Supporting Information

Porous graphene doped with Fe/N/S incorporated Fe_3O_4 nanoparticles for efficient oxygen reduction reaction

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Electrochemical evaluations

Electrode potentials conversion. Potentials measured versus Ag/AgCl electrode were converted to a reversible hydrogen electrode (RHE) scale based on the following formula [S1-S4]:

$$E_{vsRHE}=E_{vsAg/AgCl}+E^0_{Ag/AgCl}+0.059pH \text{ (in volts)} \quad (1)$$

Kouteky-Levich (K-L) plots. The working electrode was scanned cathodically at the rate of 10 mV s^{-1} with the rotation speed from 225 to 2500 rpm. The relevant K-L plots ($-J^{-1}$ *vs.* $\omega^{-1/2}$) are calculated by the following formula [S2]:

$$\frac{1}{J}=\frac{1}{J_k}+\frac{1}{J_d}=\frac{1}{J_k}+\frac{1}{B\omega^{1/2}} \quad (2)$$

$$B=0.62nFC_0D_0^{2/3}\mu^{-1/6} \quad (3)$$

where B and ω are the reciprocal of the slope and the angular velocity of the electrode, respectively, n is the transferred electrons for ORR, F is the Faraday constant (F=96485 C mol^{-1}), μ is the kinematic viscosity of the electrolyte (0.01 cm^2 s^{-1}), D_0 is the diffusion coefficient of O_2 in 0.1 M KOH (D_0=1.9×10^{-5} cm^2 s^{-1}), and C_0 is the concentration of O_2 (C_0=1.2×10^{-6} mol cm^{-3}). Furthermore, J_k is the kinetic-limiting current density, and J_d is the diffusing-limiting current density. The constant 0.62 is adopted when the rotation rate is expressed in rad s^{-1}. J_k can be calculated from J_d according to eq2 [S2]:

$$J_k=\frac{J\times J_d}{J_d-J} \quad (4)$$

The hydrogen peroxide yield (%H_2O_2) was calculated by the following equation [S4]:

$$\%H_2O_2=200\frac{{}^{i_r}/_N}{i_d+{}^{i_r}/_N}=200\frac{i_r}{Ni_d+i_r}$$

Where i_d is the disk current, i_r is ring current, and N is the Pt ring current collection efficiency (0.37). The current-time (*i-t*) chronoamperometric response curves were recorded at -0.35 V *vs.* Ag/AgCl in O_2-saturated 0.1 M KOH solution at a rotating rate of 1600 rpm.

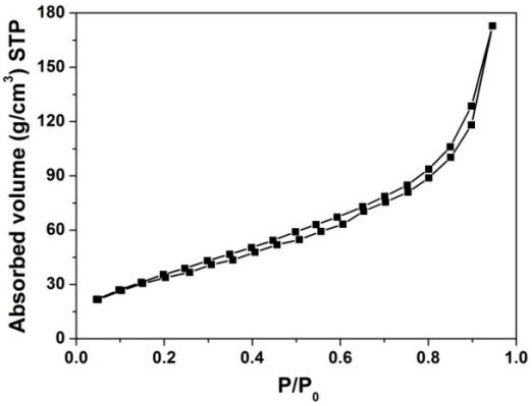

Figure S1 The N_2 adsorption/deposition isotherms of HSG.

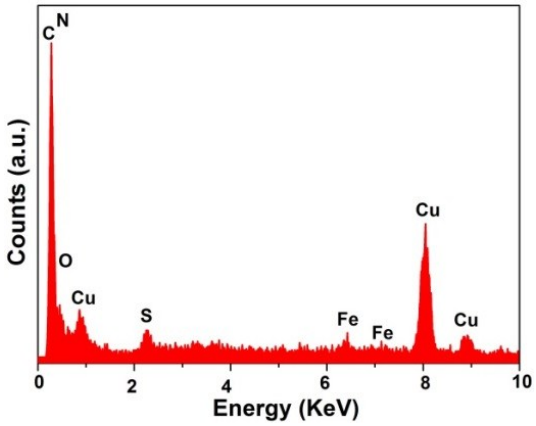

Figure S2 The EDS spectrum of the Fe_3O_4/FeNSG-3.

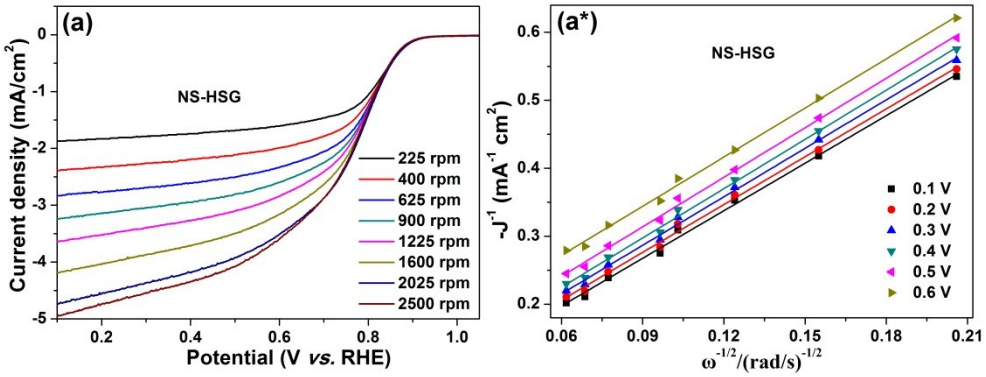

Figure S3 (a) RDE polarization curves on NS-HSG catalyst at different rotation rates, (a*) is the corresponding picture of K-L plots of $-J^{-1}$ versus $\omega^{-1/2}$.

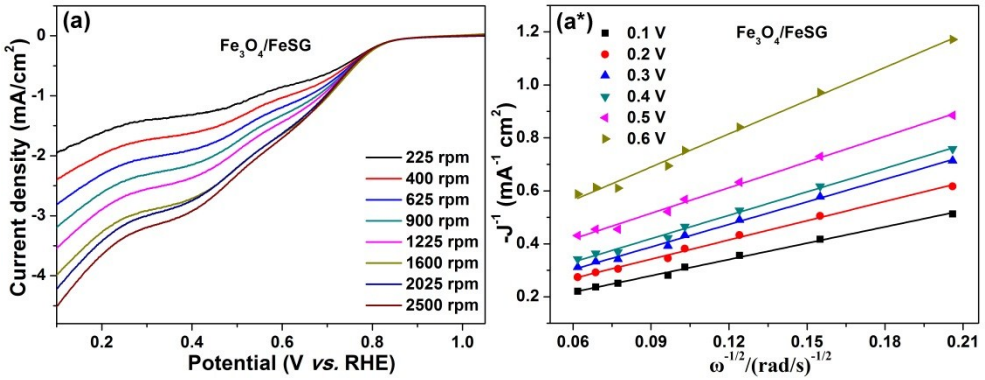

Figure S4 (a) RDE polarization curves on Fe_3O_4/FeSG catalyst at different rotation rates, (a*) is the corresponding picture of K-L plots of $-J^{-1}$ versus $\omega^{-1/2}$.

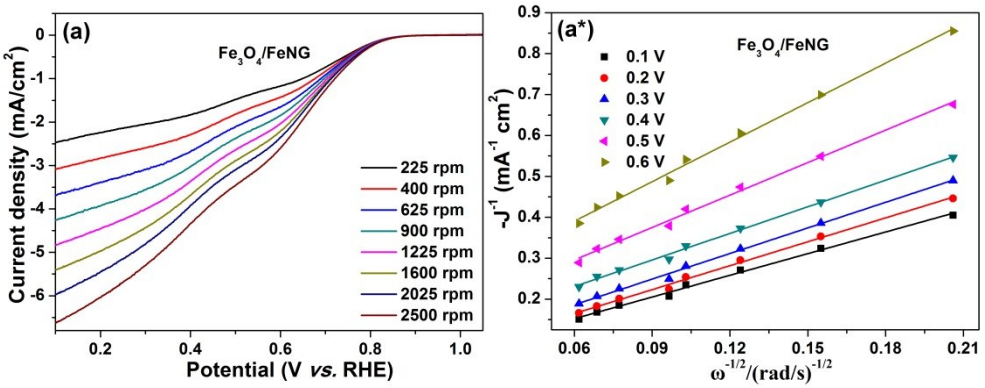

Figure S5 (a) RDE polarization curves on Fe_3O_4/FeNG catalyst at different rotation rates, (a*) is the corresponding picture of K-L plots of $-J^{-1}$ versus $\omega^{-1/2}$.

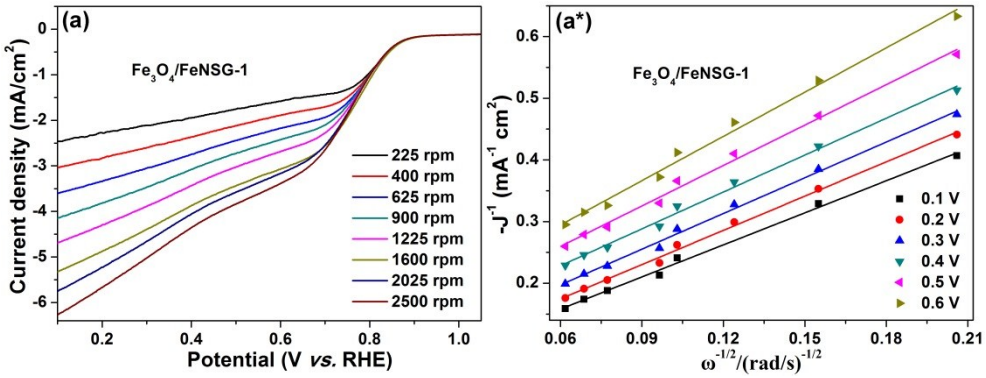

Figure S6 (a) RDE polarization curves on Fe_3O_4/FeNSG-1 catalyst at different rotation rates, (a*) is the corresponding picture of K-L plots of $-J^{-1}$ versus $\omega^{-1/2}$.

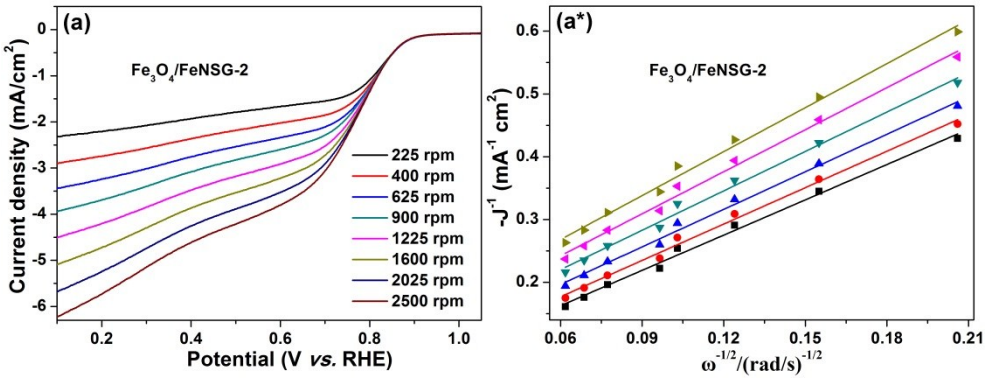

Figure S7 (a) RDE polarization curves on Fe_3O_4/FeNSG-2 catalyst at different rotation rates, (a*) is the corresponding picture of K-L plots of $-J^{-1}$ versus $\omega^{-1/2}$.

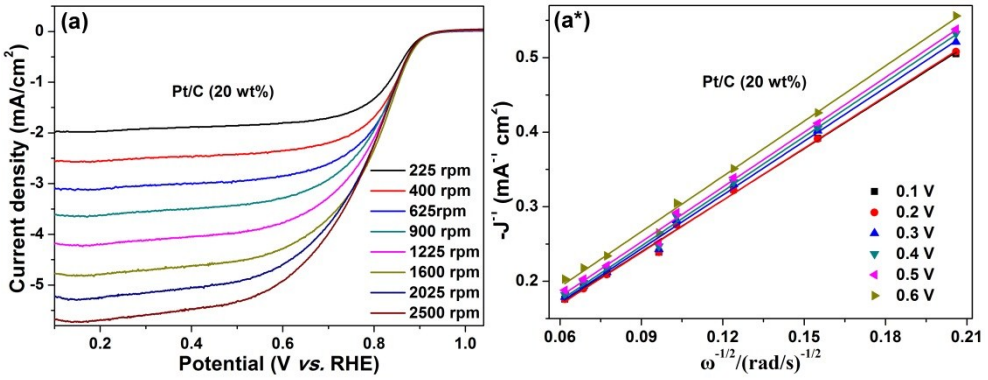

Figure S8 (a) RDE polarization curves on Fe_3O_4/FeNSG-2 catalyst at different rotation rates, (a*) is the corresponding picture of K-L plots of $-J^{-1}$ versus $\omega^{-1/2}$.

Table S1 The I_D/I_G values of different Fe_3O_4/FeNSG hybrids, for comparison, the I_D/I_G values of pure HSG and NS-HSG are also presented.

Samples	I_D/I_G
HSG	0.956
NS-HSG	1.046
Fe_3O_4/FeNSG-1	0.961
Fe_3O_4/FeNSG-2	0.992
Fe_3O_4/FeNSG-3	0.963

Table S2 Textual parameters of N_2 adsorption/deposition analysis for different samples

Sample	S_{BET} (m^2 g^{-1})	Pore vol. (cm^3 g^{-1})
NS-HSG	406.4	0.236
Fe_3O_4/FeNSG-1	429.9	0.282
Fe_3O_4/FeNSG-2	608.6	0.415
Fe_3O_4/FeNSG-3	530.5	0.311

References

[S1] Y. Li, J. Yang, N. Zhao, J. Huang, Y. Zhou, K. Xu, N. Zhao, *Appl. Catal., A*, 2017, **534**, 30-39.

[S2] Y. Zhou, C. Yen Hsu, S. Fu, G. Yang, C. Zhu, D. Du, P. Wo Ching, X. Cheng, J. Yang, C.M. Wai, Y. Lin, *Green Chem.*, 2015, **17**, 3552-3560.

[S3] Y. Li, J. Yang, J. Huang, Y. Zhou, K. Xu, N. Zhao, X. Cheng, *Carbon*, 2017, **122**, 237-246.

[S4] P. Chen, T. Zhou, L. Xing, K. Xu, Y. Tong, H. Xie, L. Zhang, W. Yan, W. Chu, C. Wu, Y. Xie, *Angew. Chem. Int. Ed.*, 2017, **56**, 610-614.