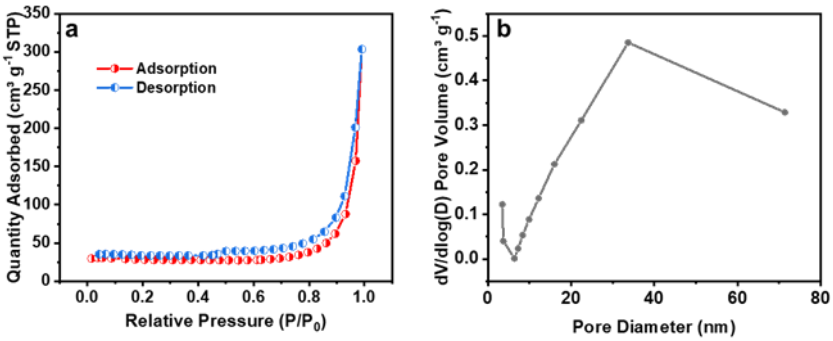

Figure S1 (a) N_2 adsorption–desorption isotherm, and (b) Barret-Joyner-Halenda (BJH) pore size distribution of Fe_3O_4/NGA-1.0-900.

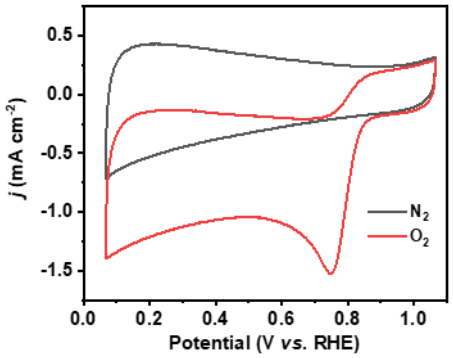

Figure S2 Cyclic voltammetry (CV) curves recorded for Fe_3O_4/NGA-1.0-900 in N_2 and O_2-saturated 0.1 M KOH at a scan rate of 50 mV s^{-1}.

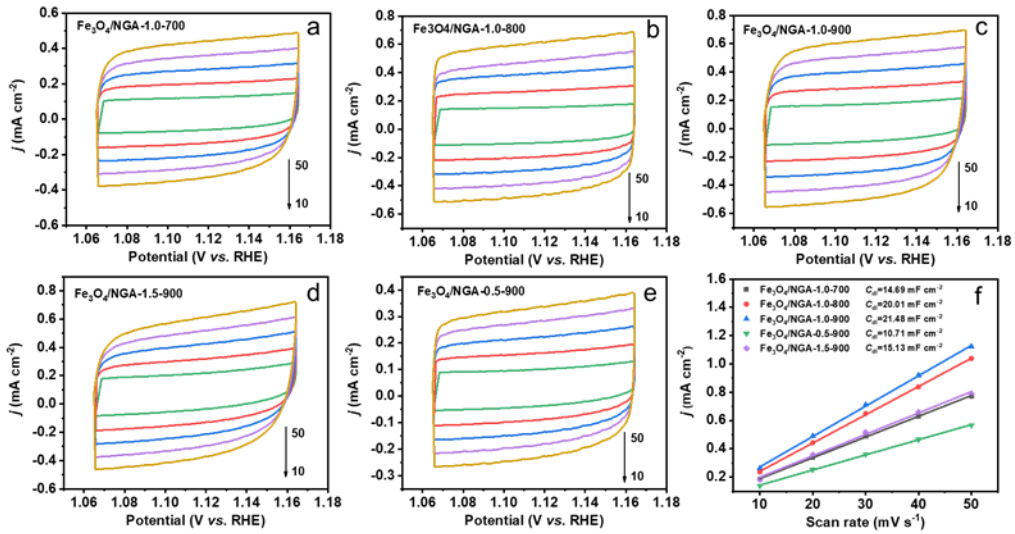

Figure S3 CV curves recorded for (a) Fe_3O_4/NGA-1.0-700, (b) Fe_3O_4/NGA-1.0-800, (c) Fe_3O_4/NGA-1.0-900, (d) Fe_3O_4/NGA-1.5-900, and (e) Fe_3O_4/NGA-0.5-900 at the scan rates of 10, 20, 30, 40, and 50 mV s^{-1}. (f) Current densities (recorded at the potential of 1.115 V) as a function of the scan rate derived from (a)–(e).

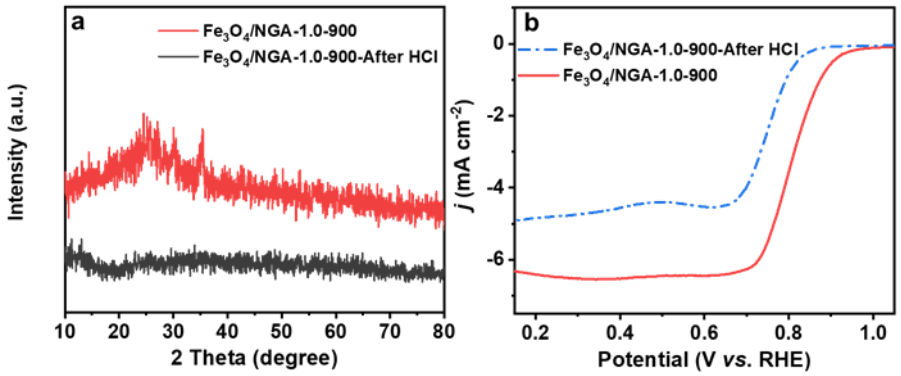

Figure S4 (a) X-ray diffraction (XRD) patterns and (b) Linear sweep voltammetry (LSV) curves recorded for Fe_3O_4/NGA-1.0-900 before and after acid etching.

Abbreviation List

Fe_3O_4/NGA: Fe_3O_4-decorated N-doped graphene aerogel.
ORR: oxygen reduction reaction.
PEMFCs: proton exchange membrane fuel cells.
2D: two-dimensional.
3D: three-dimensional.
RRDE: rotating ring disk electrode.
RDE: rotating disk electrode.
CV: cyclic voltammetry.
LSV: linear sweep voltammetry.
ECSA: electrochemical active surface area.
C_{dl}: double layer capacitance.

Table S1 Comparison of the electrochemical ORR performances of Fe_3O_4/NGA-1.0-900 recorded with the representative transition-metal-based catalysts (electrolyte is 0.1 M KOH).

Catalysts	E_{onset}[a] (V)	$E_{1/2}$[b] (V)	j[c] (mA cm^{-2})	Ref
Fe_3O_4/NGA-1.0-900	1.02	0.81	6.2	This work
Co_9S_8/C nanosheets	0.89	0.78	4.8	[1]
N-Co_3O_4@NC-2	0.89	0.77	5.9	[2]
Cu-N/C	0.91	0.81	5.5	[3]
ZnCo-HNC	1.05	0.82	6.0	[4]
FeCo NPs-N-CNTs	1.03	0.89	6.2	[5]
MgO@Phen-Fe-800-3/1	0.94	0.80	6.5	[6]
Fe/Co-CNT@MXene-8	1.02	0.85	6.0	[7]
Fe_3O_4/Fe_3N/Fe-N-C@PC-2.5	--	0.90	6.0	[8]
FeNx/Fe_2O_3-CNFs	--	0.81	6.0	[9]
Fe_3N/Fe-N-C-900	0.85	0.78	6.0	[10]

[a] Onset potential (vs. RHE). [b] Half-wave potential (vs. RHE). [c] Diffusion-limited current density.

References

1. Li L, Song L, Guo H, Xia W, Jiang C, Gao B, et al. N-Doped porous carbon nanosheets decorated with graphitized carbon layer encapsulated Co_9S_8 nanoparticles: an efficient bifunctional electrocatalyst for the OER and ORR. Nanoscale. 2019; 11: 901-907.
2. Wang Z, Xu W, Chen X, Peng Y, Song Y, Lv C, et al. Defect-rich nitrogen doped Co_3O_4/C porous nanocubes enable high-efficiency bifunctional oxygen electrocatalysis. Adv Funct Mater. 2019; 29: 1902875.
3. Lai Q, Zhu J, Zhao Y, Liang Y, He J, Chen J. MOF-based metal-doping-induced synthesis of hierarchical porous Cu-N/C oxygen reduction electrocatalysts for Zn–Air batteries. Small. 2017; 13: 1700740.
4. Lin SY, Xia LX, Cao Y, Meng HL, Zhang L, Feng JJ, et al. Electronic regulation of ZnCo Dual-ATOMIC active sites entrapped in 1D@2D hierarchical N-doped carbon for efficient synergistic catalysis of oxygen reduction in Zn-Air battery. Small. 2022; 18: e2107141.
5. Zhang T, Bian J, Zhu Y, Sun C. FeCo nanoparticles encapsulated in N-Doped carbon nanotubes coupled with layered double (Co, Fe) hydroxide as an efficient bifunctional catalyst for rechargeable Zinc-Air batteries. Small. 2021; 17: e2103737.
6. Zhan Y, Zeng H, Xie F, Zhang H, Zhang W, Jin Y, et al. Templated growth of Fe/N/C catalyst on hierarchically porous carbon for oxygen reduction reaction in proton exchange membrane fuel cells. J Power Sources. 2019; 431: 31-39.
7. Zhang C, Dong H, Chen B, Jin T, Nie J, Ma G. 3D MXene anchored carbon nanotube as bifunctional and durable oxygen catalysts for Zn–air batteries. Carbon. 2021; 185: 17-26.
8. Hao R, Chen J, Wang Z, Zhang J, Gan Q, Wang Y, et al. Iron polyphthalocyanine-derived ternary-balanced Fe3O4/Fe3N/Fe-N-C@PC as a high-performance electrocatalyst for the oxygen reduction reaction. Sci China Mater. 2021; 64: 2981-2996.
9. Yu Q, Lian S, Li J, Yu R, Xi S, Wu J, et al. FeN_x and γ-Fe_2O_3 co-functionalized hollow graphitic carbon nanofibers for efficient oxygen reduction in an alkaline medium. J Mater Chem A. 2020; 8: 6076-6082.
10. Xue N, Liu J, Wang P, Wang C, Li S, Zhu H, et al. Scalable synthesis of Fe_3N nanoparticles within N-doped carbon frameworks as efficient electrocatalysts for oxygen reduction reaction. J Colloid Interface Sci. 2020; 580: 460-469.