Supporting Information

Direct Growth of Graphene Pad on Exfoliated Hexagonal Boron Nitride Surface

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Experimental details

Growth of graphene pad

Hexagonal boron nitride (h-BN) powder was mechanically exfoliated by scotch tape method and transferred onto Si (100) wafer coated with 300 nm thermal oxide. The sample substrate was placed in a quartz tube, which was then located at the center of tube type heating furnace. The 1’ quartz tube was flushed with H_2 gas (50 sccm) for 15 min to remove air, and heated to 1000 ^oC under 50 sccm of H_2 gas at atmospheric pressure. When the furnace reached at 1000 ^oC, 50 sccm of CH_4 gas was introduced for a designated reaction time. The sample was cooled down by opening the cover of tube furnace under H_2 gas flow at 50 sccm. For various control experiments, gas flow rates and reaction temperatures were changed

Characterization

A tapping mode atomic force microscope (AFM, Nanoscope IIIa, Digital Instrument Inc.) was used to investigate the morphology and surface roughness. Histograms of surface roughness of bare h-BN, bare SiO_2/Si, and graphene on h-BN were constructed using the standard deviation of surface height distribution that was directly measured by AFM from 0.6 × 0.6 μm^2 regions of all samples. Using a micro-Raman spectroscope (Alpha 300R, WItec), Raman spectra and mapping images were obtained. Mapping images were obtained in regions 1250 – 1450 cm^{-1} for E_{2g} vibration mode of h-BN and D peak of graphene, 1500 – 1700 cm^{-1} for G peak, and 2550 – 2850 cm^{-1} for G' peak of graphene. The wavelength of diode laser was 532 nm, and the power of laser was adjusted to ~3 mW to prevent possible thermal damage. Spatially-resolved photoelectron spectroscopy (SR-XPS) data were acquired from Pohang Accelerate Laboratory (PAL, Beamline 8A1). X-ray beam (620 eV) was focused by a Fresnel zone plate and exposed to a sample in ultra high vacuum (2.0×10^{-10} torr)

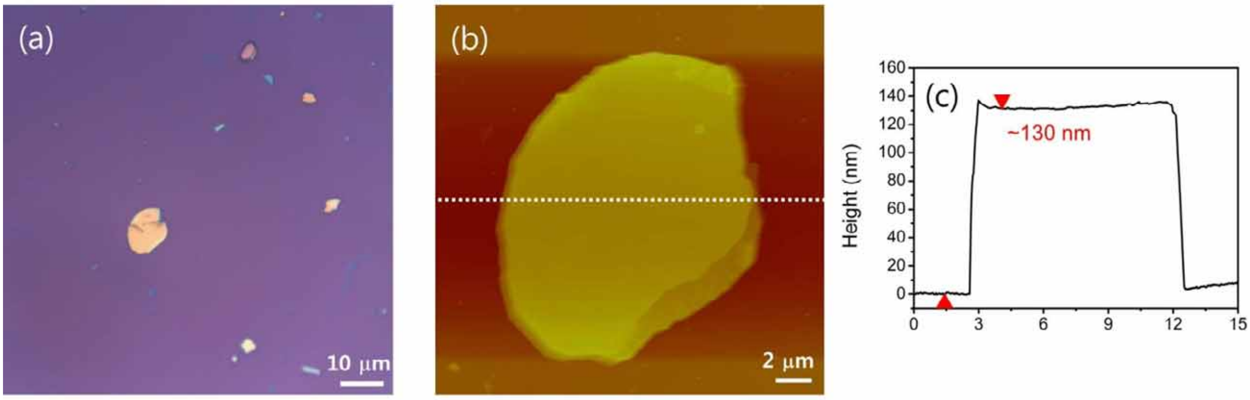

Fig. S1. (a) Optical image of exfoliated h-BN disks on a SiO_2/Si substrate. (b) Representative AFM image of an exfoliated h-BN disk. (c) Height profile along the white dotted line indicated in (b).

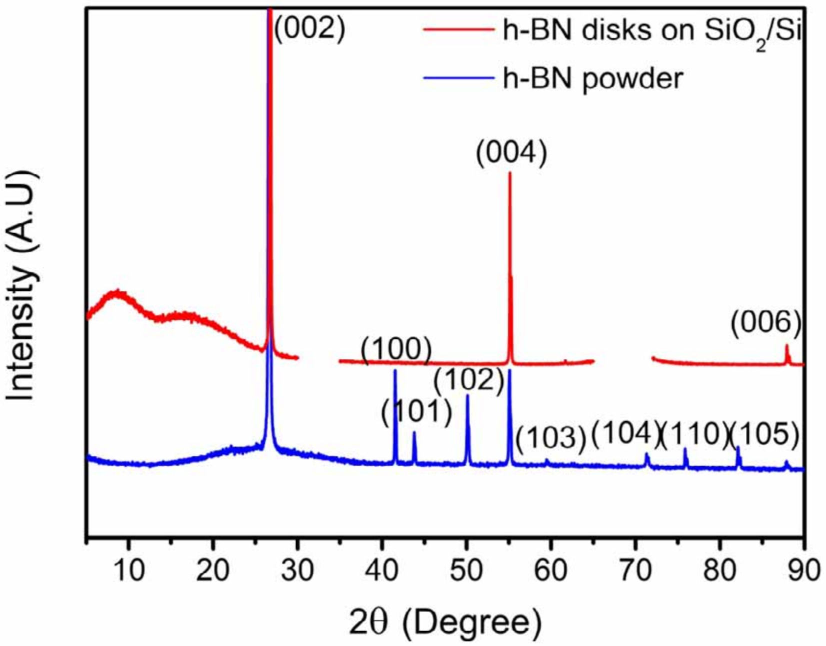

Fig. S2. Powder X-ray diffraction data of exfoliated h-BN (red) and h-BN powder (blue). The exfoliated h-BN only shows (002), (004), and (006) peaks.

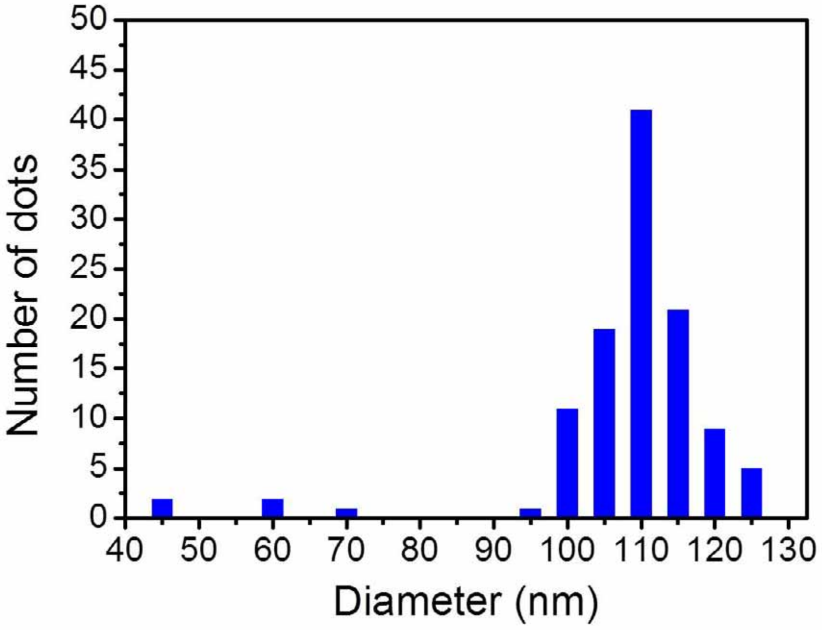

Figure S3. Size distribution of graphene pad grown for 120 min.

Quantitative XPS analysis

The atom fraction of oxygen in a graphene pads can be written as an Eq 1. (C_O is atom fraction of oxygen, I_{C1s} is area intensity of C 1s peak, I_{O1s} is area intensity of O 1s peak, S_C is atomic sensitivity factor of carbon (0.296), S_O is atomic sensitivity factor of oxygen (0.719)).[1]

$$C_O = \frac{\left(I_{O1s,graphene\ on\ h-BN} - I_{O1s\ on\ bare\ h-BN}\right)/S_O}{\left(I_{C1s,graphene\ on\ h-BN} - I_{C1s,on\ bare\ h-BN}\right)/S_C + \left(I_{O1s,graphene\ on\ h-BN} - I_{O1s,on\ bare\ h-BN}\right)/S_O}$$

(Eq. 1)

Bare h-BN shows carbon and oxygen peaks because of the surface contaminants (Fig. S4). Thus, to calculate atom fraction, C 1s and O 1s peak areas should be subtracted as Eq 1. The obtained atom fraction (3 %) was converted to the weight percentage by using Eq. 2. (C_O and C_C are atom fraction of oxygen and carbon. M_O and M_C are atomic weight of oxygen and carbon)

$$\text{Wt \% of oxygen} = \frac{C_O \times M_O}{C_O \times M_O + C_C \times M_C} \times 100\ (\%)$$

(Eq. 2)

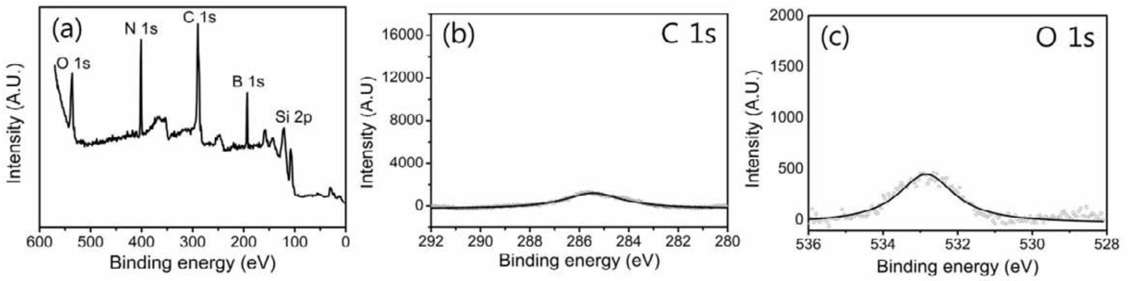

Fig. S4. Spatially resolved X-ray photoelectron spectroscopy (SR-XPS) of bare h-BN. (a) survey spectrum (a) Survey spectrum from 580 to 0 eV showing the O 1s, N 1s, C 1s, B 1s, and Si 2p. (b) SR-XPS spectrum around the C 1s showing the small amount of contaminant carbons. (c) SR-XPS spectrum around the O 1s.

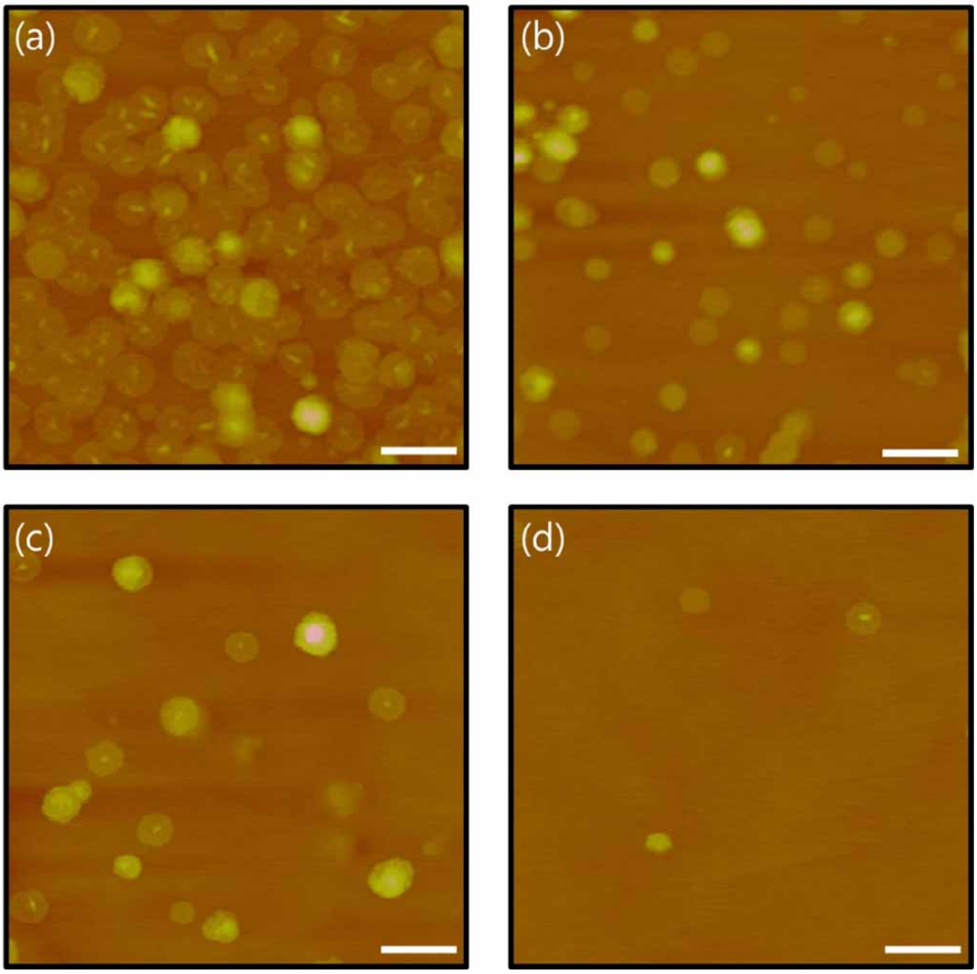

Fig. S5. The effect of temperature on the growth of graphene pad on h-BN. AFM images of graphene pads grown at (a) 1000 °C, (b) 980 °C, (c) 950 °C, and (d) 900 °C. Reaction time and precursor gas amount were fixed at 120 min and $CH_4 : H_2 = 50 : 50$ sccm, respectively. All scale bars are 200 nm. Note that (a) is identical data to the one in Fig. 1(a).

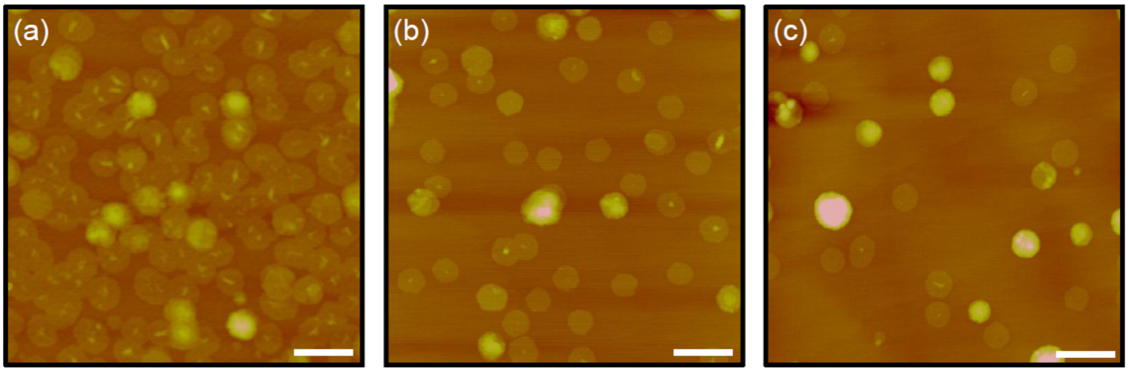

Fig. S6. The effect of precursor gas amount on the growth of graphene pad on h-BN. AFM images of graphene pads grown at the CH_4/H_2 ratio of (a) 50/50 sccm, (b) 40/50 sccm, and (c) 30/50 sccm. Reaction time and temperature were fixed at 120 min and 1000 ^{o}C, respectively. Note that (a) is identical data to the one in Fig. 1(a).

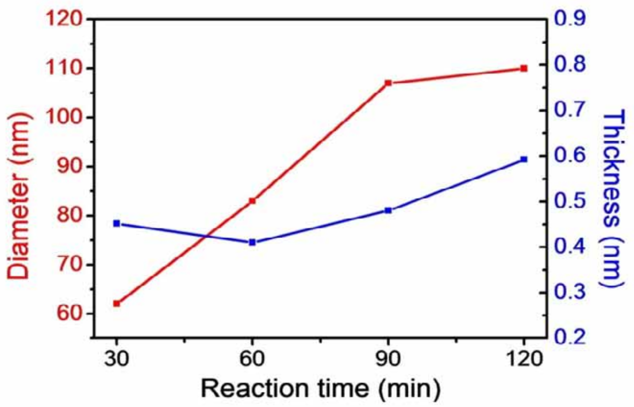

Fig. S7. Changes of average diameter and thickness of graphene pad as a function of reaction time (measured by AFM).

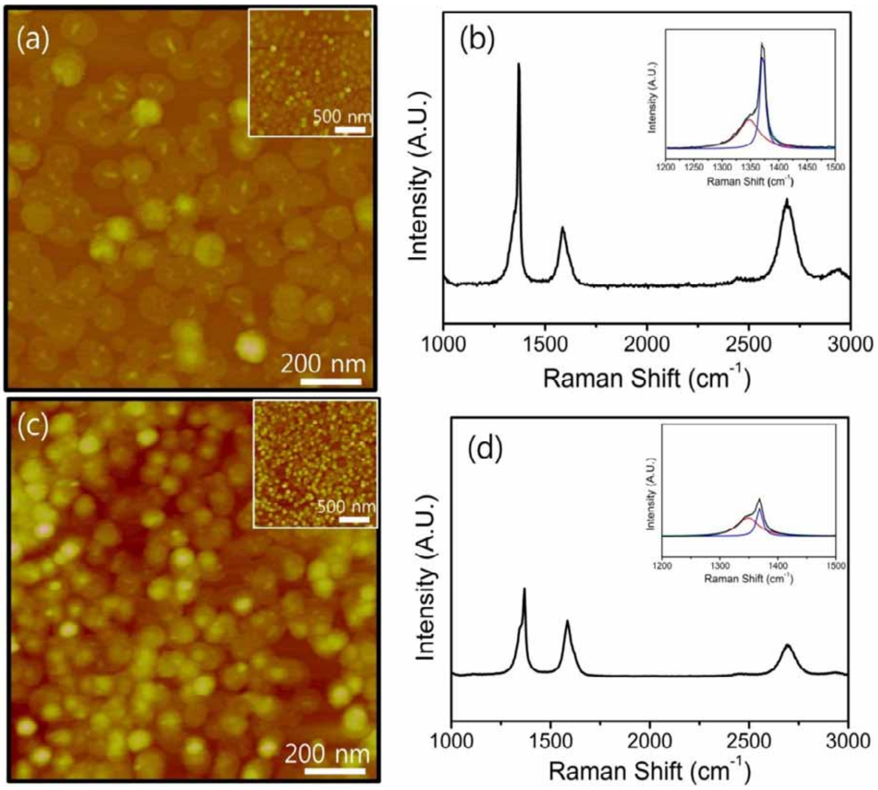

Fig. S8. AFM images of graphene pads grown for (a) 120 min and (b) 150 min. (b) and (d) Raman spectrum of (a) and (c), respectively. Insets in (c) and (d) are deconvoluted D peak and E_{2g} of h-BN. Peaks are normalized by G peak intensities. Note that (a) is identical data to the one in Fig. 1(a).

References

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1 J. F. Moulder, W. F. Stickle, P. E. Sobol, K. D. Bomber, in *Handbook of X-ray Photoelectron Spectrosopy 2nd ed.* Perkin-Elmer, Eden Prairie, **1992**, pp. 25-26.