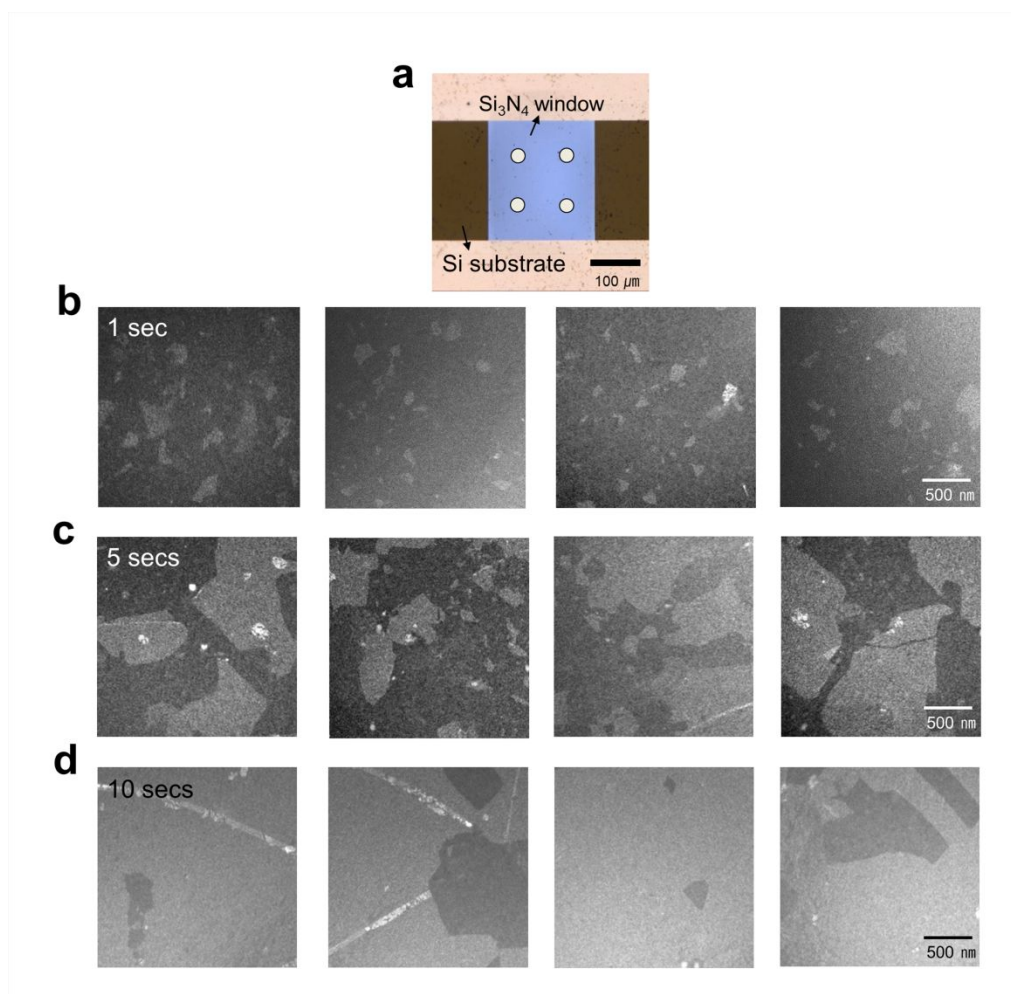


Figure S11. DF-TEM images of graphene observed at different positions of 200 μm x 200 μm Si₃N₄ supporting film. (a) Specially designed chip for observing small grain sizes of graphene. Circles in image indicate TEM observed area. (b-d) TEM images observed in (b) a second, (c) 5 seconds, and (d) 10 seconds grown graphene.

	1 second	5 seconds	10 seconds
Maximum grain size	~ 950 nm	~ 21 μm	~ 160 μm
Ratio of grain sizes less than 200nm	~ 90 %	~ 65 %	~ 25 %

Figure S12. Maximum grain size measurements at different time grown graphene during recrystallization process.

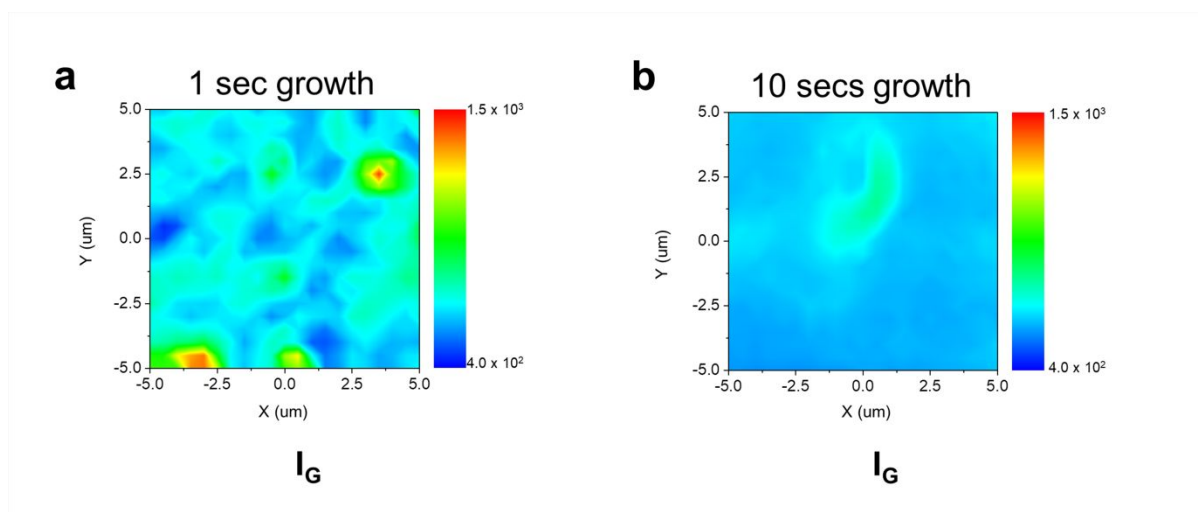


Figure S13. Two-dimensional I_G Raman spectroscopy mapping of graphene grown on HPH annealed Cu foil for (a) a second and (b) 10 seconds. Laser excitation wave length is 514 nm.