Electronic Supplementary Material (ESI) for Sustainable Energy & Fuels.

Supporting Information

Nitrogen and fluorine co-doped holey graphene hydrogel as the binder-free electrode material for flexible solid-state supercapacitors

Yulin Chen,[a] Yu Li,*[acd] Fengnan Yao,[a] Cong Peng,[a] Chen Cao,[a] Yiyu Feng,[acd] Wei Feng*[abcd]

a. School of Materials Science and Engineering, Tianjin University, Tianjin 300072, P. R China. E-mail: weifeng@tju.edu.cn; Tel: +86-22-28578269; Fax: +86-22-27404724

b. Collaborative Innovation Center of Chemical Science and Engineering (Tianjin), Tianjin 300072, P. R China

c. Key Laboratory of Advanced Ceramics and Machining Technology, Ministry of Education, Tianjin 300072, P. R China

d. Tianjin Key Laboratory of Composite and Functional Materials, Tianjin 300072, P. R China

Corresponding Author

**E-mail: weifeng@tju.edu.cn Tel:+86-22-87402059. Fax: +86-22-27404724.*

Table S1. A summary of atomic percentages of C, O, N and F

Sample	C(at%)	O(at%)	N(at%)	F(at%)
GO	71.14	28.86	0	0
RGH	87.32	12.68	0	0
HGH	85.81	14.19	0	0
NF-GH	85.82	11.79	1.87	0.52
NF-HGH	83.39	12.91	2.56	1.14

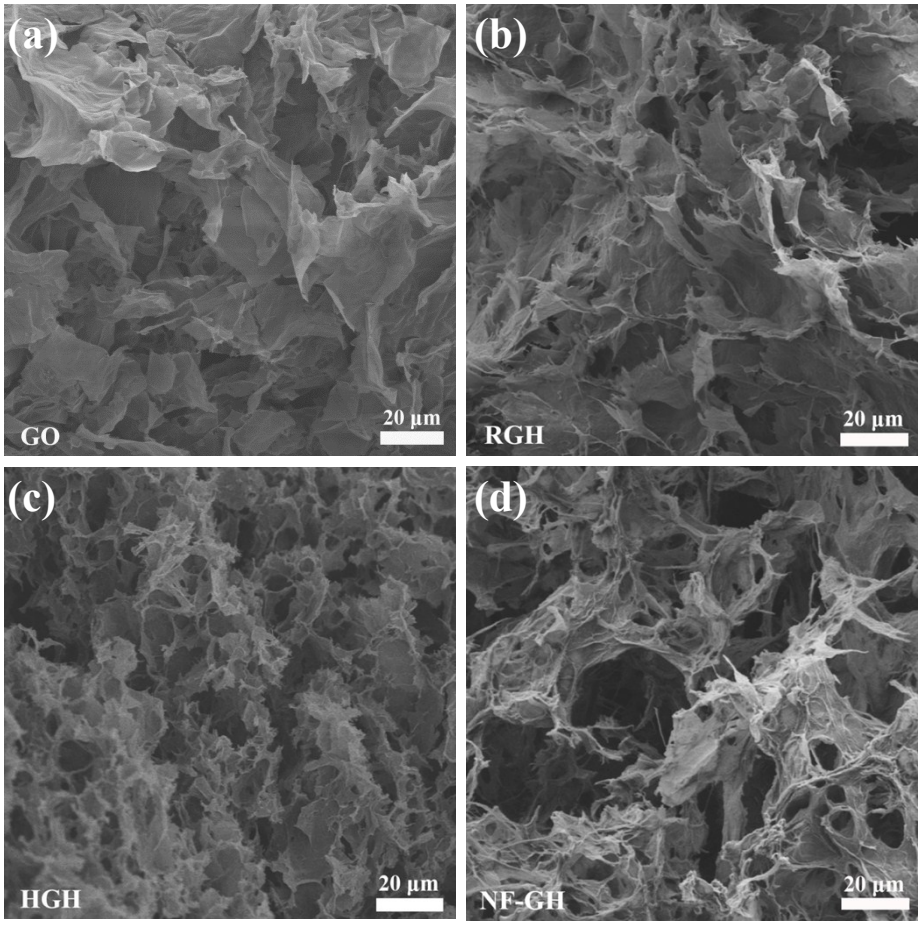

Fig. S1 SEM images of a) GO, b) RGH, c) HGH and d) NF-GH.

Fig. S2 Optical photographs of the synthesized hydrogels, from left to right, RGH, HGH, NF-GH and NF-HGH.

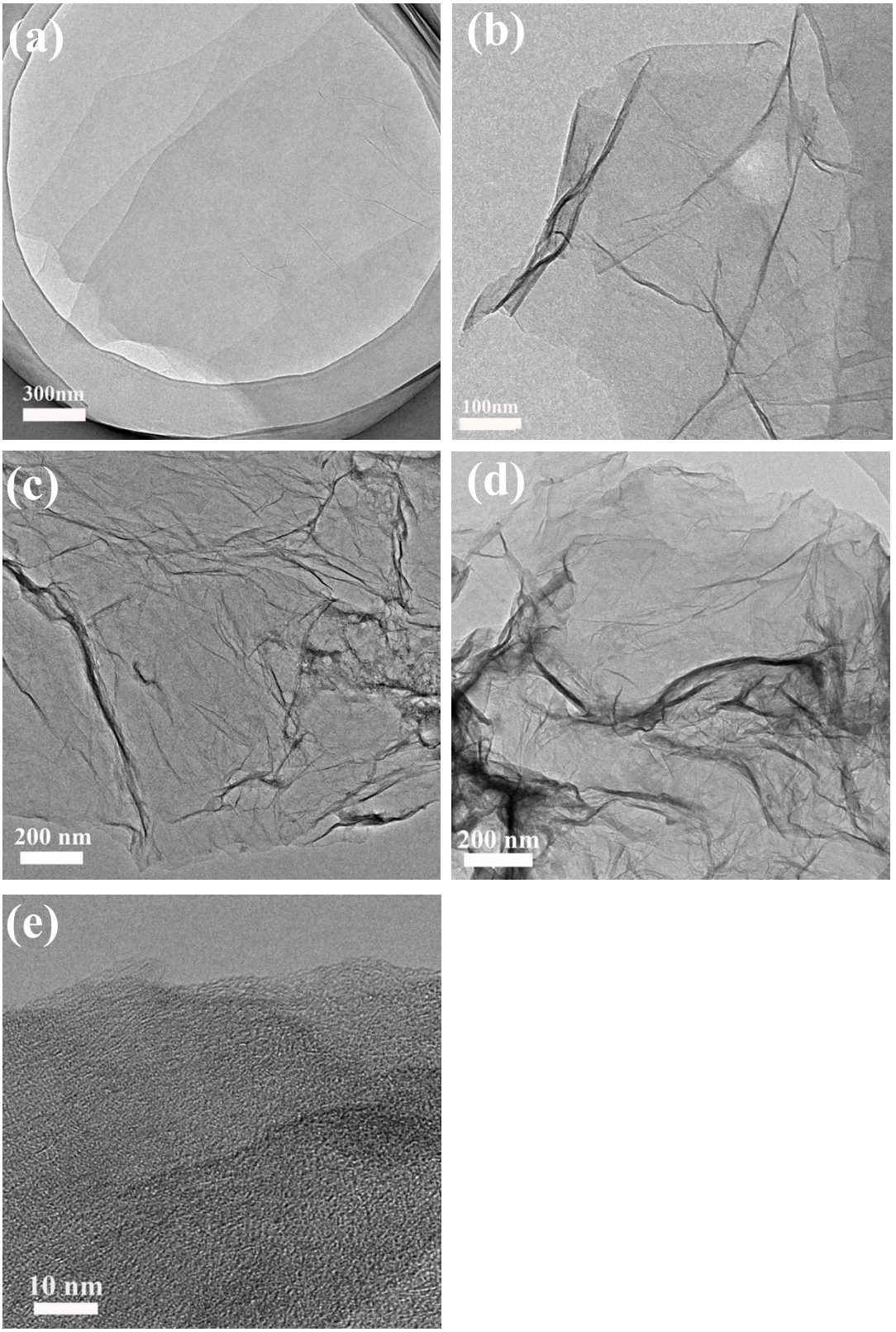

Fig. S3 TEM images of a) GO, b) RGH, c) HGH d) NF-GH and e) HRTEM images of NH-GH.

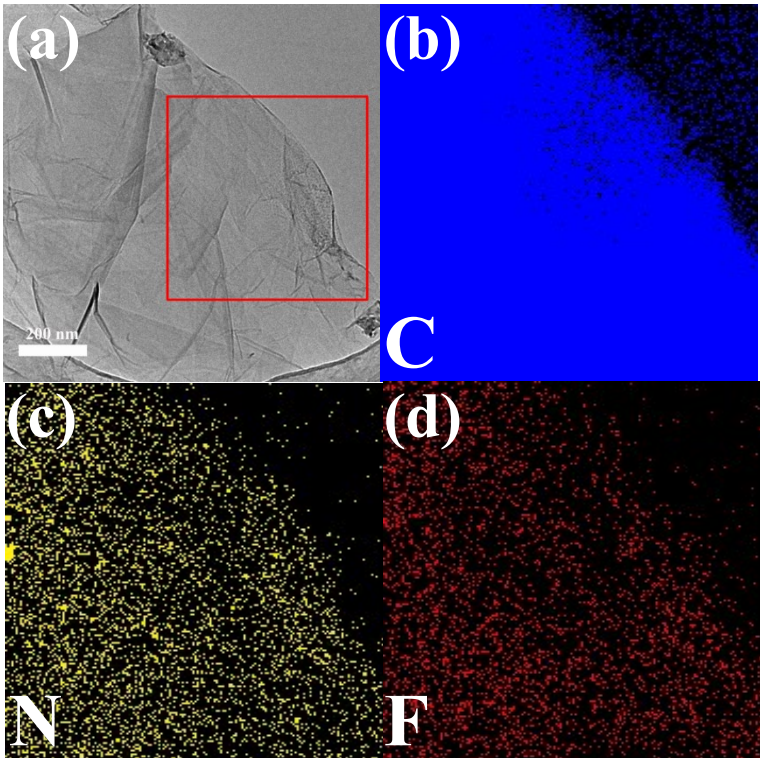

Fig. S4 a) The representive TEM image of NF-GH and the corresponding elemental mappings of b) C, c) N and d) F in the framed area.

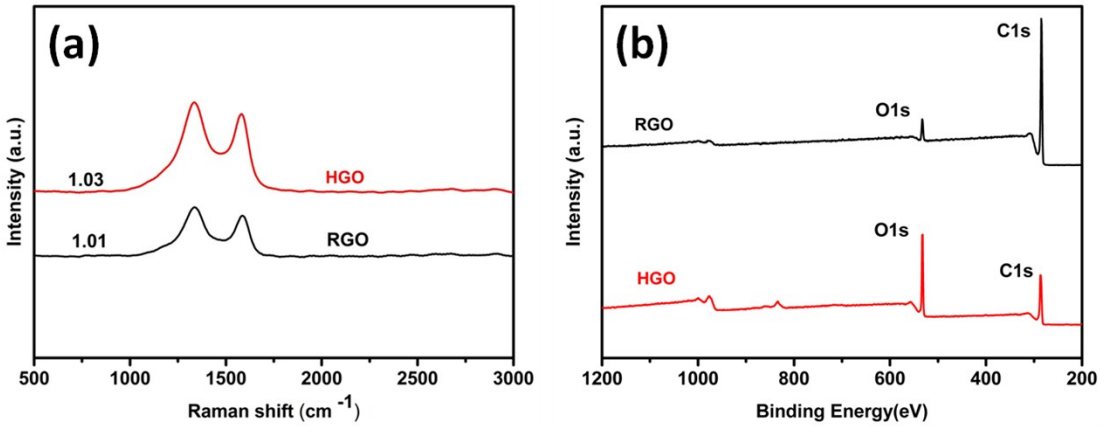

Fig. S5 Raman a) and XPS survey spectra b) of prepared precursors with and without H_2O_2 activation.

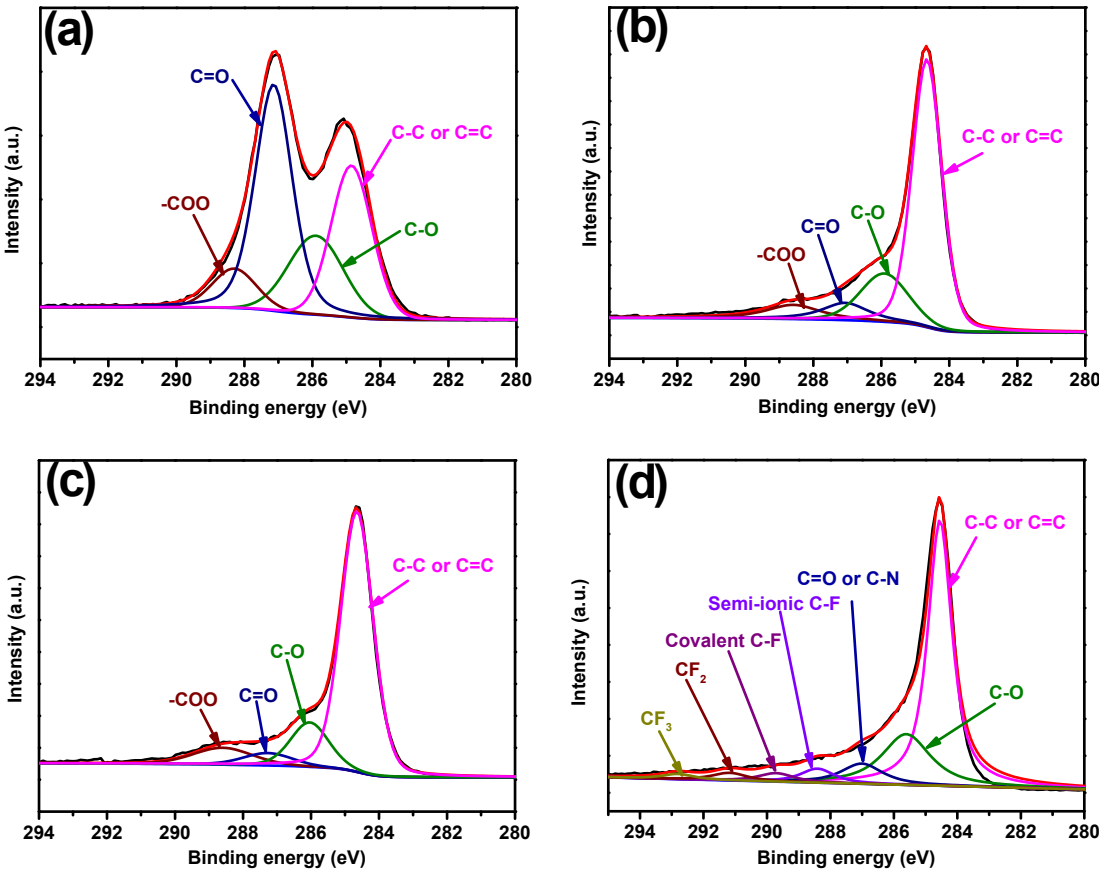

Fig. S6 High resolution C1s spectra of a) GO, b) RGH, c) HGH, and d) NF-GH.

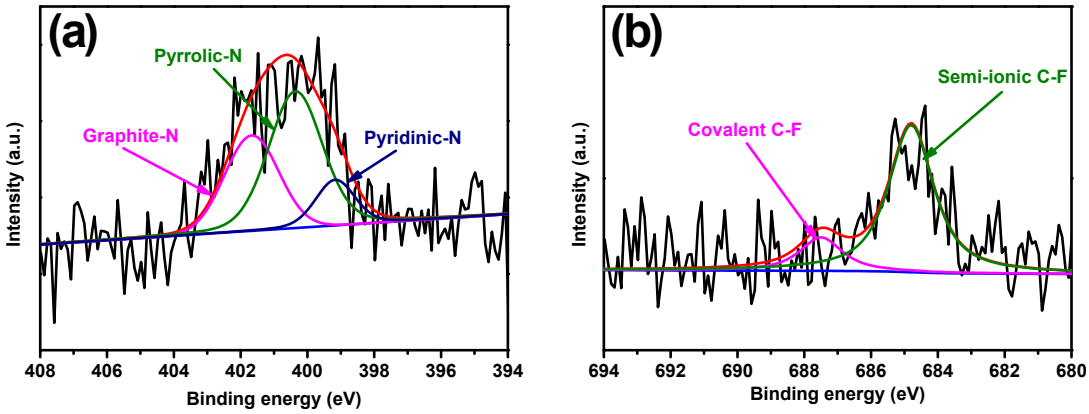

Fig. S7 High-resolution a) N 1s and b) F 1s spectra of NF-GH.

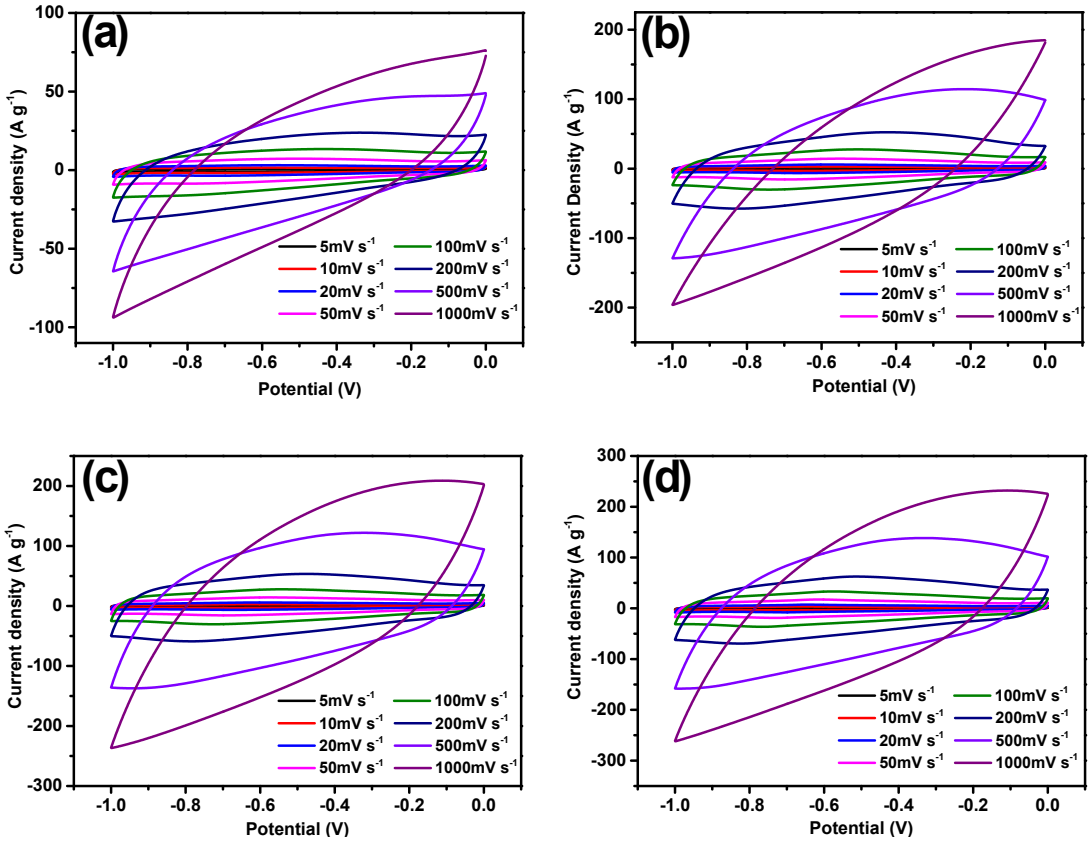

Fig. S8 CV curves of a) RGH, b) HGH, c) NF-GH, and d) NF-HGH measured at different scan rates from 5 to 1000 mV s^{-1} in the potential window from -1.0 to 0 V.

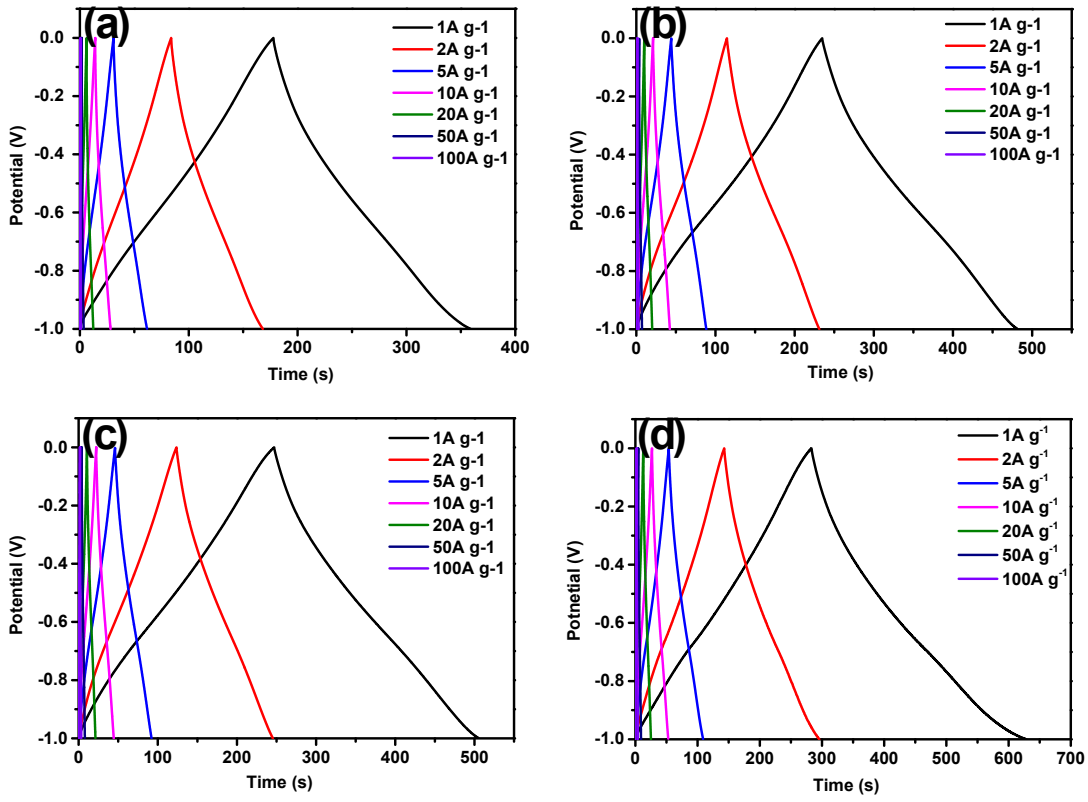

Fig. S9 Charge-discharge profiles of a) RGH, b) HGH, c) NF-GH, and d) NF-HGH at different current densities from 1 to 100 A g^{-1} in the potential range of -1.0 to 0 V.

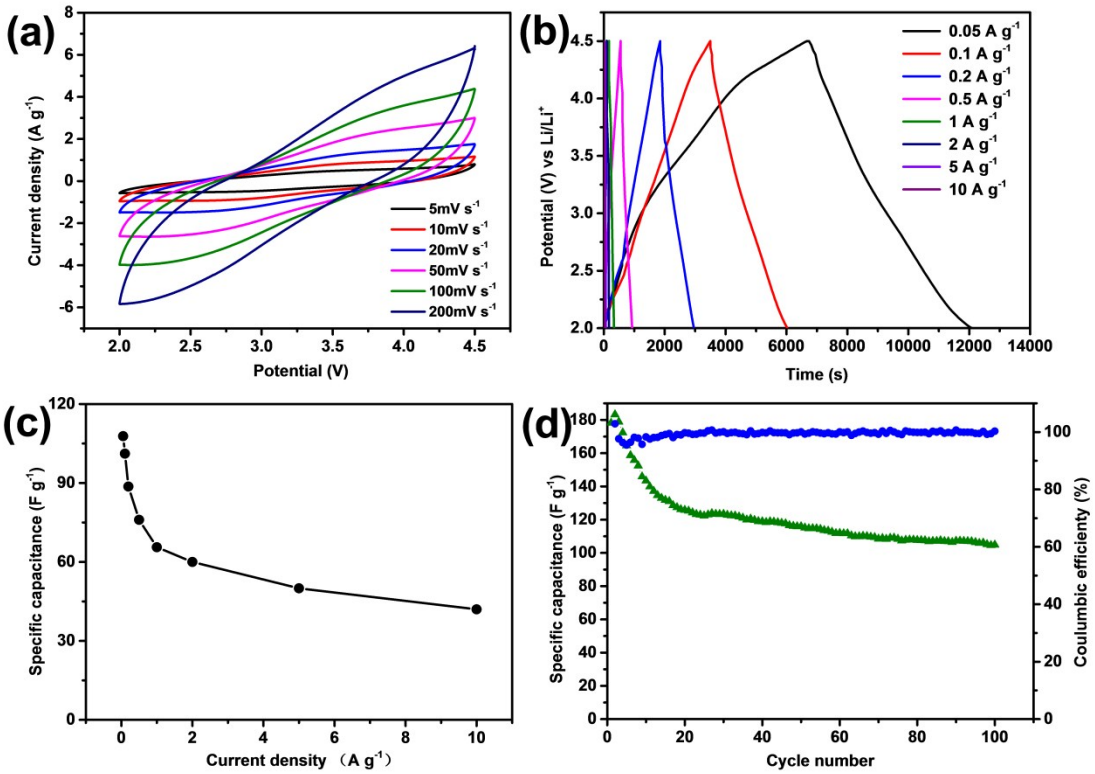

Fig. S10 Electrochemical performances NF-HGH in the potential window from 2.0 to 4.5 V in 1 M LiPF6/DC/EMC organic electrolyte: a) CV curves at the scan rate of 5, 10, 20, 50, 100, 200 mV s-1; b) The galvanostatic charge-discharge profiles at a current density of 0.05, 0.1, 0.2, 0.5, 1, 2, 5, 10 A g-1; c) Specific capacitances at different current densities; d) cycle curves of NF-HGH with 100 cycles.

The performance of NF-HGH in organic electrolyte is tested by the two-electrode system.. The work electrodes were synthesized by pressing the wet NF-HGH with the same dry weight into round nickel foams with a pressure of 10 Mpa for 30 S. The diameter of the nickel foam is 15 mm which is suitable for the CR2032-type coin cell. Then the electrodes were immersed in 1 M $LiPF_6$/DC/EMC organic electrolyte for 6 h. After that, the working electrodes and a glassy fibrous separator with organic electrolyte of 1 M $LiPF_6$/DC/EMC were assembled the cell to test. Cyclic voltammetry (CV) and galvanostatic charge/discharge (GCD) were conducted with a Zahner Zennium electrochemical workstation at room temperature. The potential range for CV and GCD testing was 2.0 to 4.5 V.

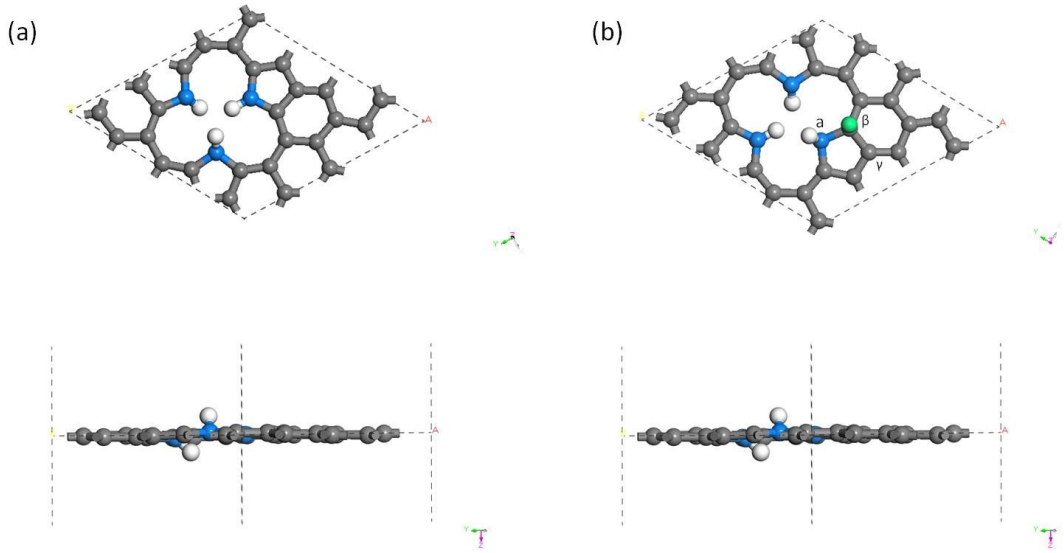

Fig. S11 schematic structural diagram and layer spacing of a) pyridine nitrogen-doped graphene and b) fluorine-doped pyridine nitrogen-doped graphene.