

Supplementary Information

Large-range Control of the Microstructures and Properties of Three-dimensional Porous Graphene

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Figure S1. Working flow of tailoring microstructure of graphene sponge by freeze casting.

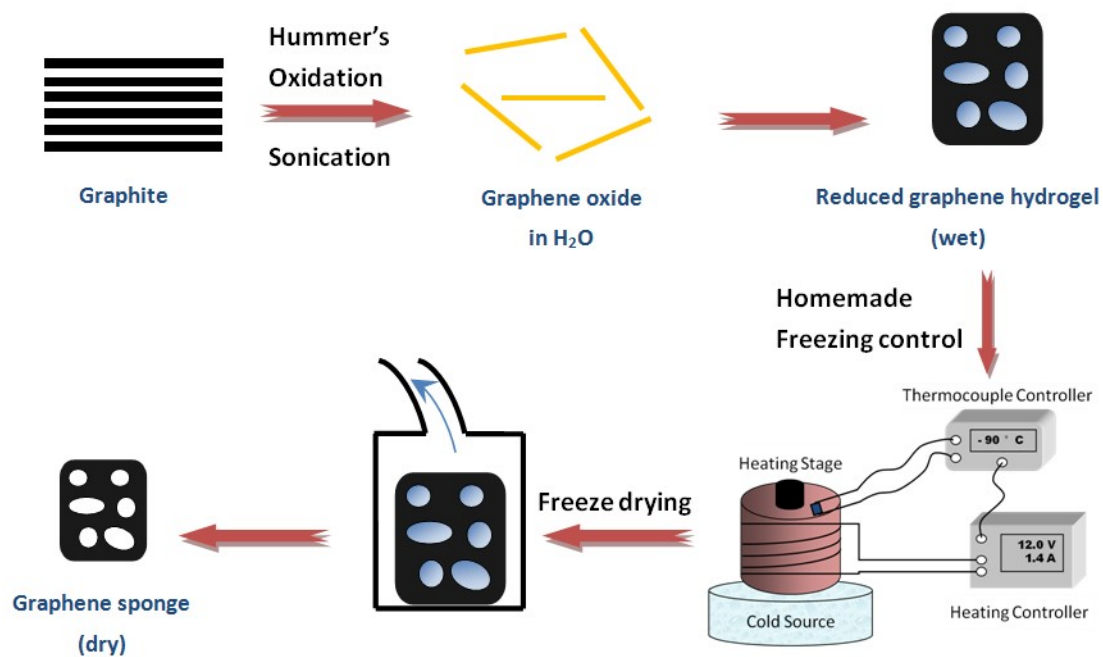


Figure S2. Higher magnification SEM images of the graphene walls in the graphene sponges. Freezing temperature: **a**, -10 °C; **b**, -20 °C; **c**, -40 °C; **d**, -50 °C; **e** and **f**, -170 °C, respectively.

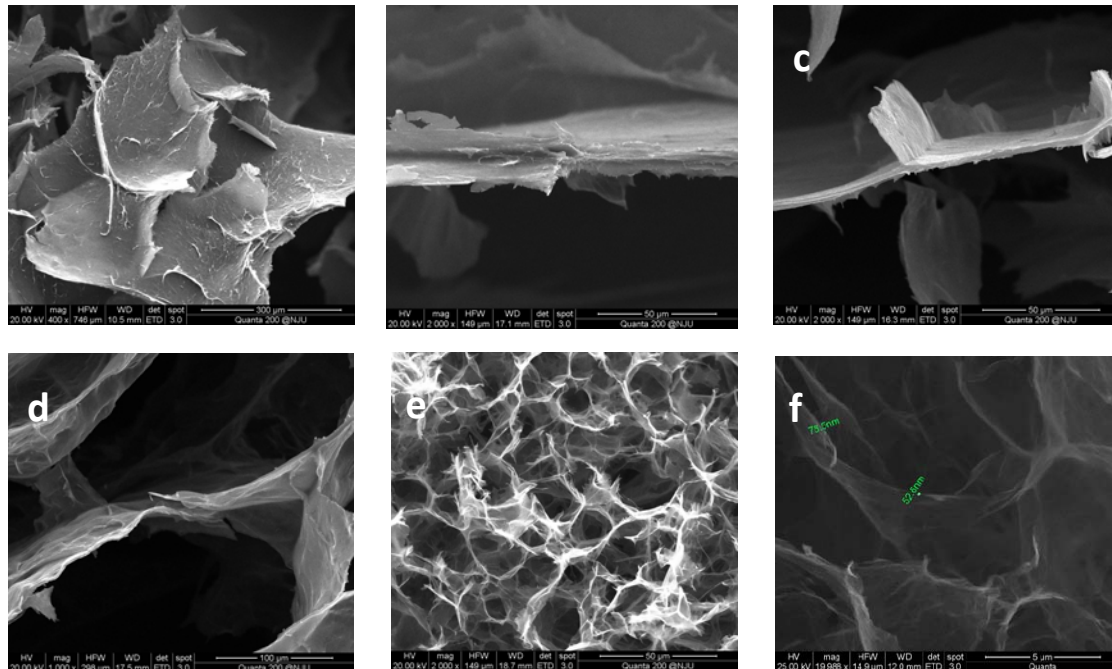


Figure S3. The relationship between nucleation/crystal growth and the morphology of the pore. The specimen was frozen at **a**, $-20\text{ }^{\circ}\text{C}$ and **b**, $-170\text{ }^{\circ}\text{C}$, showing **a**, unidirectional, lamellar growth and **b**, diffusive, uniform micropores, respectively.

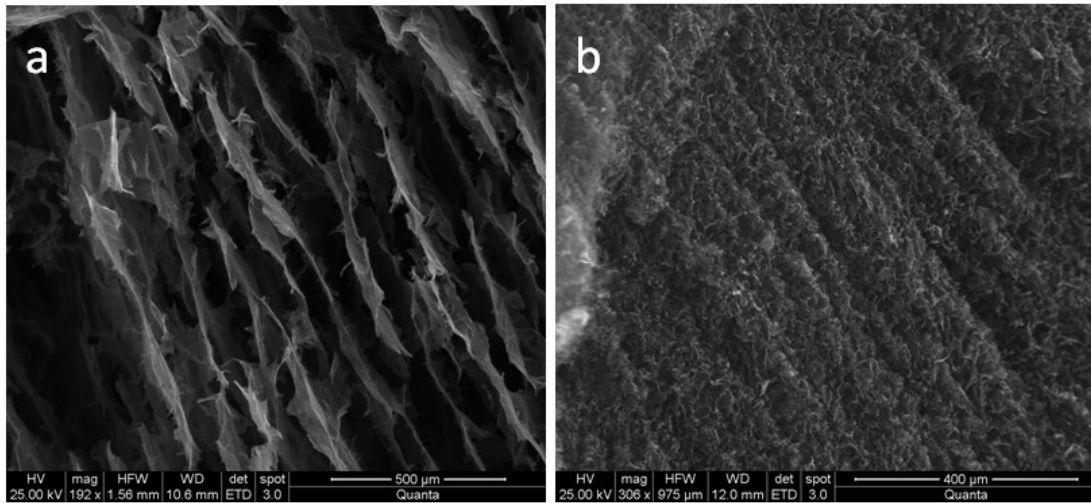
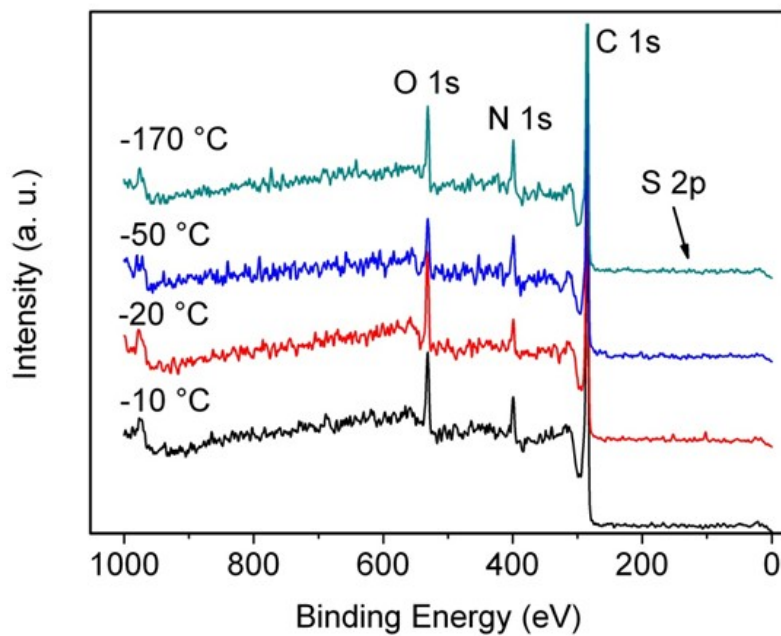


Figure S4. XPS spectra of the cross-sectional surfaces of graphene sponges which have been frozen in $-10\text{ }^{\circ}\text{C}$, $-20\text{ }^{\circ}\text{C}$, $-50\text{ }^{\circ}\text{C}$ and $-170\text{ }^{\circ}\text{C}$, respectively. No significant difference in chemical composition has been observed.



Movie S1. Mechanical compression and release of graphene sponge frozen in $-20\text{ }^{\circ}\text{C}$

Movie S2. Mechanical compression and release of graphene sponge frozen in $-170\text{ }^{\circ}\text{C}$