Electronic Supporting Information

Significantly boosting energy storage capacity of N-doped graphene by non-covalent modification of fused heterocyclic small molecules

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Preparation of working electrode

The working electrodes were prepared as follows: the active materials and the binder (SBR) (mass ratio = 39:1) were ultrasonically dispersed in ethanol aqueous solution to form a mixed slurry, and then the slurry was uniformly coated on the treated carbon cloth (CC, Diameter: 6mm). Finally, the working electrode was dried in an oven at 60 °C for 12 h. The loading amount of the active material on the carbon cloth was about 2.5 mg/cm^2.

Electrochemical calculation

For three-electrode system, the specific capacitance values (C_{S1}) of the material were calculated from CV curves according to the following formula [1]:

$$C_{S1} = \frac{\int_{E_1}^{E_2} i(E)d(E)}{2v(E_2 - E_1)}$$

Where "E_1" and "E_2" are the cutoff potentials in CV, "i(E)" is the instantaneous current, "i(E)d(E)" is the total voltammetric charge obtained by integration of the positive and negative sweeps in the CV, "v" is the scan rate, and "m" is the mass of the individual sample.

The specific capacitance (C_{S2}, F g^{-1}) of the sample was calculated from GCD curves according to the following formula[1]:

$$C_{S2} = \frac{I * \Delta t}{\Delta V * m}$$

where "C" is the specific capacitance (F g^{-1}), "I" is the charge/discharge current (A), and "Δt" is the discharge time (s), "ΔV" is the voltage window (V), and "m" is the mass

(g) of the active material.

The discharge capacity (C_{S3}, mAh g^{-1}) was calculated by the following formula[2]:

$$C_{S3} = \frac{I * \Delta t}{3.6 * m}$$

where I, Δt, and m are the discharge current (A), the discharge time (s), and the total mass (g) of the active material of the two electrodes, respectively.

For the two-electrode system, the specific capacitance (C_{S3}, F g^{-1}) was calculated by the following formula[3]:

$$C_{S3} = \frac{4 * I * \Delta t}{\Delta V * m}$$

where I, ΔV, Δt, and m are the discharge current (A), the voltage window (V), the discharge time (s), and the total mass (g) of the active material of the two electrodes, respectively.

The discharge capacity (C_{S4}, mAh g^{-1}) was calculated by the following formula:

$$C_{S4} = \frac{4 * I * \Delta t}{3.6 * m}$$

where I, Δt, and m are the discharge current (A), the discharge time (s), and the total mass (g) of the active material of the two electrodes, respectively.

The energy density (Wh Kg^{-1}) and power density (W Kg^{-1}) were calculated by the following two equations[4]:

$$E = \frac{C_{S3} * \Delta V^2}{8 * 3.6}$$

$$P = \frac{3600 * E}{\Delta t}$$

where C_{S2}, Δt, and ΔV are specific capacitances (F g^{-1}), discharge time (s) and voltage window (V).

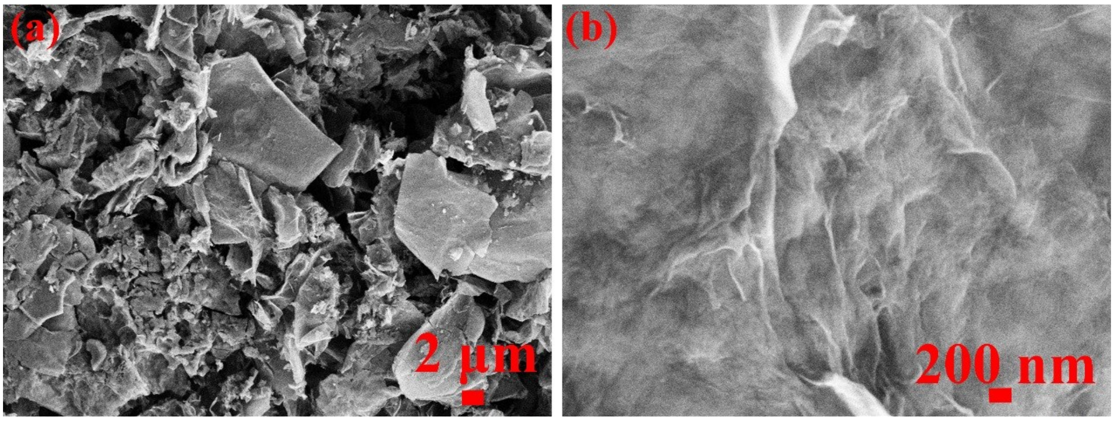

Fig. S1 SEM images of GN.

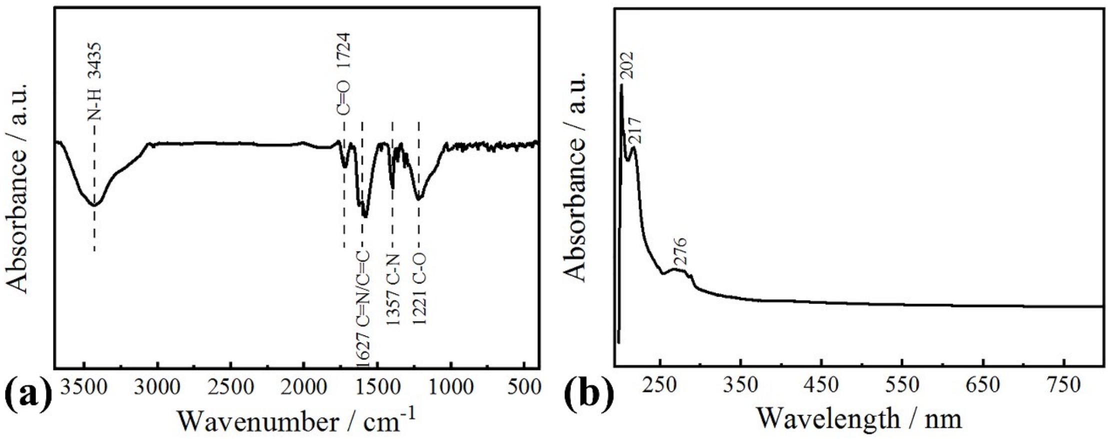

Fig. S2 FT-IR spectrum (a) and UV−vis absorption spectrum (b) of GN.

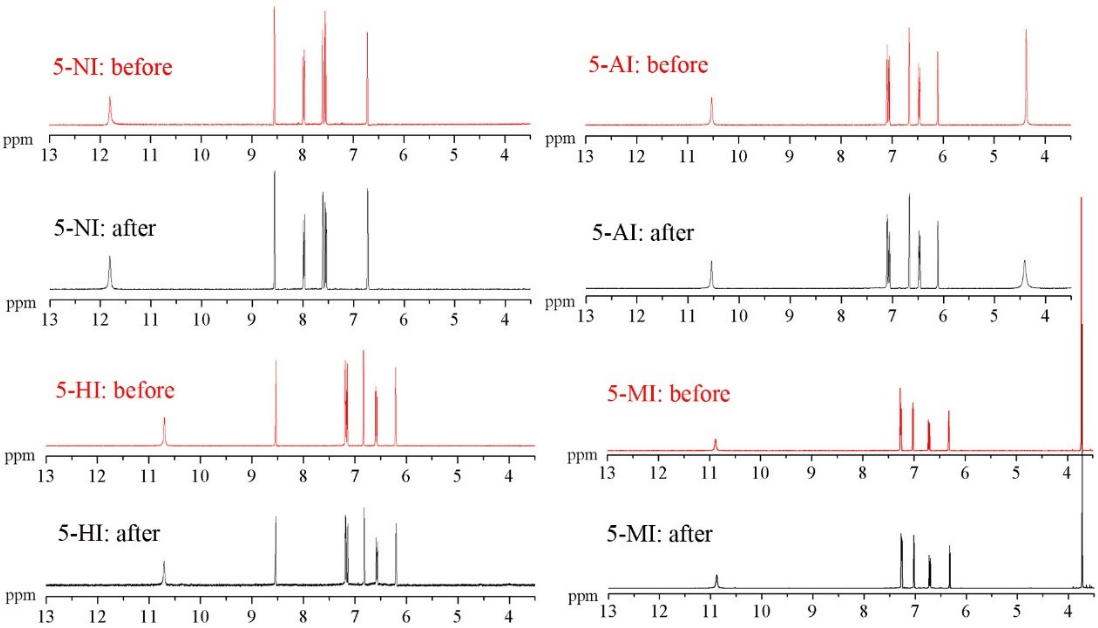

Fig. S3 ^1H NMR spectra of indole monomers before and after the hydrothermal reaction, CD_3SOCD_3 as the solvent.

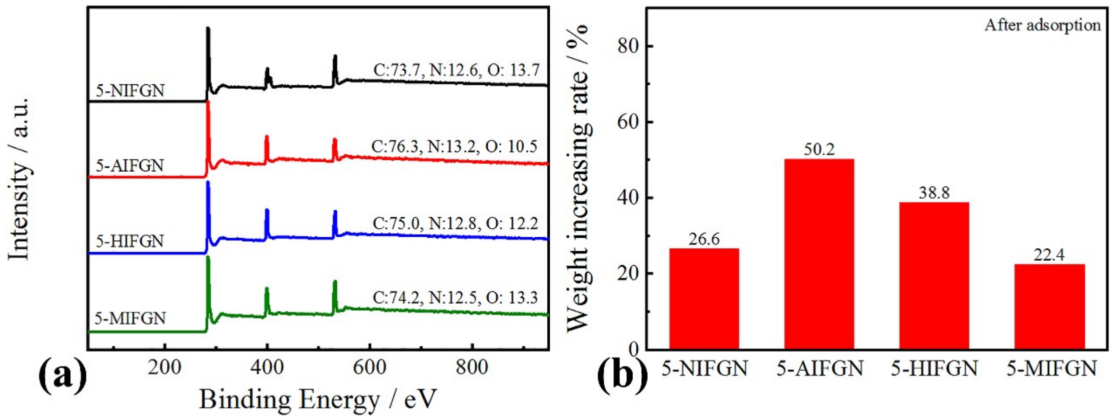

Fig. S4 XPS survey spectrum (a) and statistic of the weight increasing rates (b) of 5-NIFGN, 5-AIFGN, 5-HIFGN and 5-MIFGN.

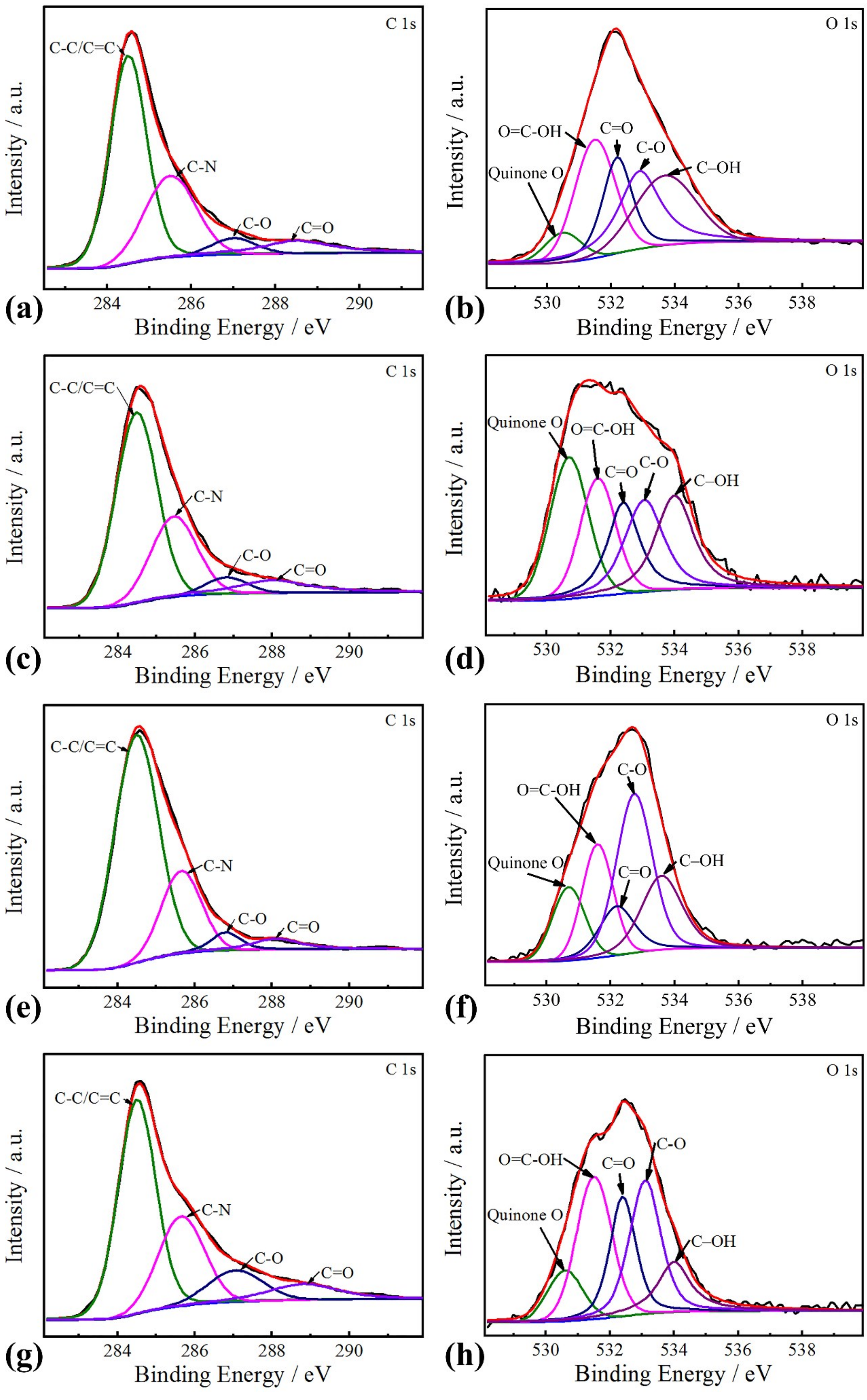

Fig. S5 XPS deconvolution of C 1s and O 1s of 5-NIFGN (a,b), 5-AIFGN (c,d), 5-

HIFGN (e,f) and 5-MIFGN (g,h).

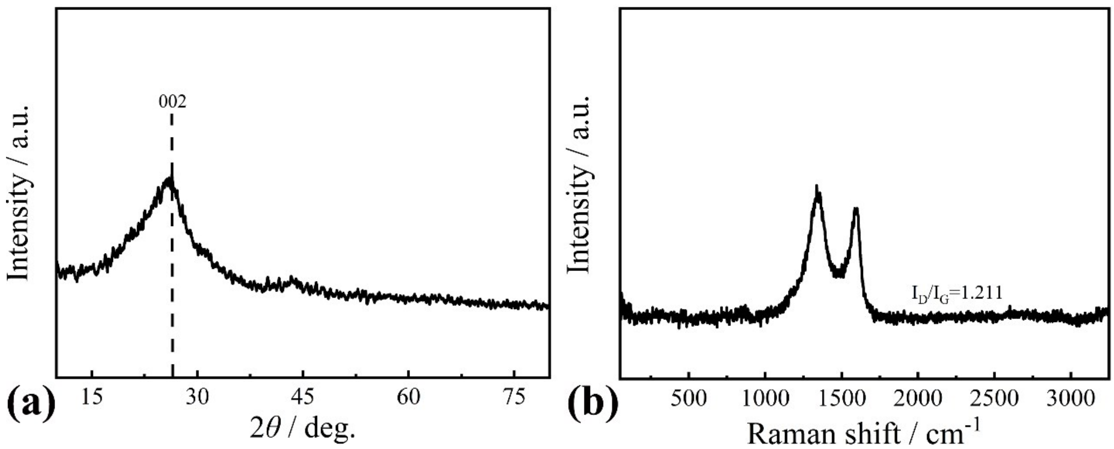

Fig. S6 XRD (a) and Raman spectrum (b) of GN.

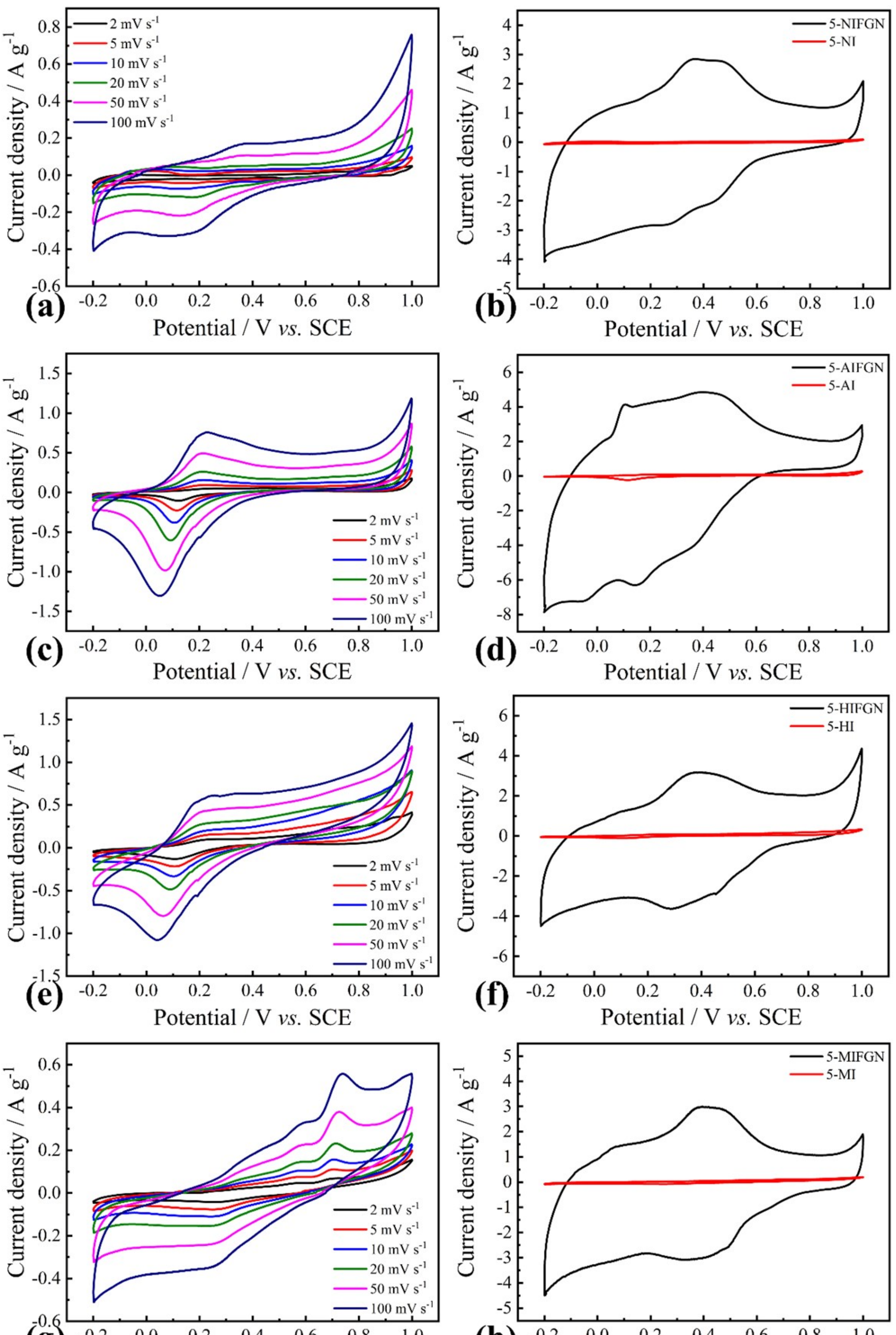

Fig. S7 Electrochemical response of pure organic molecules and comparison with the corresponding functionalized GN: (a) 5-NI, (b) 5-NI and 5-NIFGN, (c) 5-AI, (d) 5-AI and 5-AIFGN, (e) 5-HI, (f) 5-HI and 5-HIFGN, (g) 5-MI, (h) 5-MI and 5-MIFGN.

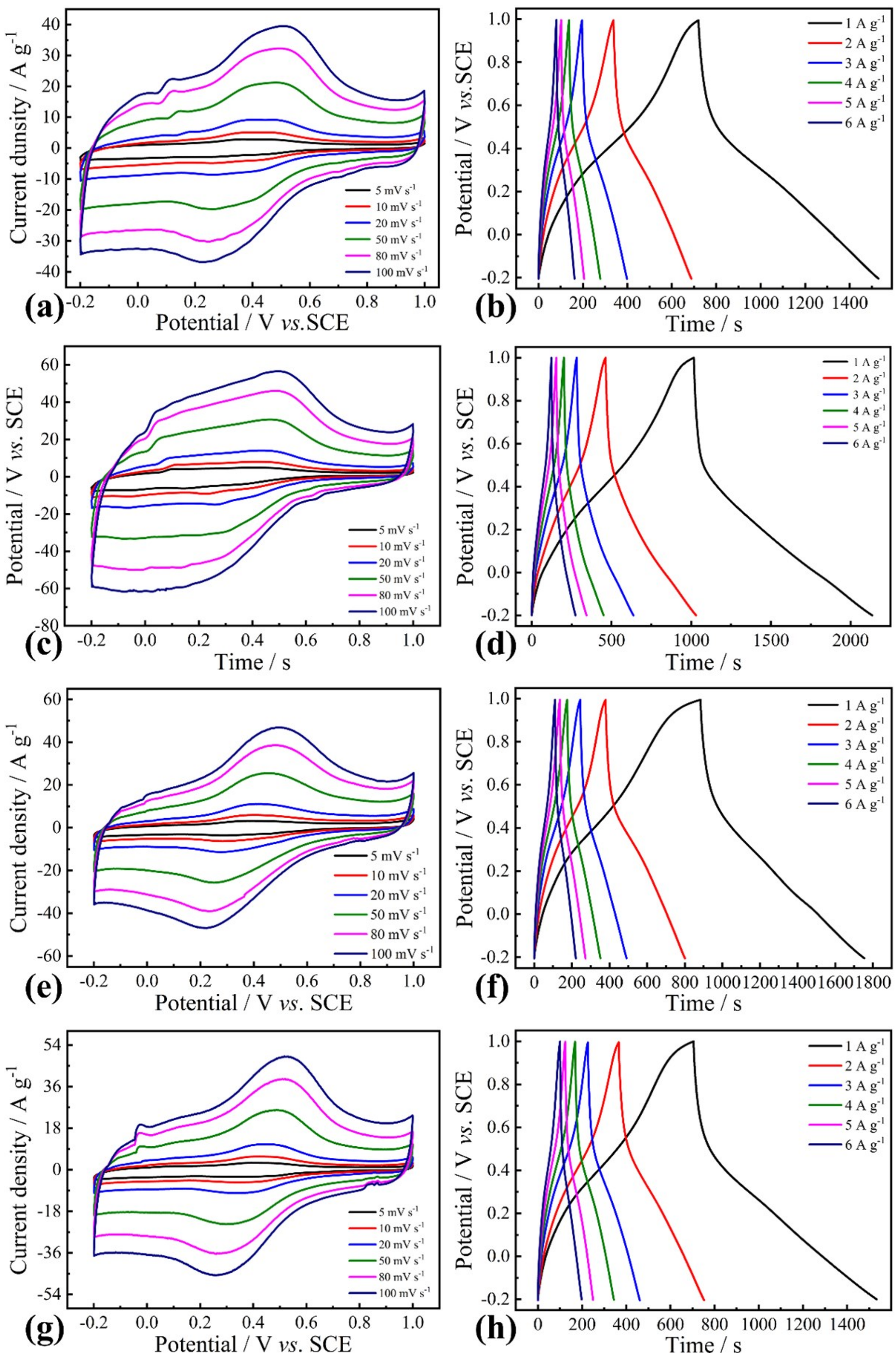

Fig. S8 Electrochemical performance of 5-NIFGN, 5-AIFGN, 5-HIFGN and 5-MIFGN under the three-electrode system: CV curves at the scan rate of 5-100 mV s^{-1} (a, c, e, g), GCD curves at the current density of 1-6 A g^{-1} (b, d, f, h),

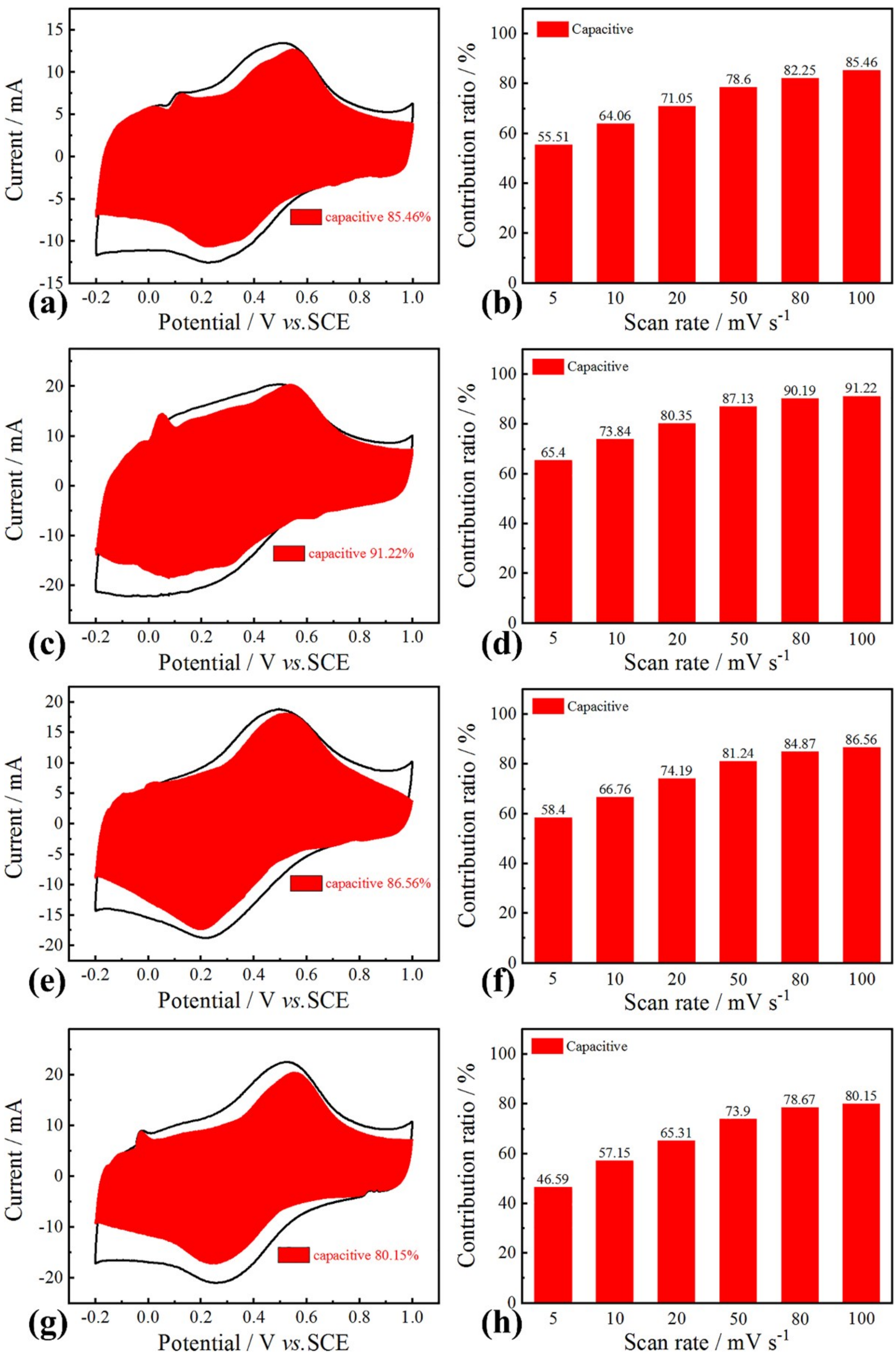

Fig. S9 The contribution of pseudo-capacitance at 100 mV s^{-1} and pseudo-capacitance contribution rates at 5-100 mV s^{-1} of 5-NIFGN (a, b), 5-AIFGN (c, d), 5-HIFGN (e, f) and 5-MIFGN (g, h).

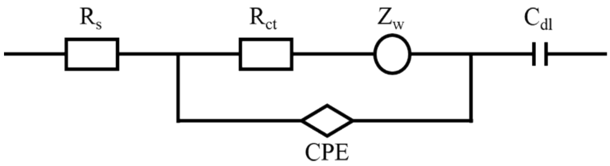

Fig. S10 Equivalent circuit diagram of GN, 5-NIFGN, 5-AIFGN, 5-HIFGN and 5-MIFGN.

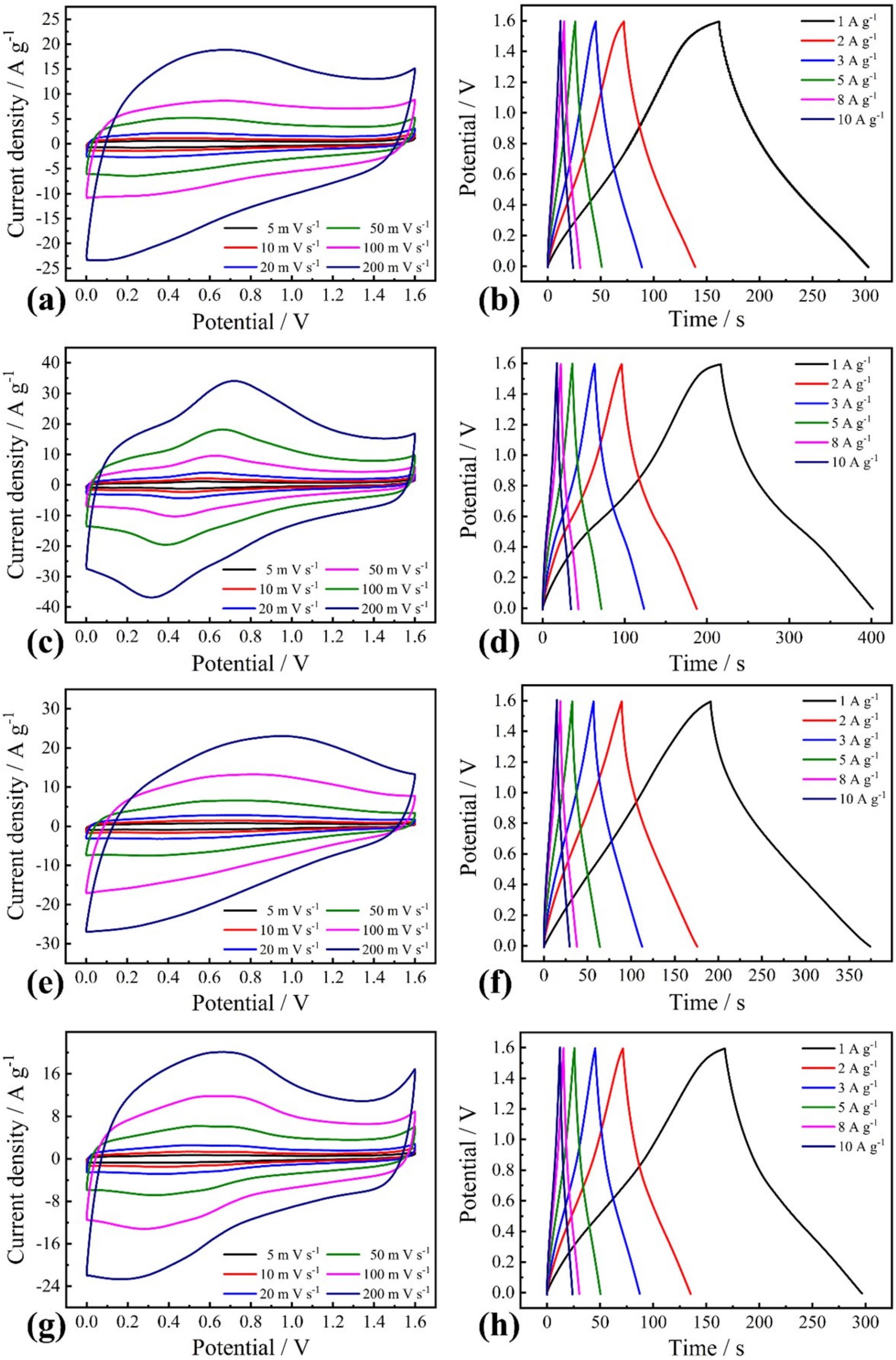

Fig. S11 Electrochemical performance of 5-NIFGN, 5-AIFGN, 5-HIFGN and 5-MIFGN under the two-electrode system: CV curves at the scan rate of 5-200 mV s^{-1} (a, c, e, g), GCD curves at the current density of 1-10 A g^{-1} (b, d, f, h).

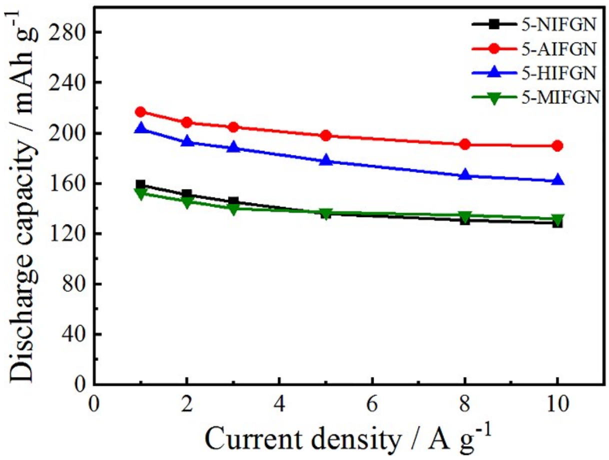

Fig. S12 Plots of discharge capacity versus current densities of 5-NIFGN, 5-AIFGN, 5-HIFGN and 5-MIFGN.

Supporting References

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3. Y.B. Zou, W.B. Zhong, S.C. Li, J.W. Luo, C.L. Xiong, W.T. Yang, Structure of functionalized nitrogen-doped graphene hydrogels derived from isomers of phenylenediamine and graphene oxide based on their high electrochemical performance, *Electrochim. Acta*, 2016, **212**, 828-838.

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