Supporting Infromation

Thermally-Driven Fabrication of Single-Atom Catalysts from Bulk Metal Foil for Oxygen Reduction

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The crystalline phase of the products was analyzed using powder XRD measurements conducted with a Bruker D8 Advance X-ray diffractometer. The microstructure of the samples was characterized by TEM, measured at 200 kV using a FEI TECNAI G20 field-emission TEM. Raman spectrum analysis, employing a Horiba HR Evolution instrument with laser excitation at 633 nm and a sweep range from 200 to 1800 cm-1, was utilized to discern the characteristic vibrational modes of the synthesized materials, particularly focusing on the D-band and G-band of the carbonized samples. The specific surface area of the catalysts was determined through Brunauer-Emmett-Teller (BET) analysis, performed with a Micromeritics Tristar II 3020 instrument. The surface elemental states of the samples were examined by XPS, utilizing an Escalab 250Xi X-ray photoelectron spectrometer (Thermo Fisher) equipped with Mg Ka X-ray as the excitation source. The X-ray absorption fine structure spectra (Fe K-edge) were acquired at the beamline BL14W1 station of the Shanghai Synchrotron Radiation Facility, China. The data was gathered in transmission mode, employing an ionization chamber for Fe foil, FeO, and Fe2O3 in fluorescence excitation mode. All spectra were recorded under ambient conditions.

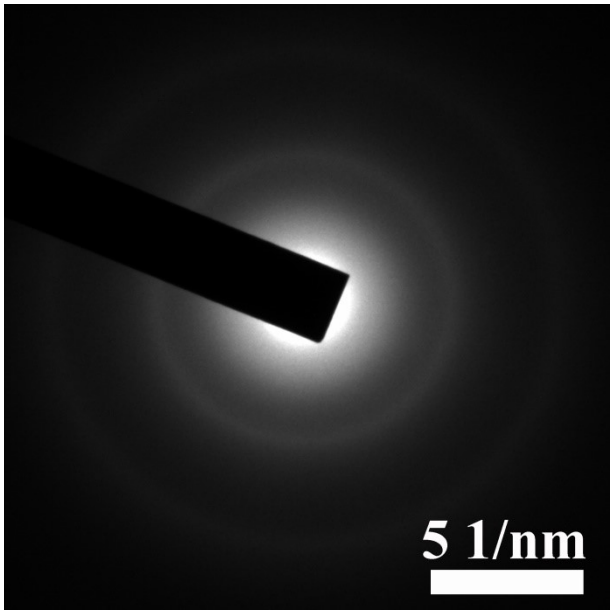

Figure S1. Selected-area electron diffraction pattern (SAED) of Fe-800.

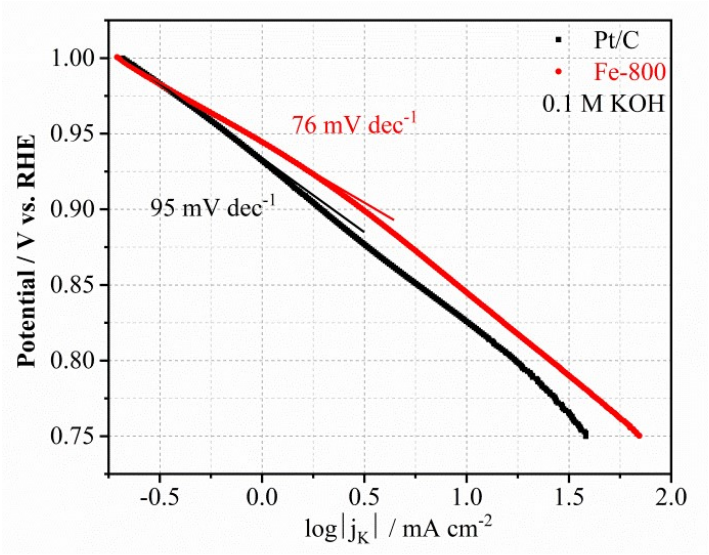

Figure S2. Tafel plots of Fe-800 and Pt/C.

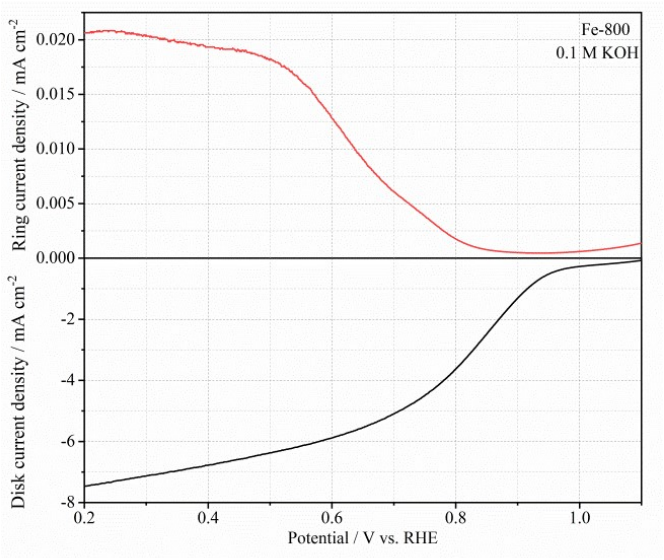

Figure S3. LSV curves obtained by the rotating ring disk electrode (RRDE) for Fe-800

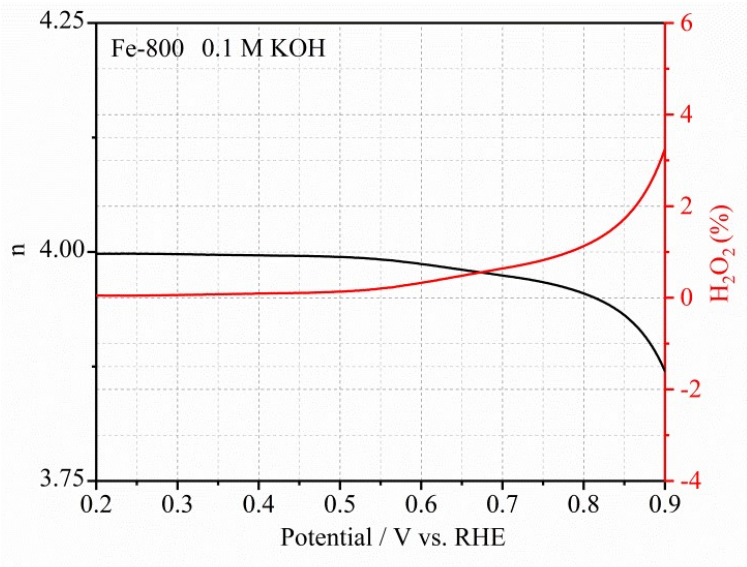

Figure S4. The electron transfer number and H_2O_2 yield measured by RRDE for Fe-800 within the potential range of 0.2 to 0.9 V.

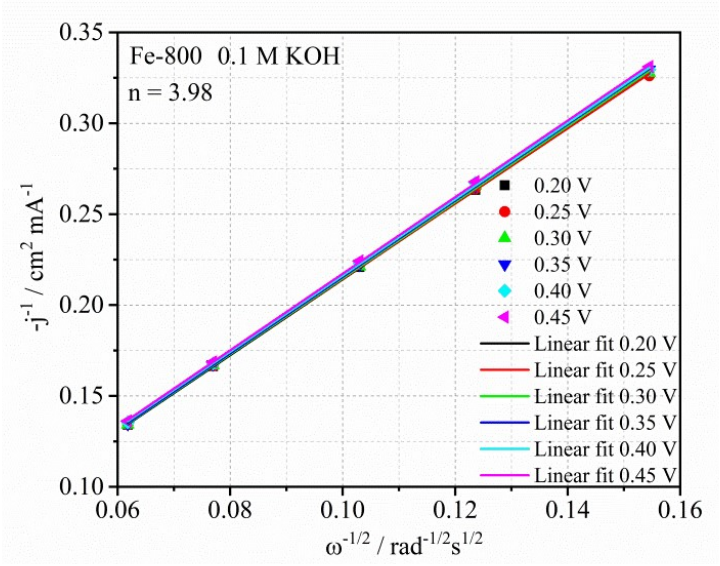

Figure S5. Koutecky–Levich (K–L) plots of Fe-800 in 0.1 M KOH

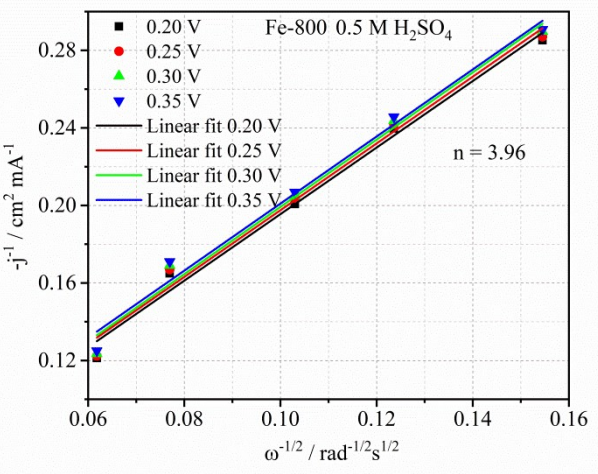

Figure S6. Koutecky–Levich (K–L) plots of Fe-800 in 0.1 M H_2SO_4

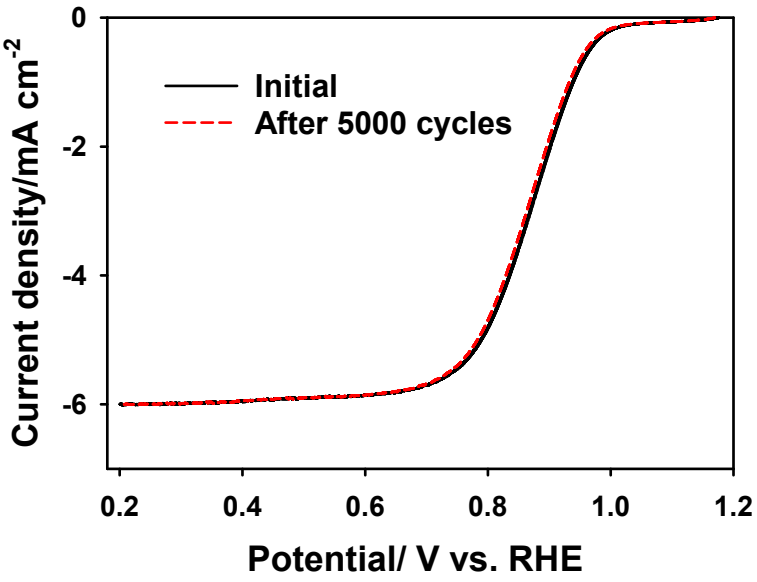

Figure S7. RDE polarization curves of Fe SAs/N-C before and after 5,000 cycles (0.6 to 1.0 V (vs. RHE)).

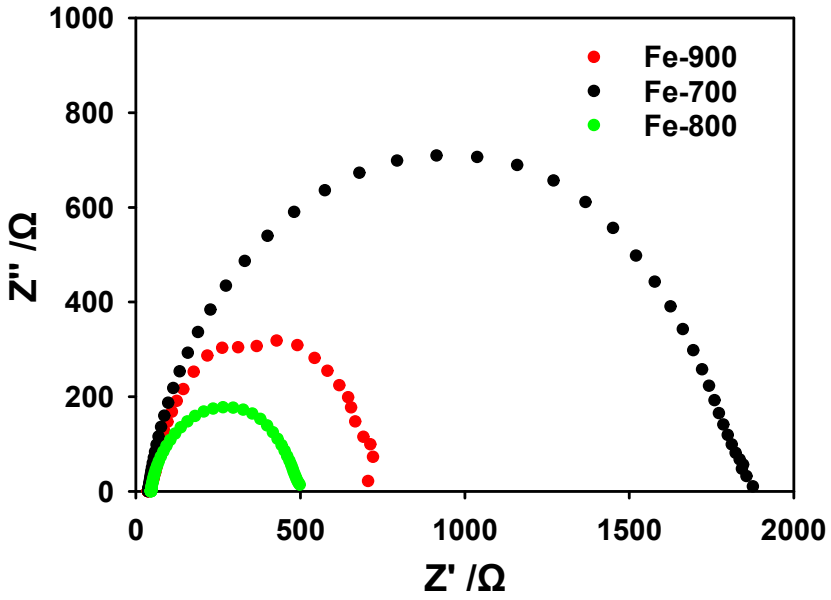

Figure S8. EIS analyses.

Table S1. Structural parameters extracted from the Fe K-edge EXAFS fitting. (S_0^2=0.85)

sample	Scattering pair	CN	R(Å)	$\sigma^2(10^{-3}Å^2)$	ΔE_0(eV)	R factor
Fe-800	Fe-N	4.1	2.00	4.9	2.0	0.003

S_0^2 is the amplitude reduction factor; CN is the coordination number; R is interatomic distance (the bond length between central atoms and surrounding coordination atoms); σ^2 is Debye-Waller factor (a measure of thermal and static disorder in absorber-scatterer distances); ΔE_0 is edge-energy shift (the difference between the zero kinetic energy value of the sample and that of the theoretical model). R factor is used to value the goodness of the fitting.

Error bounds that characterize the structural parameters obtained by EXAFS spectroscopy were estimated as N ± 20%; R ± 1%; σ^2 ± 20%; ΔE_0 ± 20%.

Table S2 Comparison of the half-wave potential in recently reported single-atom catalysts.

Catalyst	Half-wave potential（V vs. RHE）	Reference
Fe-800	0.87	This work
CHS	0.78	1
Fe SACs/NC	0.89	1
ZnN_4	0.82	2
Zn-SAs/NCNS	0.63	3
Zn-Sas/UNCNS-850	0.87	3
$Cu_{0.6}$-N-CNFs	0.81	4

REFERENCS

1. Z. Han, S. Chai, Y. Zhu, X. Yao, L. Yang, C. Du, X. Ma, C. Cao and M. Zou, *Chemical Engineering Journal*, 2024, **502**, 157943.
2. X. Sun, X. Li, H. Huang, W. Lu, X. Xu, X. Cui, L. Li, X. Zou, W. Zheng and X. Zhao, *Nano Letters*, 2024, **24**, 14602-14609.
3. T. Zhang, F. Wang, C. Yang, X. Han, C. Liang, Z. Zhang, Y. Li, A. Han, J. Liu and B. Liu, *Chem Catalysis*, 2022, **2**, 836-852.
4. F. Yan, L. Dong, J. Su, X. Liu, X. Han, J. Zang and Y. Wang, *Electrochemistry Communications*, 2022, **136**, 107245.

5. X. Ge, G. Su, W. Che, J. Yang, X. Zhou, Z. Wang, Y. Qu, T. Yao, W. Liu and Y. Wu, *ACS Catalysis*, 2020, **10**, 10468-10475.