

## Supporting Information

### Porous B-doped Graphene Inspired by Fried-Ice for Supercapacitors and Metal-Free Catalysts

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The specific capacitance of these samples from galvanostatic charge/discharge curves are calculated as

$$C_s = \frac{I\Delta t}{m\Delta V}$$

where  $I$  is the constant current and  $m$  is the total mass for both carbon electrodes,  $\Delta t$  is the discharge time and  $\Delta V$  is the voltage change during the discharge process.

The specific capacitance derived from cyclic voltammograms as

$$C_s = \frac{A}{f \cdot \Delta V \cdot m}$$

where  $A$  is the integral areas of the cyclic voltammogram loops,  $f$  is the scan rate,  $\Delta V$  is the voltage window, and  $m$  is the mass of the electrode.

The electron transfer numbers at these three electrodes can be derived from the equation of Koutechy-Levich plot <sup>1,2</sup> showing blow:

$$\frac{1}{j} = \frac{1}{j_k} + \frac{1}{B\omega^{0.5}} \quad (1)$$

in which  $j$  is the current density at appointed voltage,  $j_k$  is the kinetic current and  $\omega$  is the electrode rotating rate (rpm). The parameter  $B$  at different applied voltage could be obtained from the slope of the K-L plots in Figure S7. Meanwhile, the electron transfer number at different voltage is connected with parameter  $B$  according to the Levich equation as following in the alkaline aqueous solution <sup>2</sup>:

$$B = 0.2nF(D_{O_2})^{2/3}\nu^{-1/6}C_{O_2} \quad (2)$$

where  $n$  represents the overall electron transfer number per oxygen molecule,  $F$  is the Faraday constant with the value of  $96485 \text{ C mol}^{-1}$ ,  $D_{O_2}$  is the diffusion coefficient of  $O_2$  in  $0.1 \text{ M KOH}$  ( $1.9 \times 10^{-5} \text{ cm}^2 \text{ s}^{-1}$ ),  $\nu$  is the kinetic viscosity ( $0.01 \text{ cm}^2 \text{ s}^{-1}$ ), and  $C_{O_2}$  is the bulk concentration of  $O_2$  ( $1.2 \times 10^{-6} \text{ mol cm}^{-3}$ ). The constant 0.2 is adopted when the rotation speed is expressed in rpm in alkaline aqueous solution.

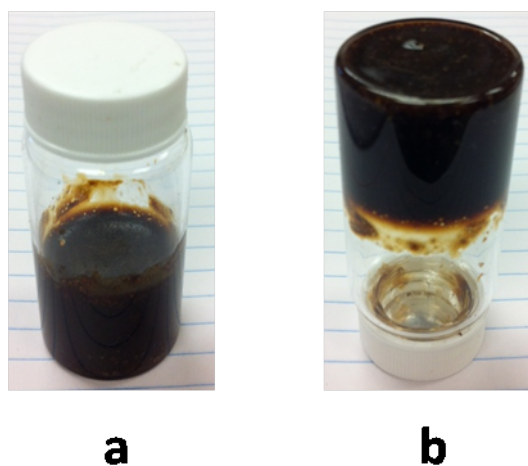


Figure S1. Photo image of the gel-like GO sediment before and after freezing.

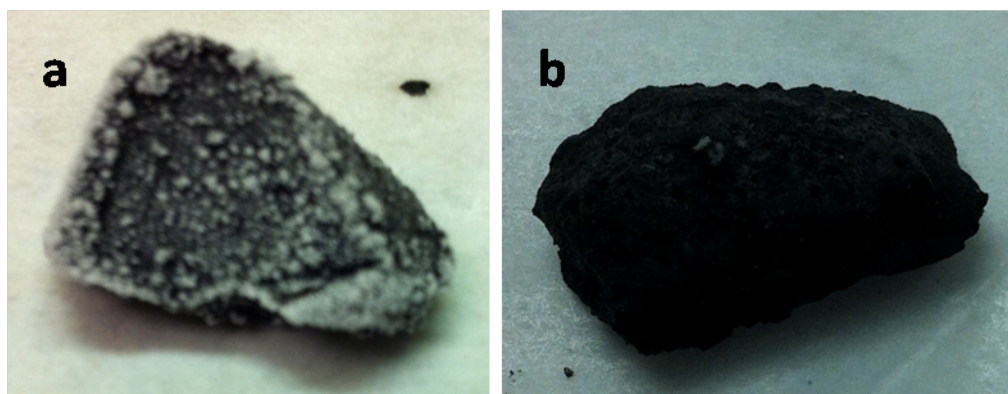


Figure S2. Photo image of the frozen GO monolith before thermal treatment (a) and the as-prepared porous graphene monolith after thermal treatment (b)

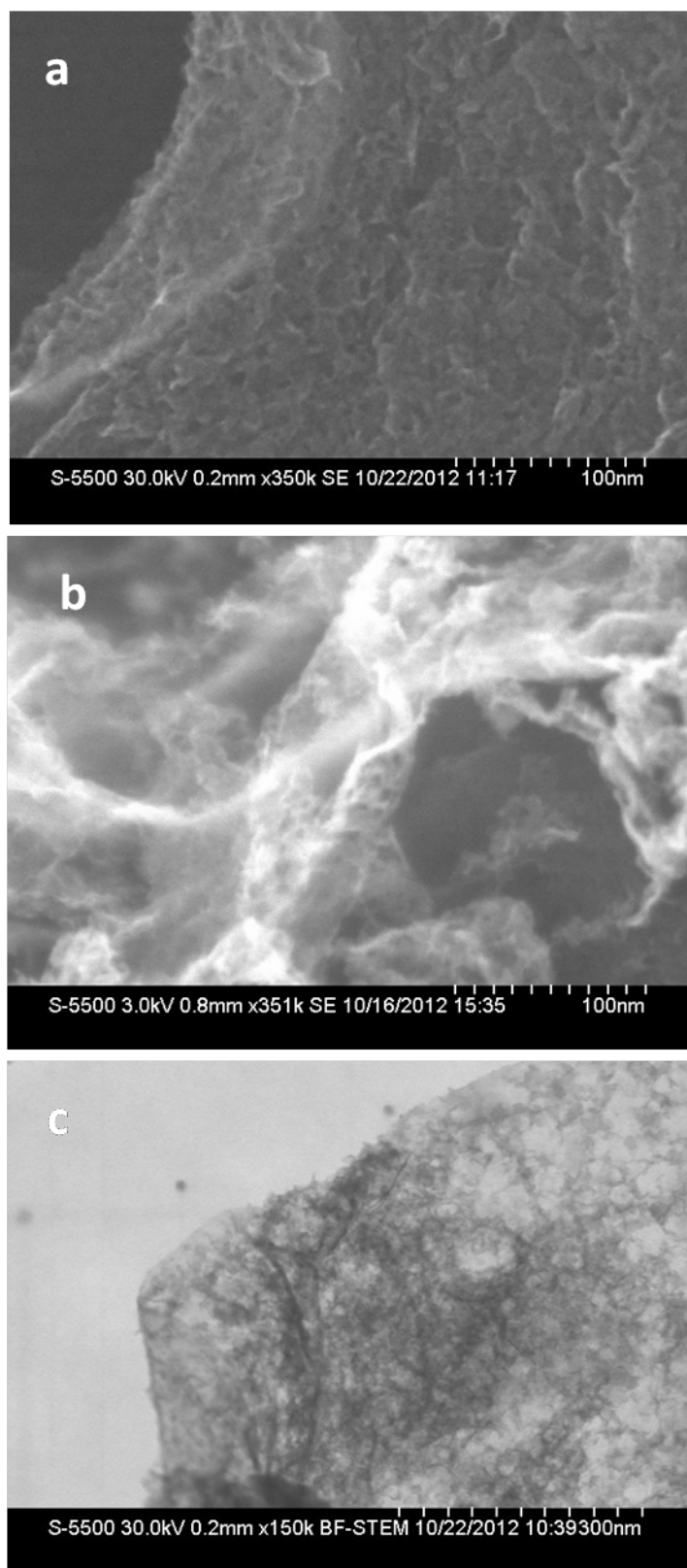


Figure S3. Magnified SEM images of B-G 600 (a), B-G 800 (b), and STEM image of B-G 800 (c).

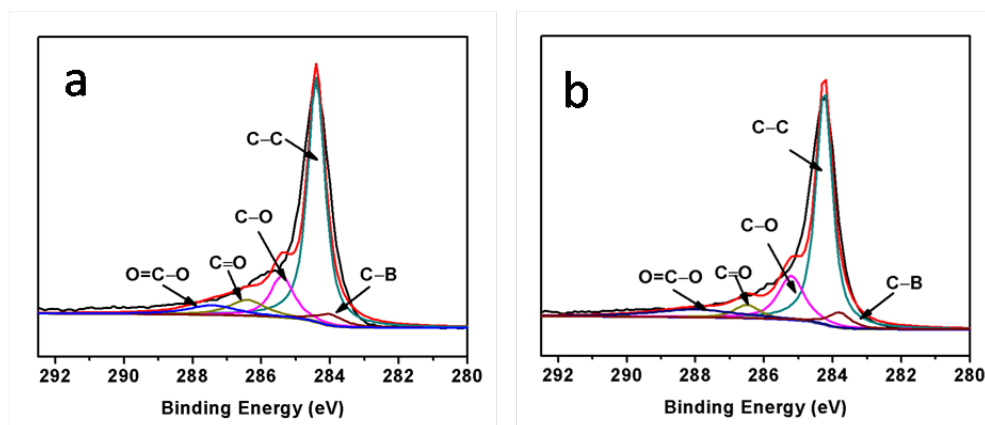


Figure S4. C1s spectrum of (a) B-G 600 and (b) B-G 800.

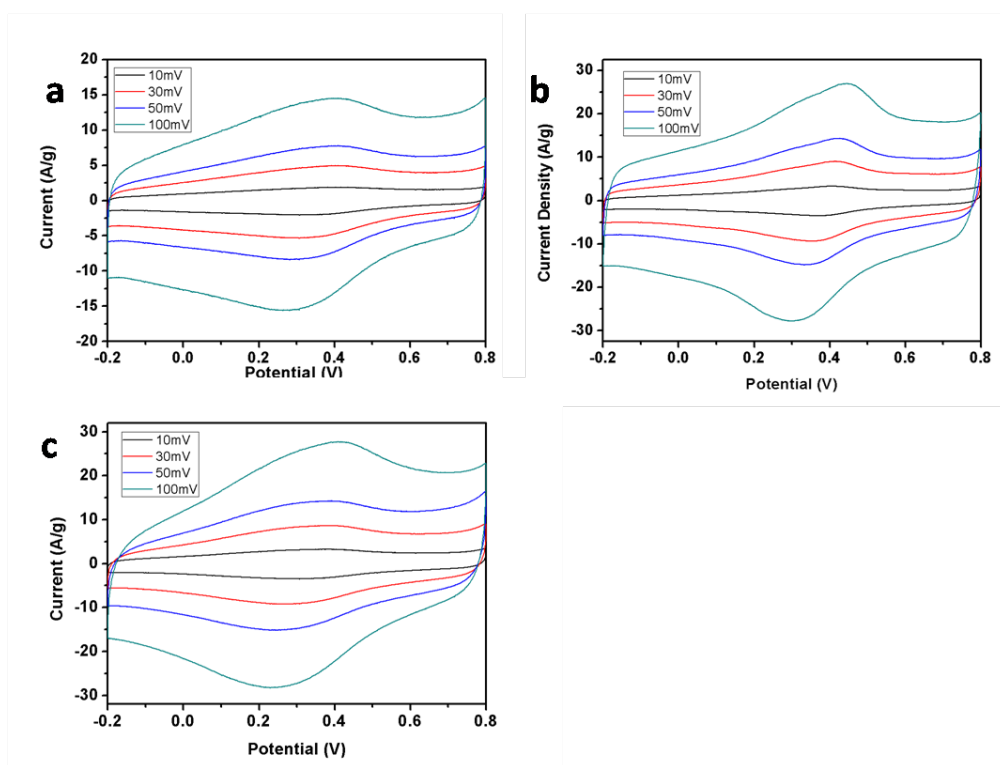


Figure S5. Cyclic voltammetry (CV) curves of 3-D porous graphene framework in a three electrode system in 2 M  $\text{H}_2\text{SO}_4$  solution at the scan rate of 10, 30, 50, and 100  $\text{mV s}^{-1}$ . (a) B-G 400, (b) B-G 600, and (c) B-G 800.

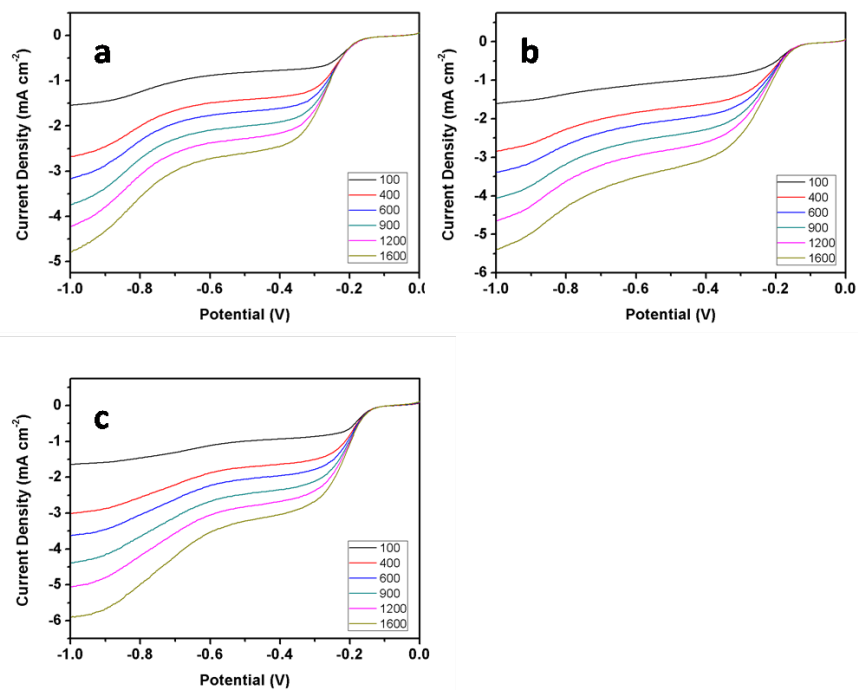


Figure S6. Linear sweep voltammetry curves of ORR at several designed rotation speed in the O<sub>2</sub>-saturated 0.1 M KOH solution with the scan rate of 10 mV s<sup>-1</sup>. a) B-G 400, (b) B-G 600, and (c) B-G 800.

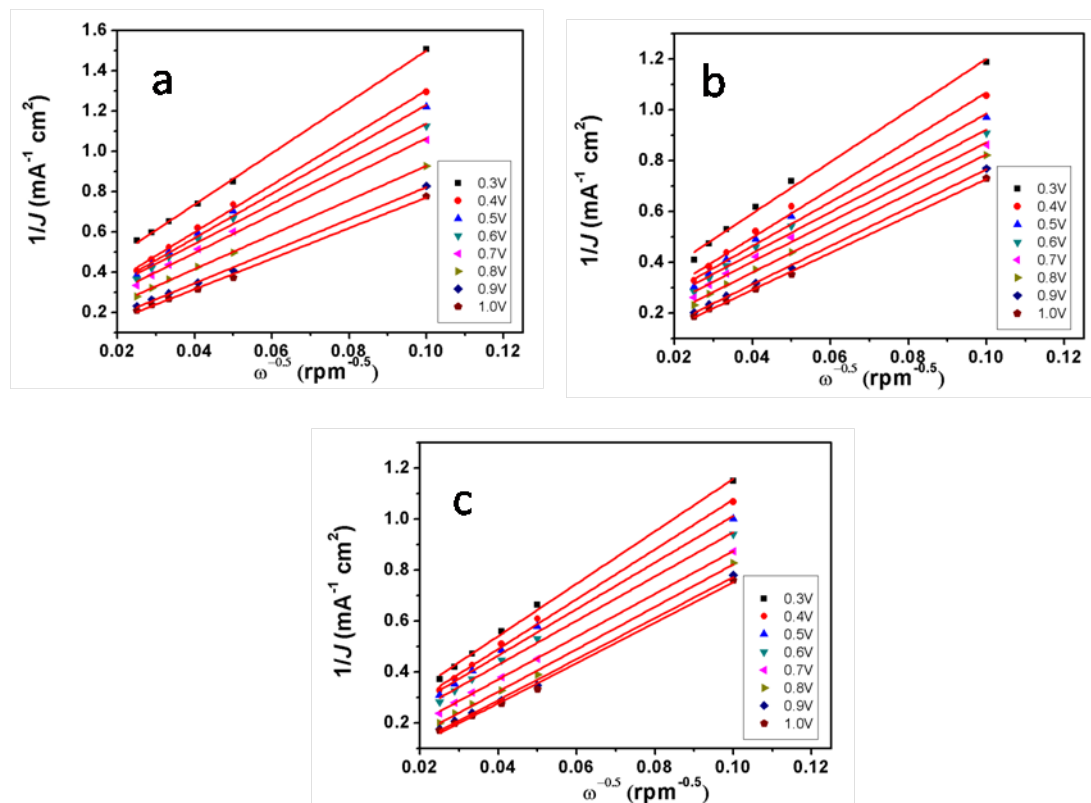


Figure S7. K-L plots of ORR at different applied potential of B-G 400 (a), B-G 600 (b), and B-G 800 (c).

## References:

1. A. J. Bard, L. R. Faulkner, *Electrochemical Methods: Fundamentals and Applications.*; Wiley: New York, 1980.
2. S. Wang, D. Yu, L. Dai, D. W. Chang and J.-B. Baek, Polyelectrolyte-Functionalized Graphene as Metal-Free Electrocatalysts for Oxygen Reduction. *ACS Nano* **2011**, *5*, 6202