Supporting Information

Transformation of ZIF-8 Nanoparticles into 3D Nitrogen-Doped Hierarchically Porous Carbon for Li-S Batteries

Guiqiang Cao,[a] Da Bi,[a] Jingxiang Zhao,[b] Jing Zheng,[c] Zhikang Wang,[a] Qingxue Lai*,[a], and Yanyu Liang;*,[a]

[a]College of Materials Science and Technology, Nanjing University of Aeronautics and Astronautics, Nanjing 210016, P. R. China.

[b] College of Chemistry and Chemical Engineering, and Key Laboratory of Photonic and Electronic Bandgap Materials, Ministry of Education, Harbin Normal University, Harbin, 150025, China

[c] Department of Chemistry and Materials Science, College of Science, Nanjing Forestry University, Nanjing 210037, China.

*Corresponding authors: laiqingxue@126.com; liangyy403@126.com

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Models and Methods

All the calculations were performed by using the spin-polarized DFT methods implemented in the Vienna Ab Initio Simulation Package (VASP).[1] The exchange correlation energy was represented by the Perdew-Burke-Ernzerhof (PBE) functional within the generalized gradient approximation (GGA).[2] The electron-ion interactions were described by the projector augmented wave (PAW) method.[3] A Monkhorst-Pack k-point mesh of 5 × 5 ×1 was used to sample the Brillouin zone and the plane-wave cut-off energy was set to be 500 eV. To accurately determine the van der Waals (vdW) interactions in these layered systems, the DFT-D3 method with Grimme's scheme was employed.[4] All structures were fully relaxed until the forces on each atom were less than 0.02 eV $Å^{-1}$ and the convergence criterion for the electronic structure iteration was set to be 10^{-5} eV. The vacuum space was set to be 20 Å in the z direction, which was large enough to minimize the interaction between periodic images.

To model these N-doped graphenes, we used a supercell consisting of 5 × 5 graphene unit cells. The binding energy (E_b) of soluble Li_2S_6 species on the anchoring materials (AM) was determined by: $E_b = (E_{Li2S6} + E_{AM}) - E_{Li2S6\text{-}AM}$, where E_{Li2S6}, E_{AM}, and $E_{Li2S6\text{-}AM}$ represent the total electronic energies of isolated Li_2S_6 cluster, pristine AM, and the adsorbed Li_2S_6 systems, respectively. According to this definition, a more positive binding energy indicates a stronger binding strength of soluble Li_2S_n species on the anchoring materials.

Figure Captions in Supporting Information

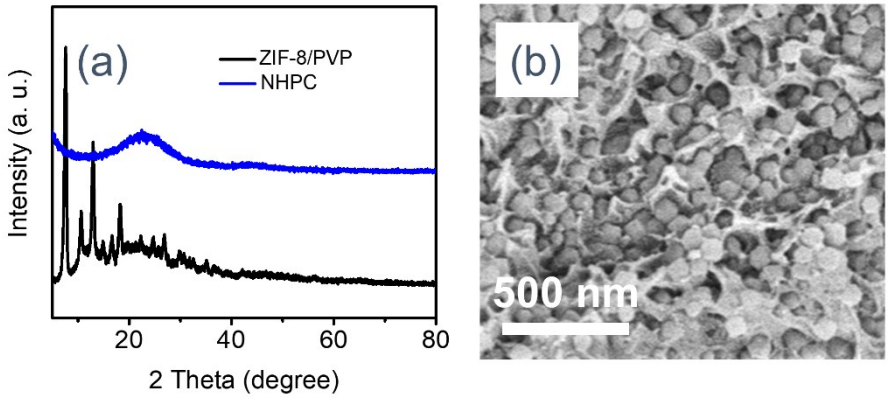

Figure S1. XRD pattern of ZIF-8/PVP and NHPC and SEM image of ZIF-8/PVP.

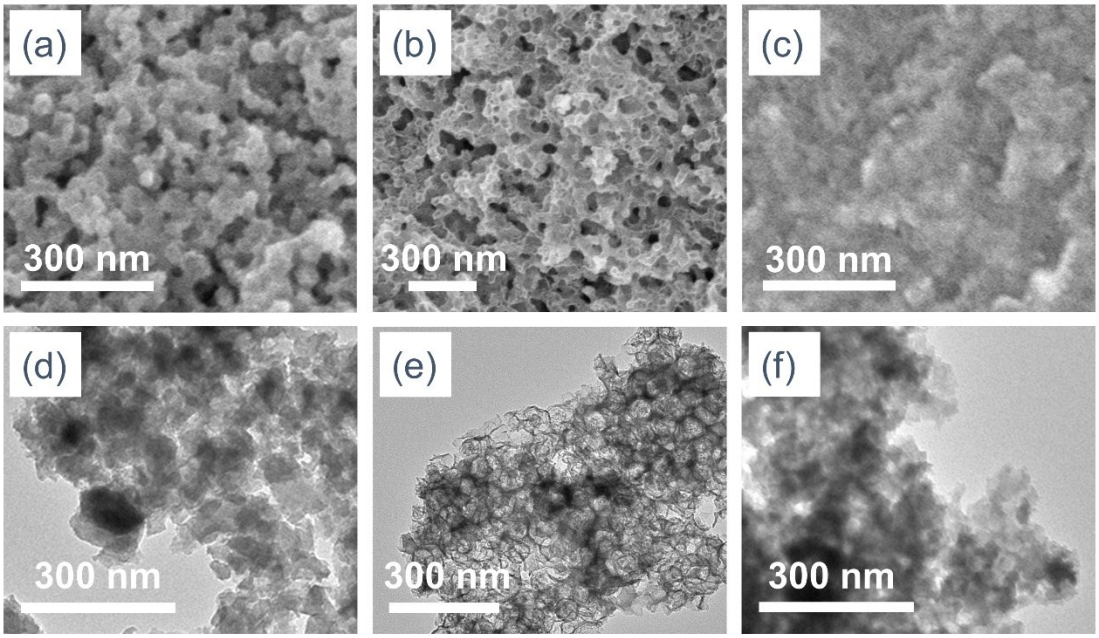

Figure S2. SEM image of NHPC with a ZIF-8/PVP ratio of a) 1:1, b) 1:2, c) 1:3 and corresponding TEM image d), e), f), respectively.

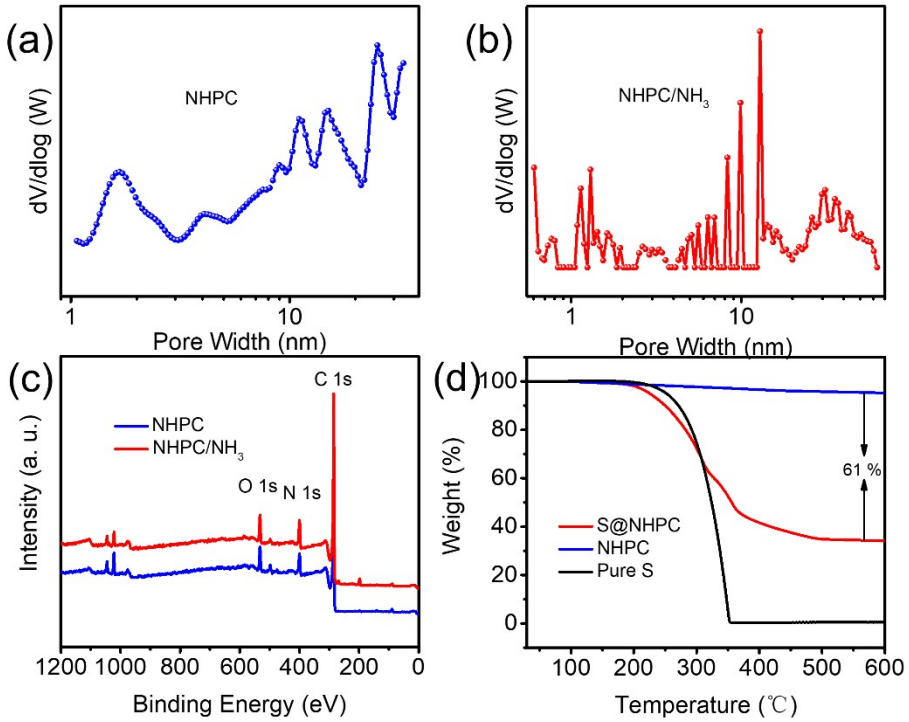

Figure S3. a-b) Pore distribution of NHPC and NHPC/NH_3, c) XPS study of NHPC and NHPC/NH_3, d) TG curves of S@NHPC and NHPC and Pure S.

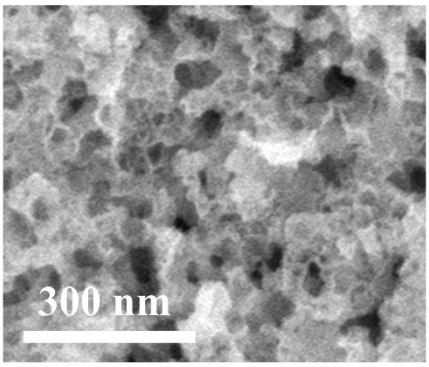

Figure S4. SEM image of S@NHPC/NH_3.

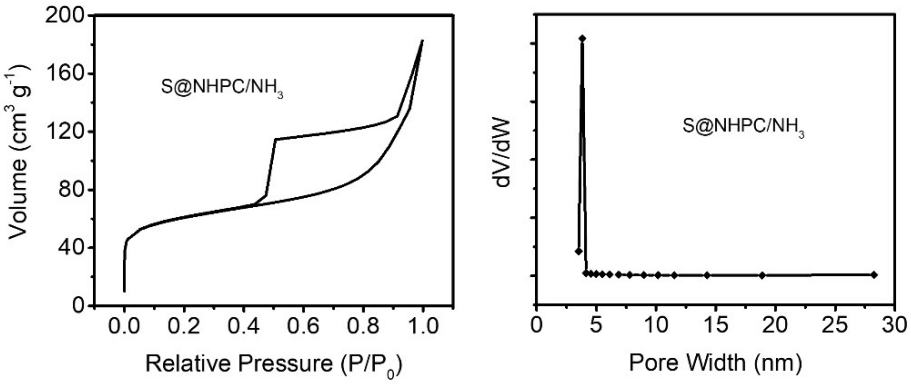

Figure S5. Nitrogen adsorption/desorption isotherm and pore size distributions of the S@NHPC/NH_3.

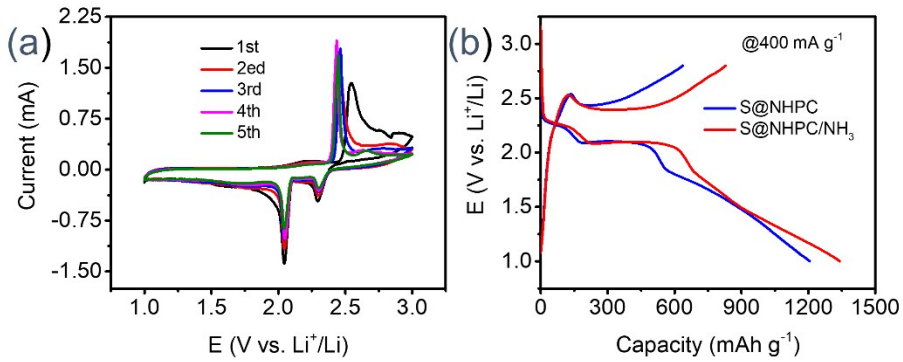

Figure S6. a) Cyclic voltammetry of the S@NHPC cathode. b) Charge/discharge profiles of the S@NHPC and the S@NHPC/NH_3 cathodes at 400 mA g^{-1}.

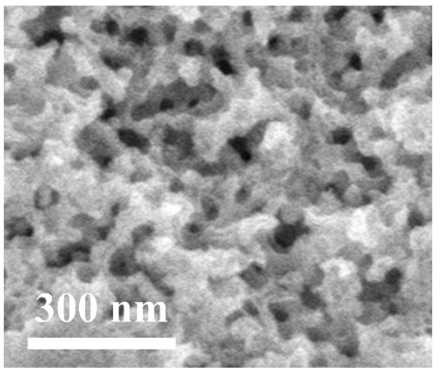

Figure S7. SEM image of S@NHPC.

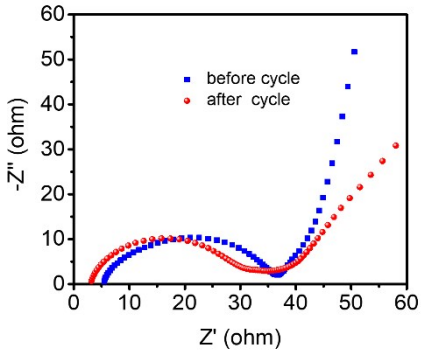

Figure S8. Electrochemical impedance spectra of S@NHPC/NH_3 cathodes before cycle and after cycle at 200 mA g^{-1}.

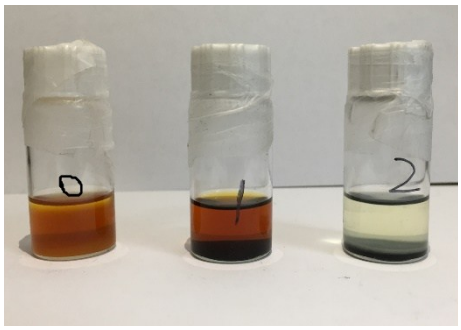

Figure S9. The image of a Li_2S_6 solution before (0) and after the addition of NHPC (1) and NHPC/NH_3 (2) for 3 h.

References in Supporting Information

[1] G. Kresse and J. Furthmüller, Physical Review B, 1996, 54, 11169-11186.

[2] J. P. Perdew, K. Burke and M. Ernzerhof, Physical Review Letters, 1996, 77, 3865-3868.

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[4] S. Grimme, J. Antony, S. Ehrlich and H. Krieg, Journal of Chemical Physics, 2010, 132, 154104.