## Nitrogen-Doped Holey Graphene Foams for High-Performance Lithium Storage

Jiaguang Sun, Lan Wang, Ranran Song, Shubin Yang\*

Key Laboratory of Aerospace Advanced Materials and Performance of Ministry of Education, School of Materials Science and Engineering, Beihang University, 100191, Beijing, China \*Email:yangshubin@buaa.edu.cn

## 1. Experimental of Absorbance of methylene blue (MB)

The absorption value at 310 nm of methylene blue (MB) ethanol solution was used to evaluate the macroporous surface areas of NHGFs <sup>1</sup> through the UV-vis spectra. Specifically, 0.55 cm<sup>2</sup> of as-prepared Holey graphene foams were added into 0.1 mg mL<sup>-1</sup> MB ethanol solution and kept for 10 days at room temperature to allow adsorption of MB on the surface of holey graphene foams. UV-vis spectra adsorption experiments were recorded after this long-time absorbance and pure MB solution. The surface area can be calculated through the variation of the absorption value at 310 nm of MB ethanol solution. 1 mg of adsorbed MB represents 2.54 m<sup>2</sup> of covered surface area<sup>2</sup>.

## 2. Calculation of diffusion coefficient (D) of lithium

As shown in Figure 6b in main text, Warburg impedance coefficient,  $\sigma_w$  can be facily calculated. And then, the diffusion coefficient (D) of lithium could be calculated by using equation (1):

$$D = R^2 T^2 / 2A^2 n^4 F^4 C^2 \sigma_{\rm w}^2 \tag{1}$$

Where *R* is the gas contant, *F* is the Faraday constant, *T* is the temperature, *A* is the surface area, *n* is the number of moles of charge per ion, and *C* is the maximum ion concentration, the value of  $\sigma_w$  is shown in Table S1.



Figure S1. XRD patterns of NHGFs, se-NGFs, ne-NGFs, HGFs.



**Figure S2.** UV-vis spectra of NHGFs, as compared to se-NGFs and HGFs recorded after 10 days until absorption equilibrium during MB adsorption experiments.



**Figure S3.** Typical cyclic voltammograms of (a) NHGFs, (b) se-NGFs, (c) ne-NGFs and (d) NGFs.



Figure S4. Coulombic efficiency of nitrogen-doped holey graphene foams at 0.1 C.



**Figure S5.** Randles equivalent circuit for NHGFs electrode/electrolyte interface.  $R_e$  is the electrolyte resistance, and  $C_f$  and  $R_f$  are the capacitance and resistance of the surface film formed on the electrodes, respectively.  $C_{dl}$  and  $R_{ct}$  are the double-layer capacitance and charge-transfer resistance, respectively,  $Z_w$  is the Warburg impedance related to the diffusion of lithium ions into the bulk electrodes.



**Figure S6.** a) Cycle performances of NHGFs-800, NHGFs-900 and NHGFs at a current rate of 0.1C. b) Comparison of rate capacities of NHGFs-800, NHGFs-900 and NHGFs.



Figure S7. Nyquist plots of NHGFs-800, NHGFs-900 and NHGFs.

Samples	$\sigma_{ m w}$
NHGFs	38.3
se-NGFs	40.1
ne-NGFs	64.2
HGFs	47.4

**Table S1.** Warburg impedane coefficient,  $\sigma_w$ , derived from the slope of each line in Fig. 6 in main text.

Samples	R <sub>e</sub> (Ohms)	$R_{\rm f}({\rm Ohms})$	$R_{\rm ct}$ (Ohms)
NHGFs	4.0	9.4	26.7
se-NGFs	5.5	11.1	32.0
ne-NGFs	9.3	14.0	45.8
HGFs	5.3	12.9	39.4
NHGFs-900	7.2	10.8	34.5
NHGFs-800	8.5	16.2	43.5

**Table S2.** The kinetic parameters of NHGFs, se-NGFs, ne-NGFs and NGFs.

## References

- 1 J. Duan, S. Chen, M. Jaroniec and S. Z. Qiao, *ACS Nano*, 2015, **9**, 931-940.
- 2 M. J. McAllister, J. L. Li, D. H. Adamson, H. C. Schniepp, A. A. Abdala, J. Liu, M. Herrera-Alonso, D. L. Milius, R. Car, R. K. Prud'homme and I. A. Aksay, *Chem. Mater.*, 2007, **19**, 4396-4404.