Electronic Supplementary Material (ESI) for Journal of Materials Chemistry A.

Supplementary Information

Experimental Section

***Preparation of N-doped carbon nanotubes on graphene framework (N-CNTs–GF)*:** For preparation of N-CNTs–GF, 5 g melamine and Fe_2O_3/GF (200 mg) were mixed together in a ceramic boat and treated under the same conditions as the preparation of nitrogen-doped CNTs–holey graphene framework (N-CNTs–HGF). The N-CNTs–GF was obtained after removing the iron particles with 12 M HCl and washing the residue with water until the pH is equal to 7.0.

***Preparation of N-doped holey graphene framework (N-HGF)*:** For preparation of N-HGF, the as-prepared Fe_2O_3/HGF with porous structures was immersed into concentrated HCl solution (12 M) to remove the residual iron species after annealing at 850 ^oC for 2 h with a heating speed of 5 ^oC min^{-1} under Ar atmosphere. After washing and freeze-drying process, the prepared HGF was heated with melamine under the same conditions as previous preparation of N-CNTs–HGF. The N-HGF was obtained after the furnace cooling down to room temperature.

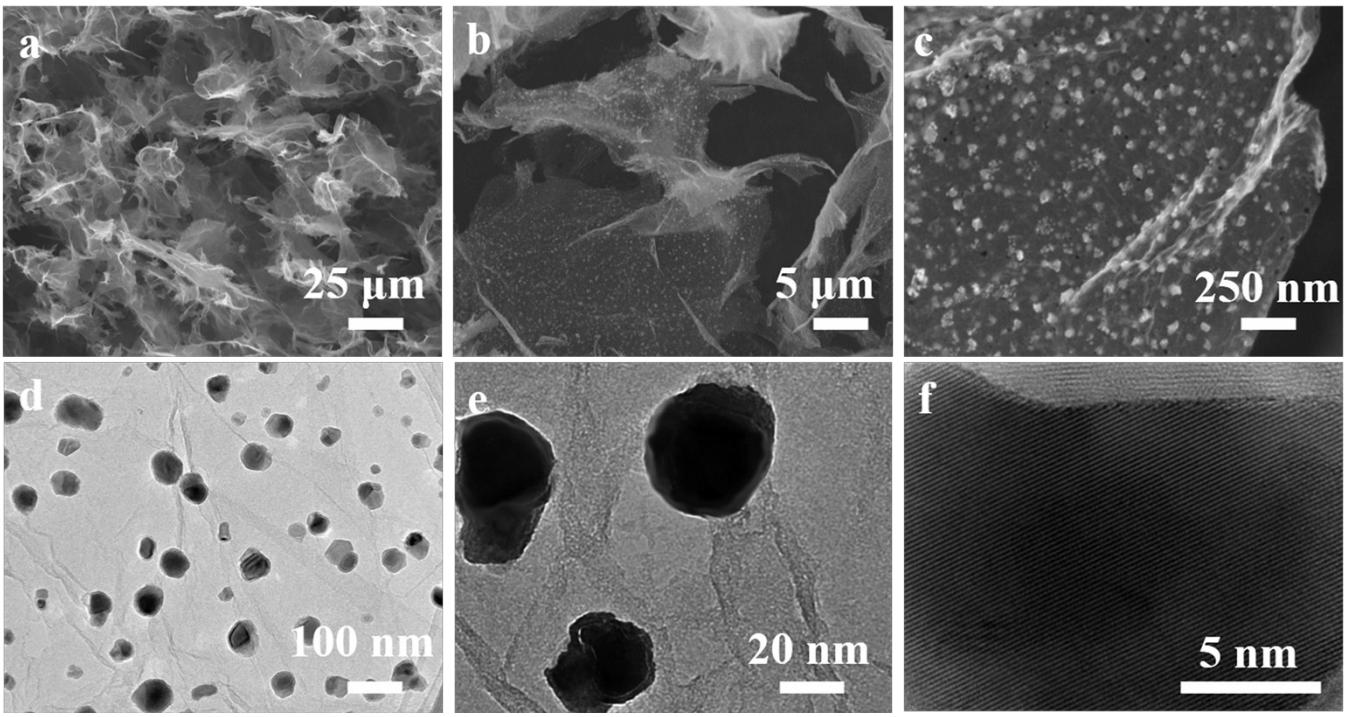

Figure S1. The SEM and TEM images of the as-prepared holey graphene framework decorated with well-distributed Fe_2O_3 nanoparticles at different scale bars: a) 25 μm; b) 5 μm; c) 250 nm; d) 100 nm; e) 20 nm; f) 5 nm.

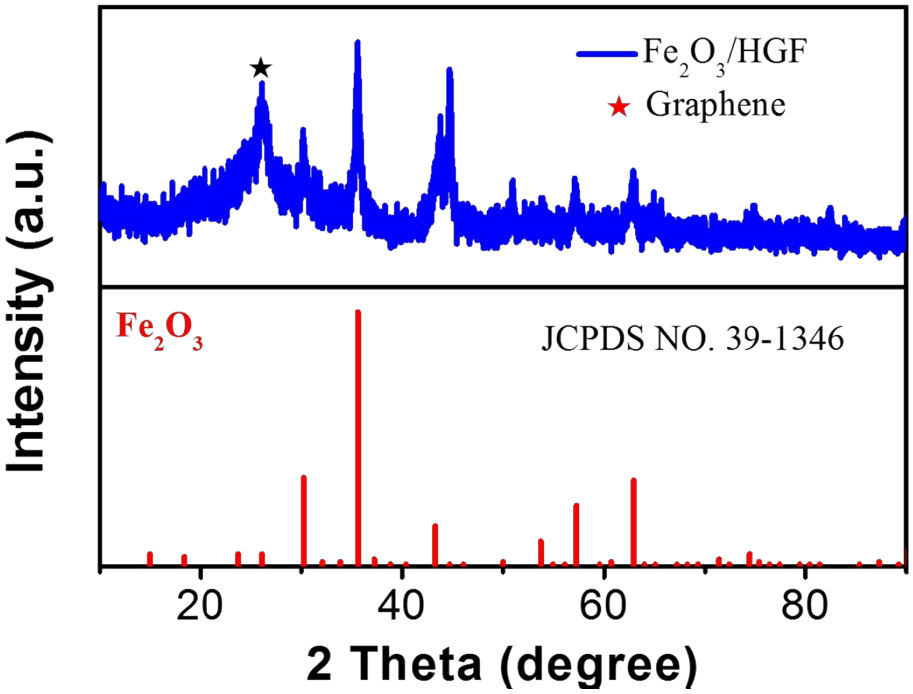

Figure S2. The XRD data of the Fe_2O_3/HGF and the corresponding standard Fe_2O_3 XRD pattern (JCPDS No. 39-1346).

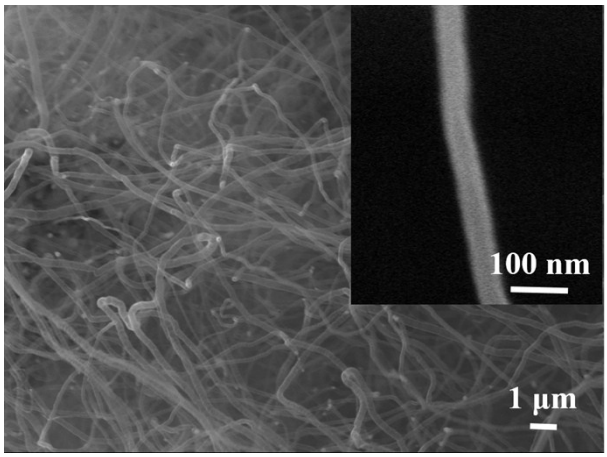

Figure S3. The SEM image of crooked bamboo-like CNTs, insert: the high-resolution image of a single CNT.

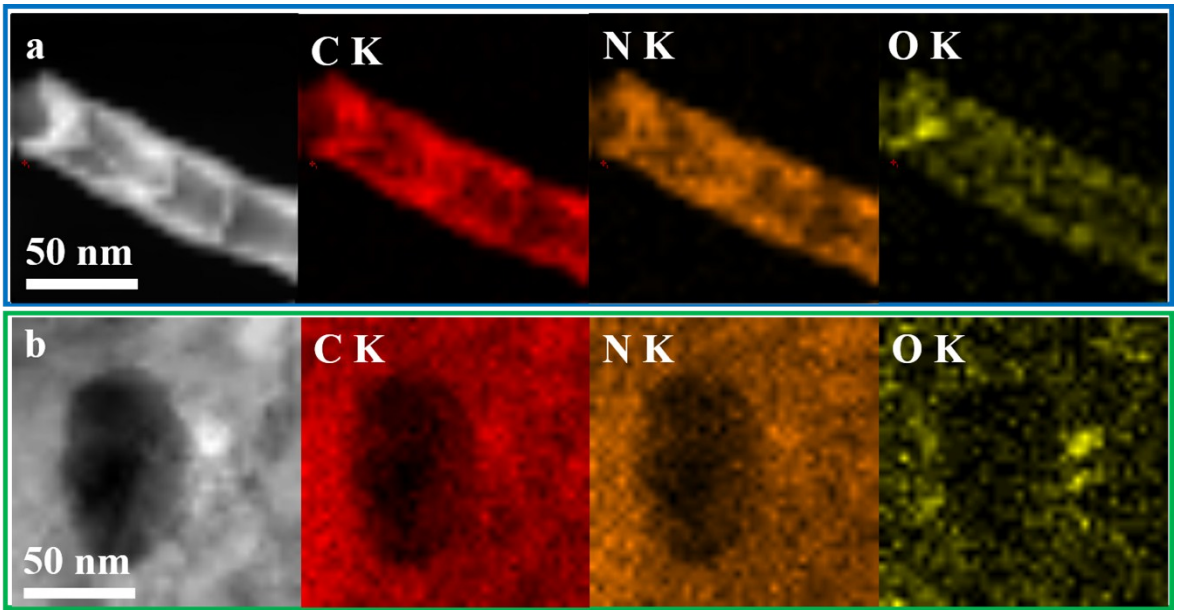

Figure S4. The STEM images of a) N-CNT and b) graphene nanohole with carbon (C), nitrogen (N) and oxygen (O) element mappings.

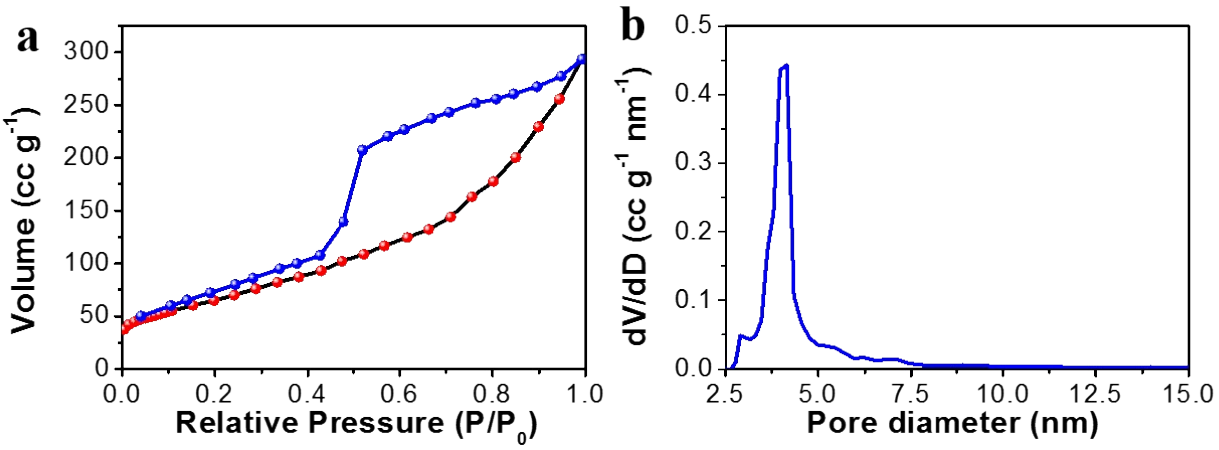

Figure S5. a) The nitrogen adsorption-desorption isotherms of the N-HGF; b) The corresponding pore size distribution obtained by using Density-Functional-Theory (DFT) method.

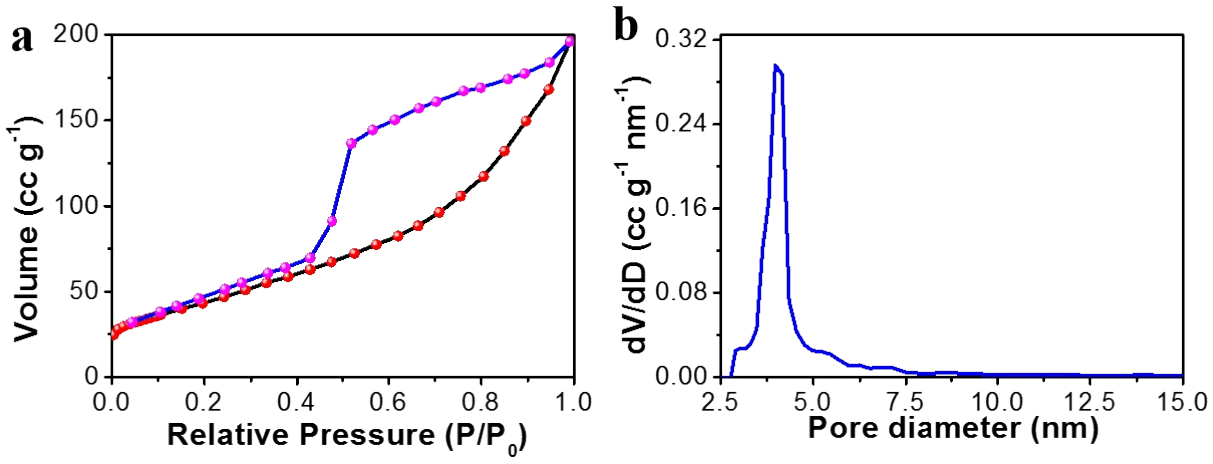

Figure S6. a) The nitrogen adsorption-desorption isotherms of the N-CNTs–GF; b) The corresponding pore size distribution obtained by using DFT method.

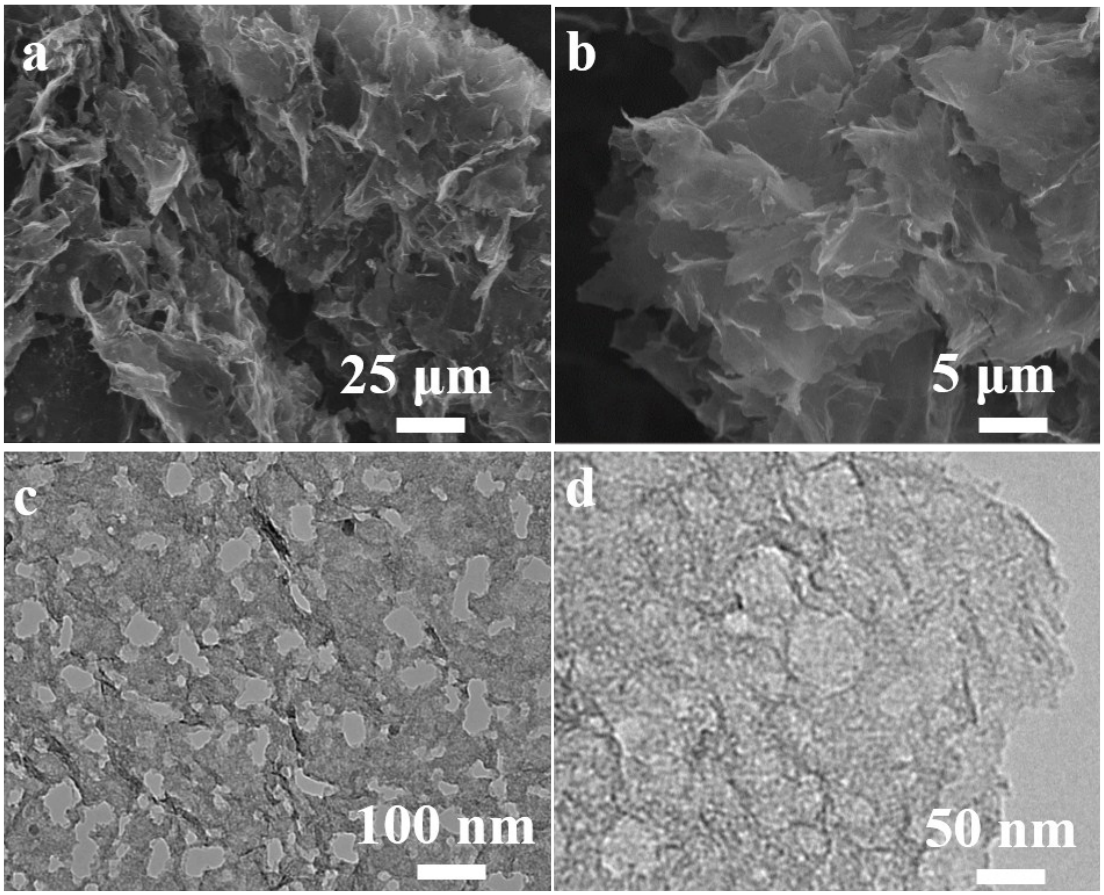

Figure S7. The morphological images of as-synthesized N-HGF. a, b) SEM images of the N-HGF; c) TEM image of N-HGF with obvious holes on graphene sheets; d) The enlarged image of holey graphene sheets.

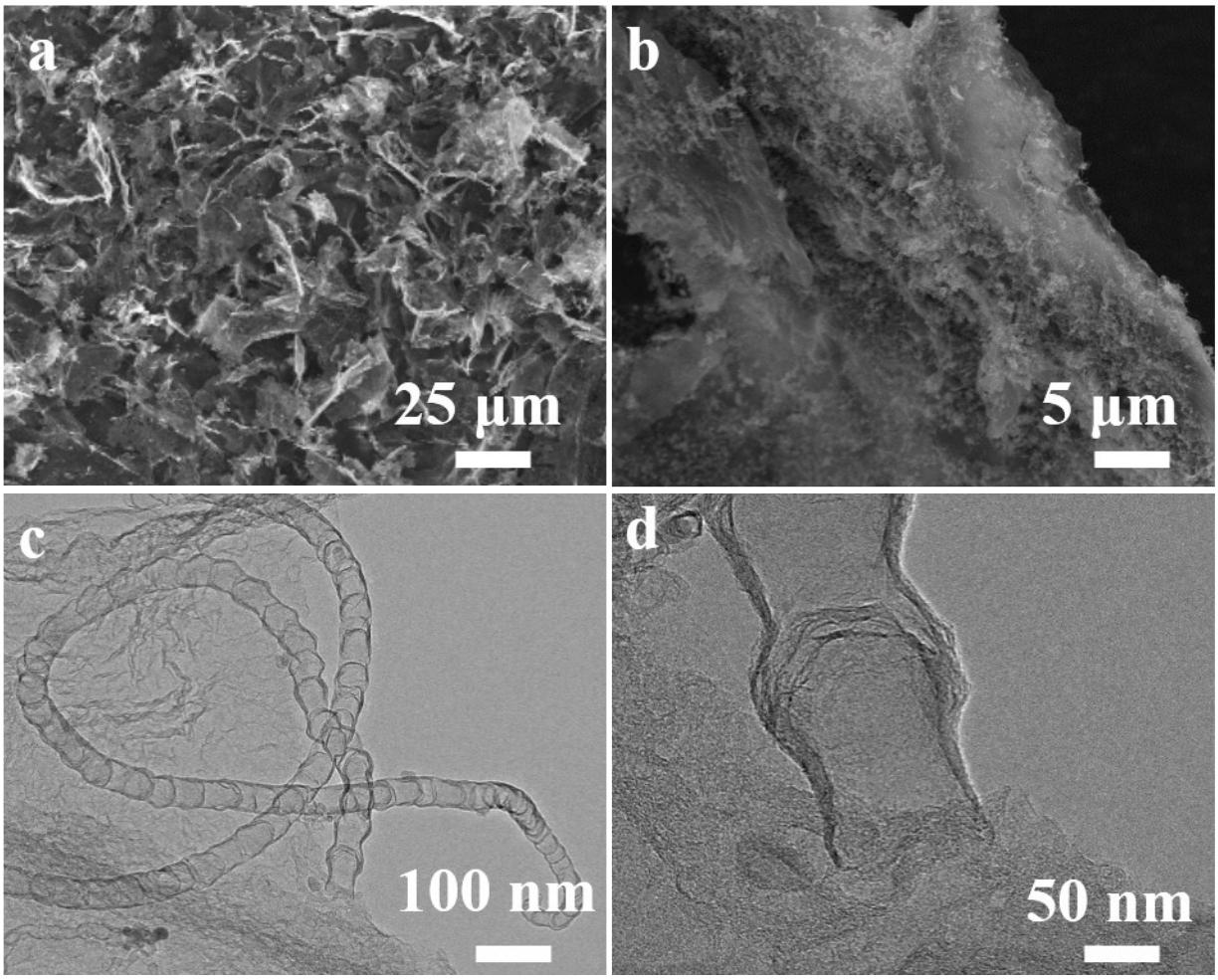

Figure S8. The morphological images of as-synthesized N-CNTs–GF. a, b) SEM images of the N-CNTs–GF; c) TEM image of N-CNTs–GF where the bamboo-like N-CNTs grown on graphene sheets; d) The enlarged TEM image of a N-CNT grown on graphene sheet.

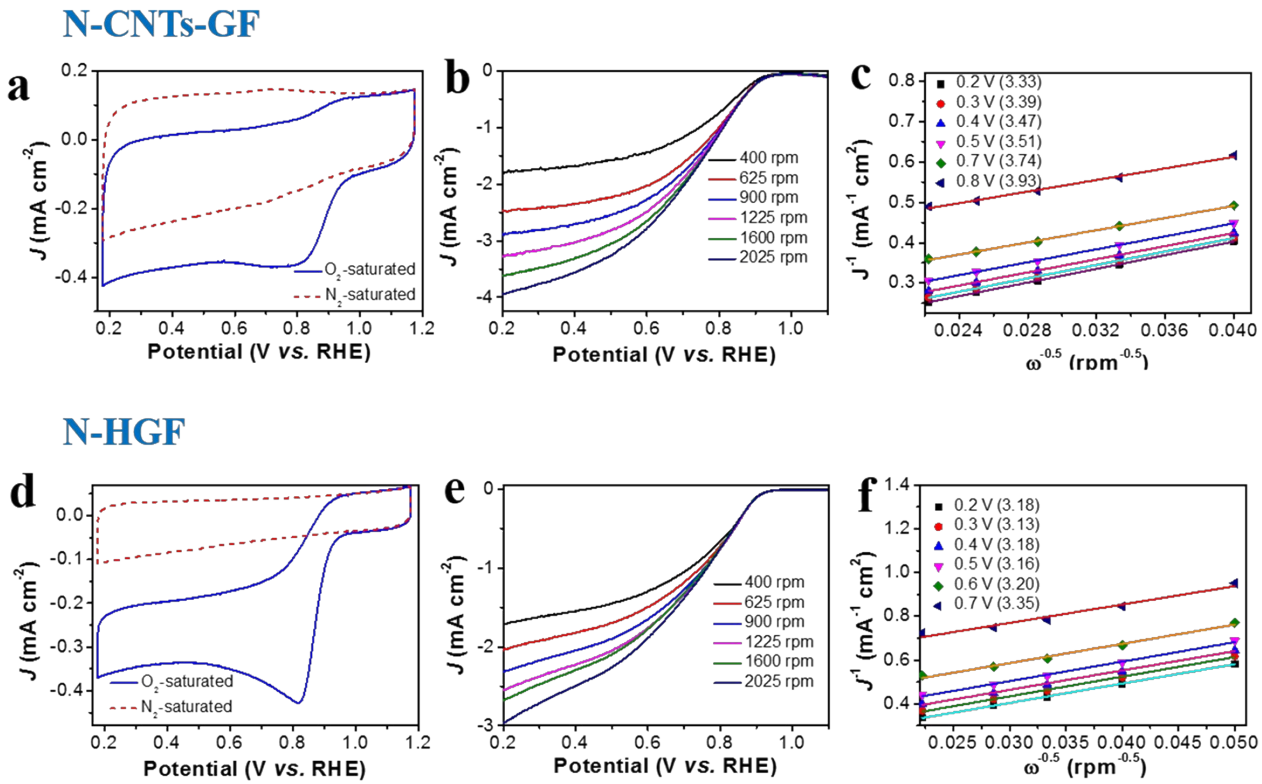

Figure S9. Cycle voltammetry (CV) curves of a) N-CNTs–GF and d) N-HGF in O_2-saturated (solid line) and N_2-saturated (short dash line) 0.1 M KOH aqueous solution, scan rate: 10 mV s^{-1}; The polarization curves with various rotation speeds for b) N-CNTs–GF and e) N-HGF, and the corresponding K-L plots of c) N-CNTs–GF and f) N-HGF.

Table S1: The electronic conductivity of all samples

Sample	Conductivity (S m^{-1})
N-CNTs–HGF	1469
N-CNTs–GF	1508
N-HGF	914

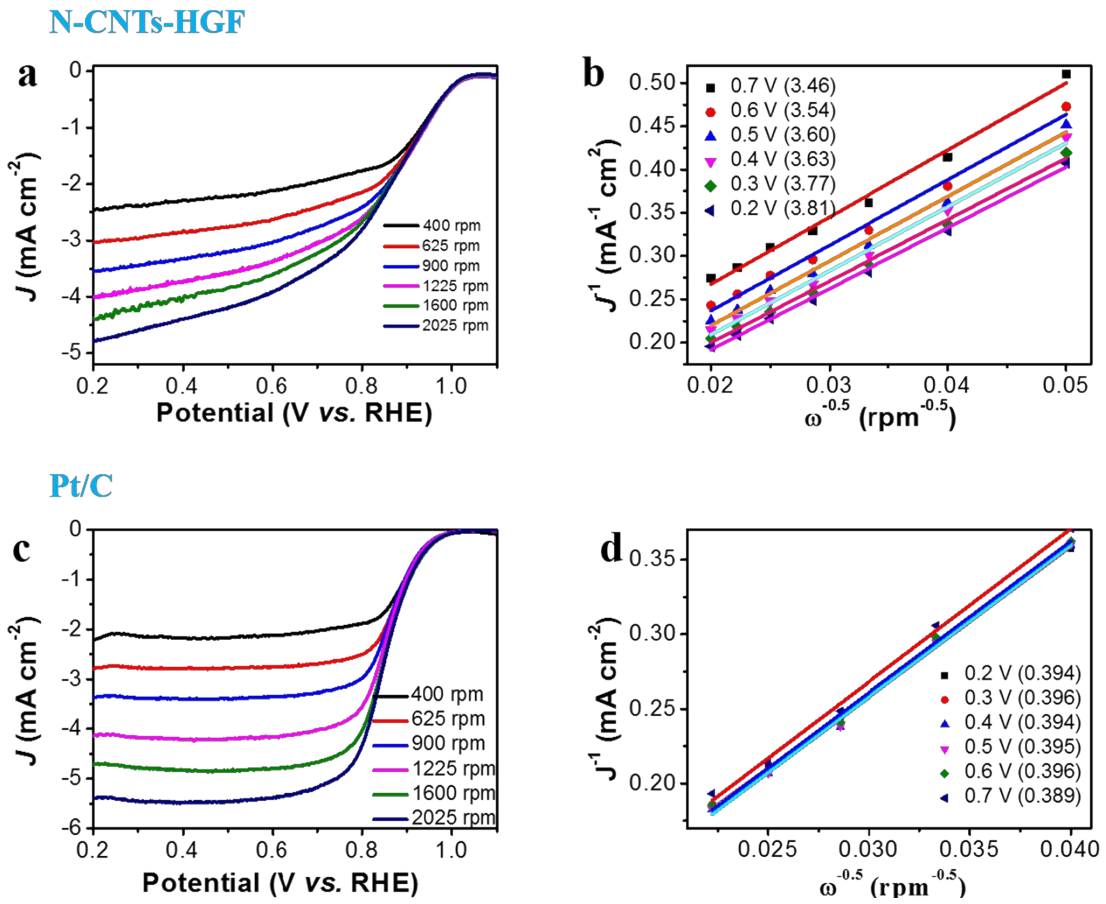

Figure S10. The polarization curves with different rotation speeds for a) N-CNTs–HGF and c) Pt/C; and the corresponding K-L plots for b) N-CNTs–HGF and d) Pt/C.

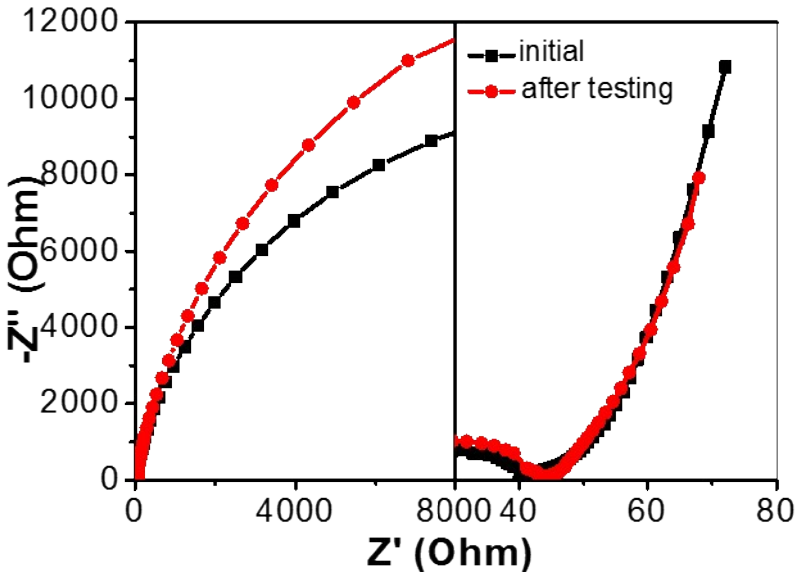

Figure S11. Nyquist plots of N-CNTs–HGF before and after electrochemical tests.

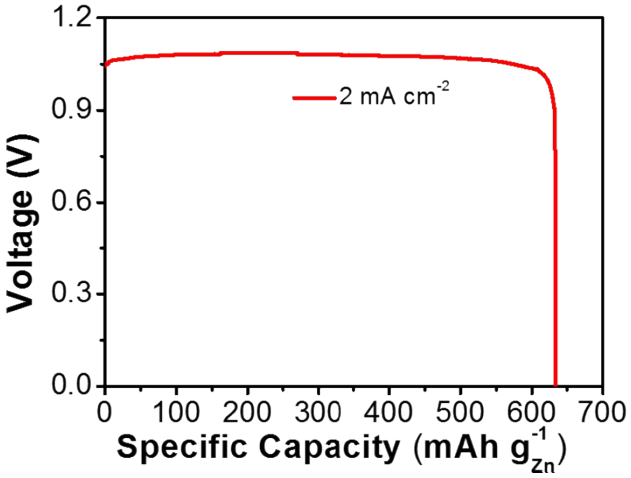

Figure S12. The specific capacity of the N-CNTs–HGF at the current density of 2 mA cm^{-2}.