Electronic Supplementary Information

## **High-Pressure Hydrogenation of Graphene: Towards Graphane**

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Figure S1. SEM micrographs of (A) G-H: [60 bar/220 °C], (B) G-H: [100 bar/200 °C], (C) G-H: [150 bar/220 °C], (D) G-H: [100 bar/350 °C], (E) G-H: [100 bar/500 °C] and (F) G-Ar: [100 bar/350 °C]. Scale bars (a) 10 μm, (b) 1 μm and (c) 100 nm.



Figure S2. TEM images of (A) G-H: [60 bar/220 °C], (B) G-H: [100 bar/200 °C], (C) G-H: [150 bar/220 °C], (D) G-H: [100 bar/350 °C], (E) G-H: [100 bar/500 °C] and (F) G-Ar: [100 bar/350 °C]. Scale bars (a) 200 nm, (b) 50 nm.



Figure S3. Survey XPS spectra of hydrogenated graphenes under (A) constant temperature of  $\sim 220$  °C and pressure of (a) 60, (b) 100 and (c) 150 bar and under (B) constant pressure of 100 bar and varying temperature of (a) 200, (b) 350 and (c) 500 °C. Also shown XPS spectra of graphite oxide exfoliated in Ar atmosphere (d).



Figure S4. High-resolution XPS of hydrogenated graphene materials.