Electronic Supplementary Information for

Two-Dimensional Hexagonal Boron-Carbon-Nitrogen Atomic Layers

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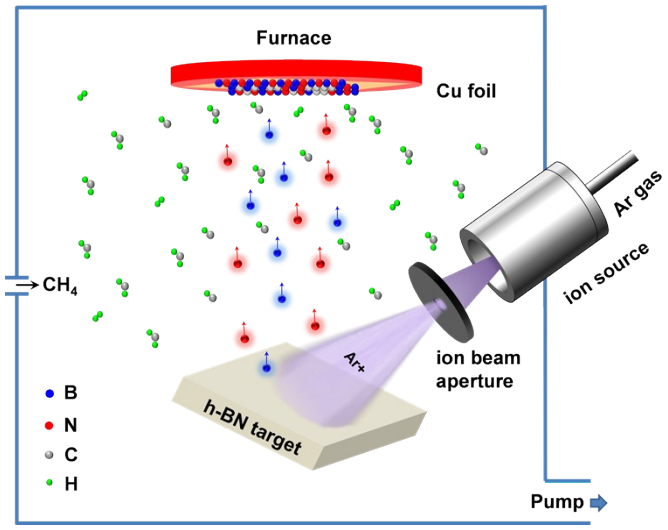

Fig. S1. Schematic diagram of the IBSD system. Boron and nitrogen species were sputtered from a pure h-BN target by a 1.0-keV Ar ion beam, while CH_4 was introduced into the sputtering atmosphere during the growth process. The beam aperture is situated between ion source and target, and the sputtering rate of BN can be adjusted by using ion beam aperture with different sizes. Thus, it provides an alternative approach for controlling C content. In this work, two ion beam apertures with different diameters (10 and 20 mm) were adopted.

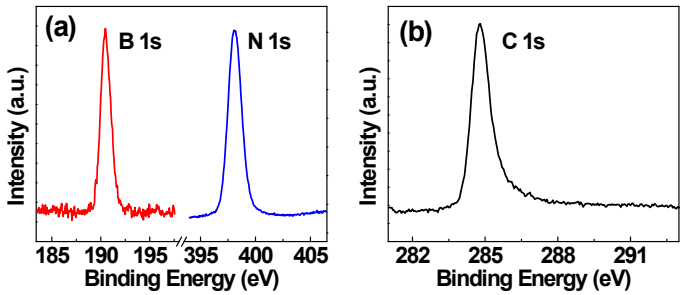

Fig. S2. XPS core level spectra of (a) B 1s and N 1s for pristine h-BN, (b) C 1s for pristine graphene.

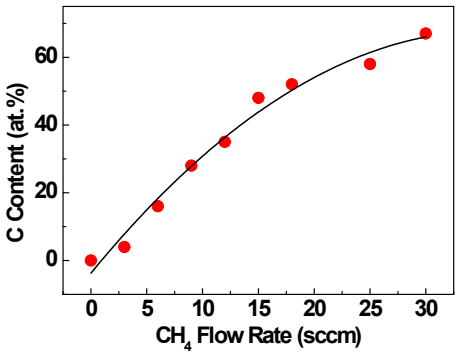

Fig. S3. Dependence of the C content in h-BCN layers on the CH_4 flow rate.

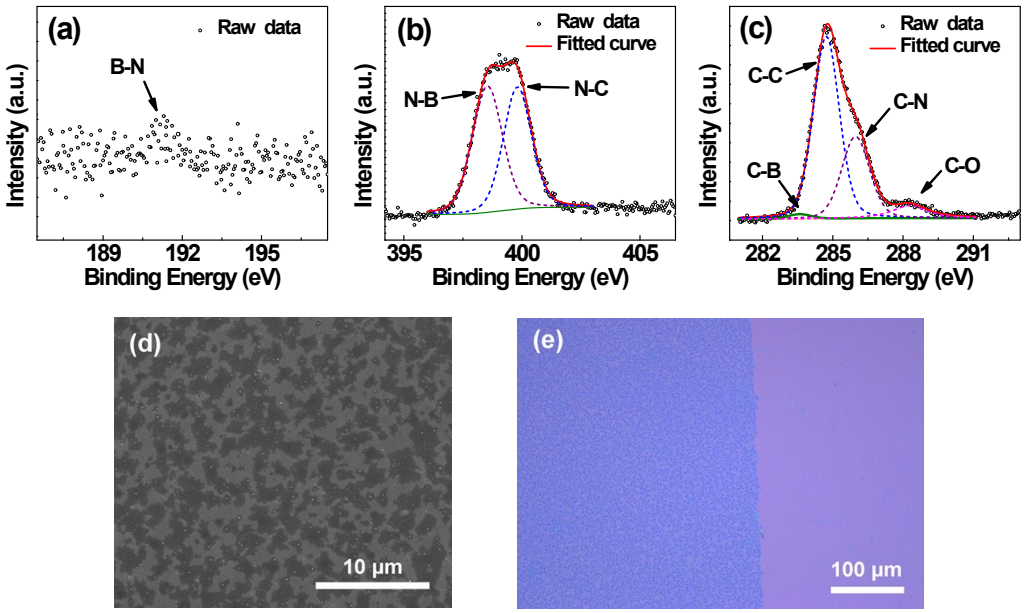

Fig. S4. Characterization of the h-BCN layer with 82 at.% C. XPS core level spectra of (a) B 1s, (b) N 1s, and (c) C 1s for the h-BCN sample grown with an ion beam aperture of 10 mm and 20 sccm CH_4. The C content is determined to be 82 at.% in this sample. (d) SEM image of the h-BCN film with 82 at.% C. The formation of segregated graphene domains embedded in h-BCN matrix was observed. (e) Optical microscopy image of h-BCN film with 82 at.% C on a SiO_2/Si substrate.

In the present IBSD system, the maximum allowable CH_4 flow rate is about 30 sccm, because when a large amount of gas is introduced into vacuum chamber, the raised pressure will lead to the abnormal discharge of Kaufman ion source. As a result, the maximum C content is about 50%, when the larger ion beam aperture of 20 mm is adopted. In order to further increase C content, a small ion beam aperture of 10 mm was used. As illustrated in Fig. S1, the smaller beam aperture situated between ion source and target can reduce sputtering rate of BN, while the dissociation efficiency of CH_4 remains unchanged since the ion current density keeps a constant value. Therefore, the h-BCN layers with full range of compositions can be prepared by adjusting the CH_4 flow rate and the size of ion beam aperture.

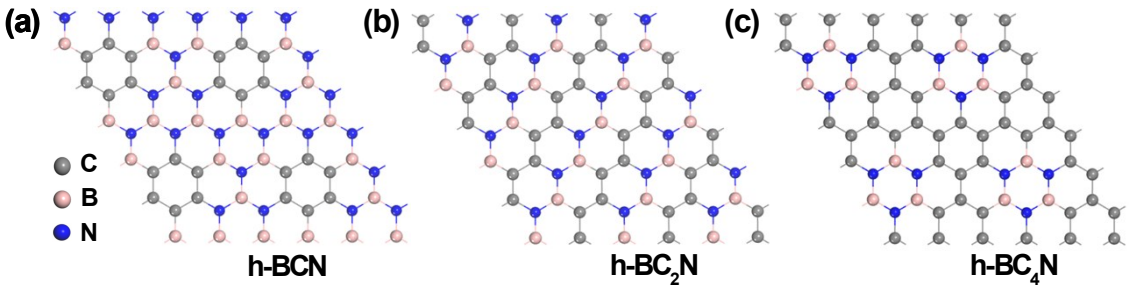

Fig. S5. Geometries of (a) h-BCN, (b) h-BC_2N, and (c) h-BC_4N with the lowest formation energies with respect to the chemical potential of h-BN and graphene.

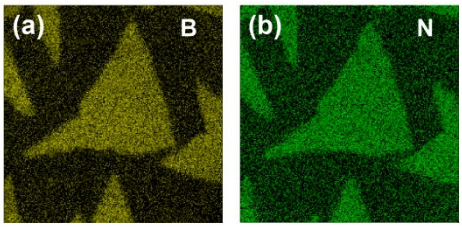

Fig. S6. (a) B (KLL) and (b) N (KLL) Auger electron maps of pristine h-BN flakes.

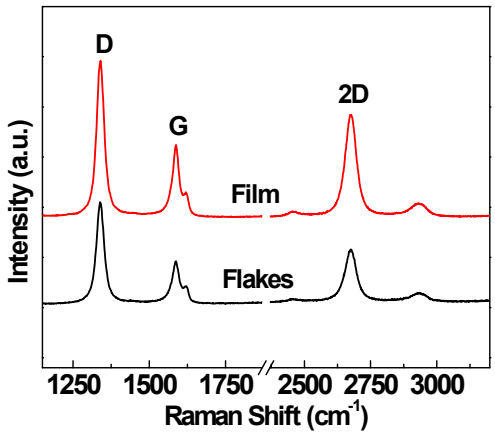

Fig. S7. Raman spectra of pristine graphene flakes and continuous film.

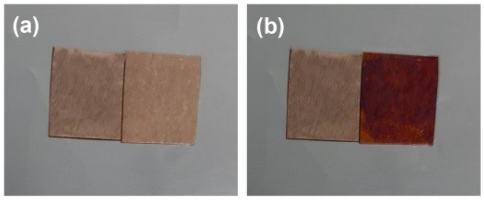

Fig. S8. Photograph of two samples on Cu foils (a) before and (b) after oxidation treatment in air. The two samples were grown under the same conditions (1050 ^{o}C, 15 sccm CH_4) with the sole exception that the Kaufman ion source was switched off (the right sample) or on (the left sample) during the process. The change in color indicates that there was no graphene coating (the right sample), while the graphene coating (the left sample) can protect the Cu foil from oxidation at 500 ^{o}C.

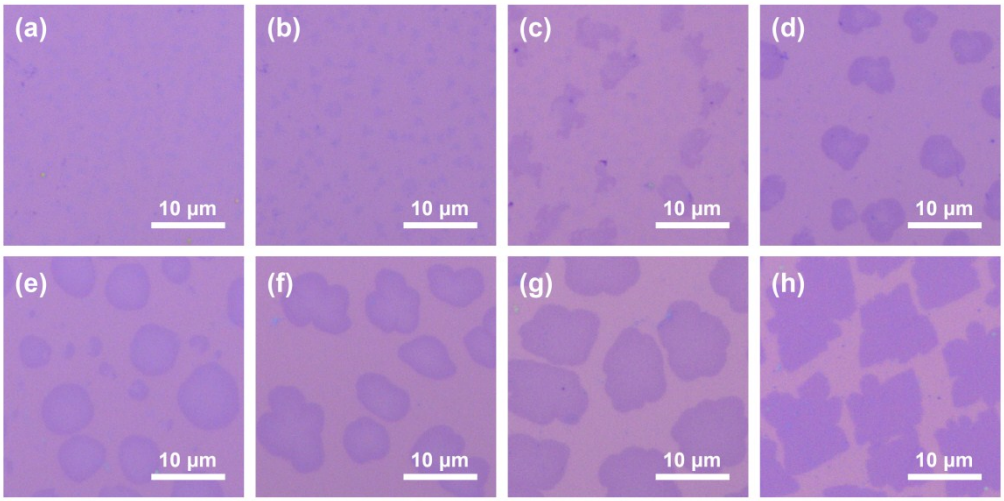

Fig. S9. Optical microscopy image of (a) pristine h-BN, and h-BCN flakes with (b) 4 at.%, (c) 16 at.%, (d) 35 at.%, (e) 48 at.%, (f) 58 at.%, (g) 67 at.%, as well as (h) pristine graphene transferred on SiO_2/Si substrates.

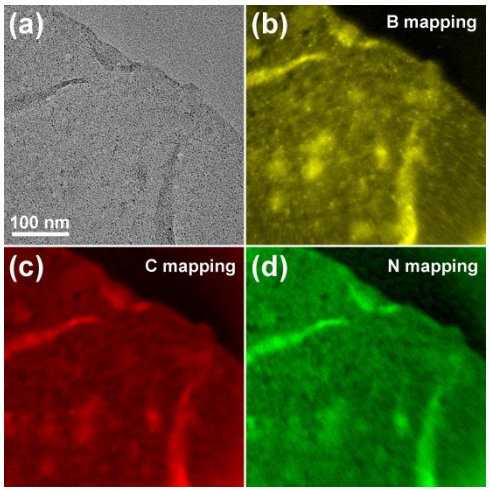

Fig. S10. (a) TEM image of an h-BNC film with 48 at.% C. The corresponding EELS mapping for (b) B, (c) C and (d) N in the h-BCN film.

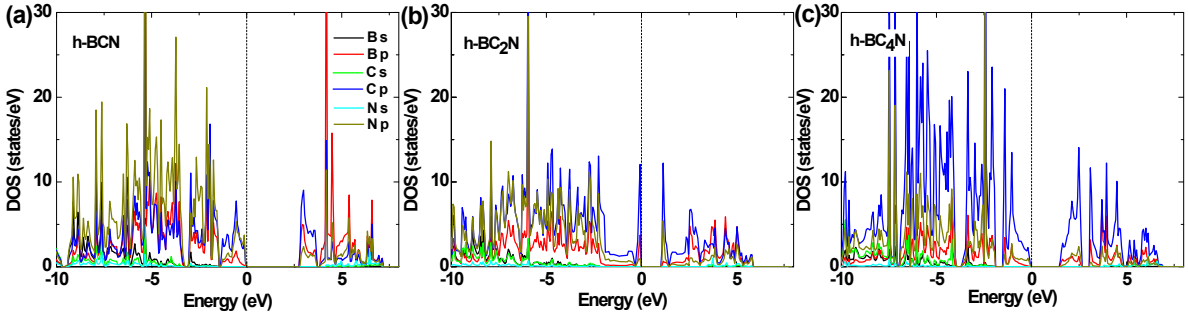

Fig. S11. The density of states (DOSs) of the (a) h-BCN, (b) h-BC_2N, and (c) h-BC_4N.

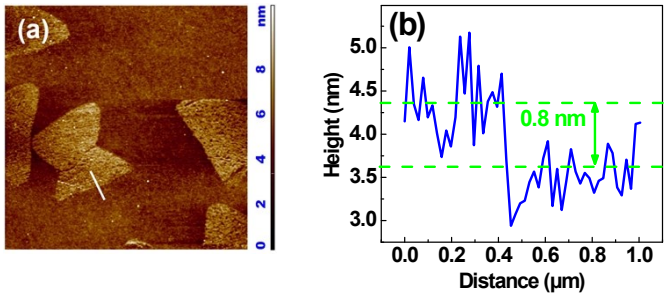

Fig. S12. (a) AFM image of pristine h-BN flakes transferred onto a SiO_2/Si substrate. (b) Height profiles along the white lines drawn in (a), showing a thickness of 0.8 nm.

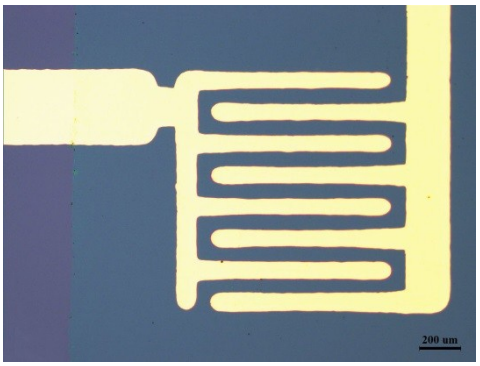

Fig. S13. Optical microscopy image of the h-BCN layers with interdigitated Ti/Au electrodes.