# Supporting Information

# Rational Fabrication of Defect-Rich and Hierarchically Porous Fe-N-C Nanosheets as Highly Efficient Oxygen Reduction Electrocatalysts for Zinc-Air Battery

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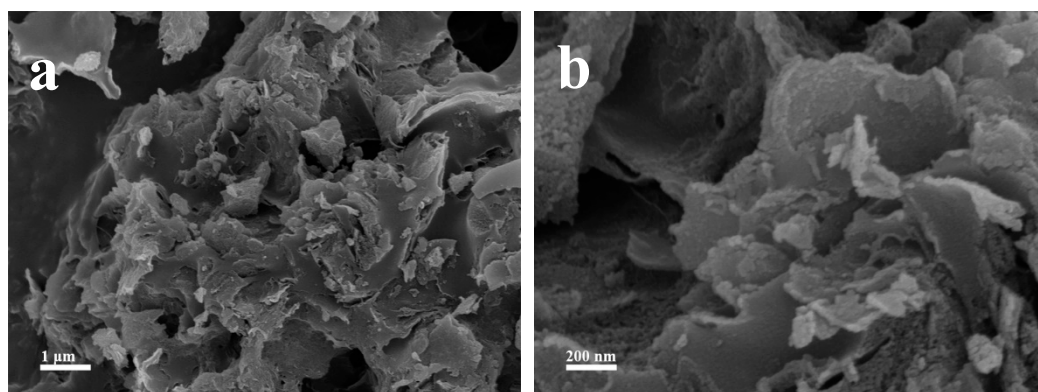* Correspondence: wuxy90@xju.edu.cn (X.W.); jxguo1012@163.com (J.G.)
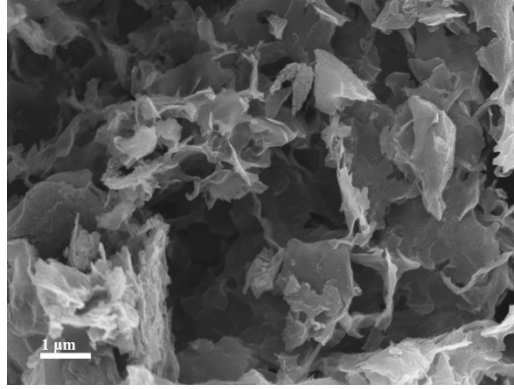
**Figure S1.** SEM images of Fe-N-CNSs-800.

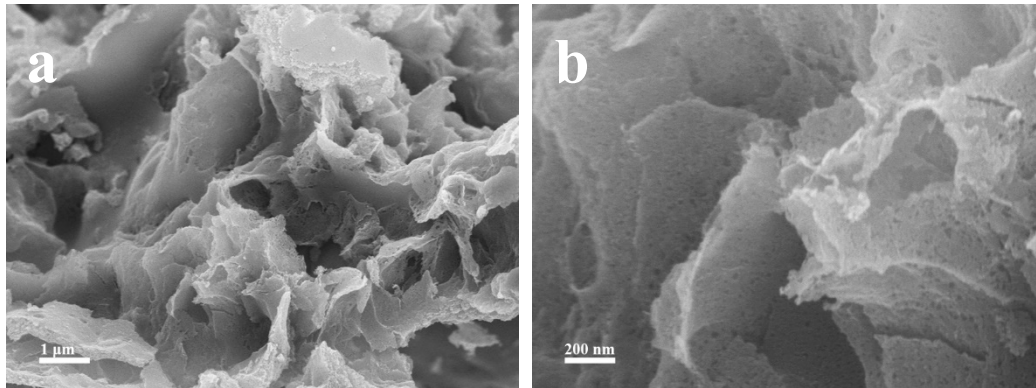**Figure S2**. SEM image of Fe-N-CNSs-900.



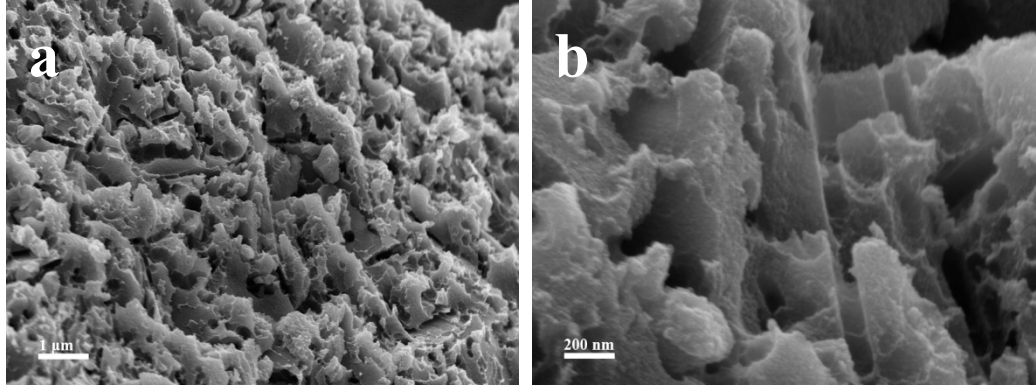**Figure S3**. SEM images of Fe-N-CNSs-1000



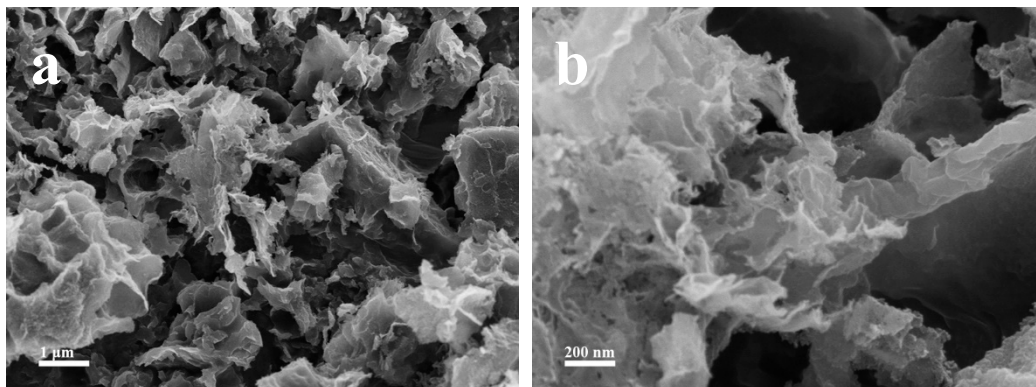**Figure S4.** SEM images of Fe-N-C-900.



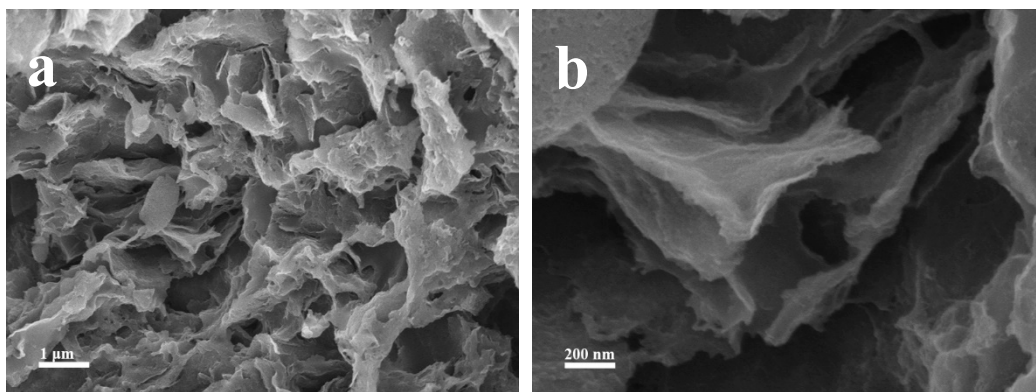**Figure S5.** SEM images of Fe-N-CNSs-15.

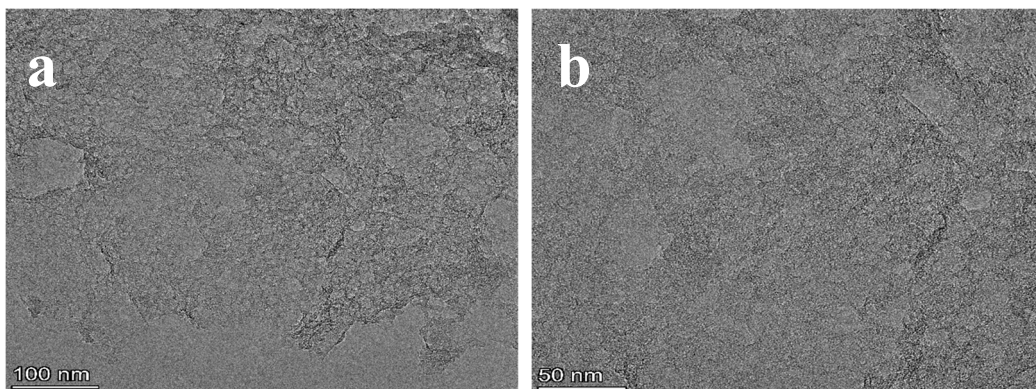**Figure S6.** SEM images of Fe-N-CNSs-60.



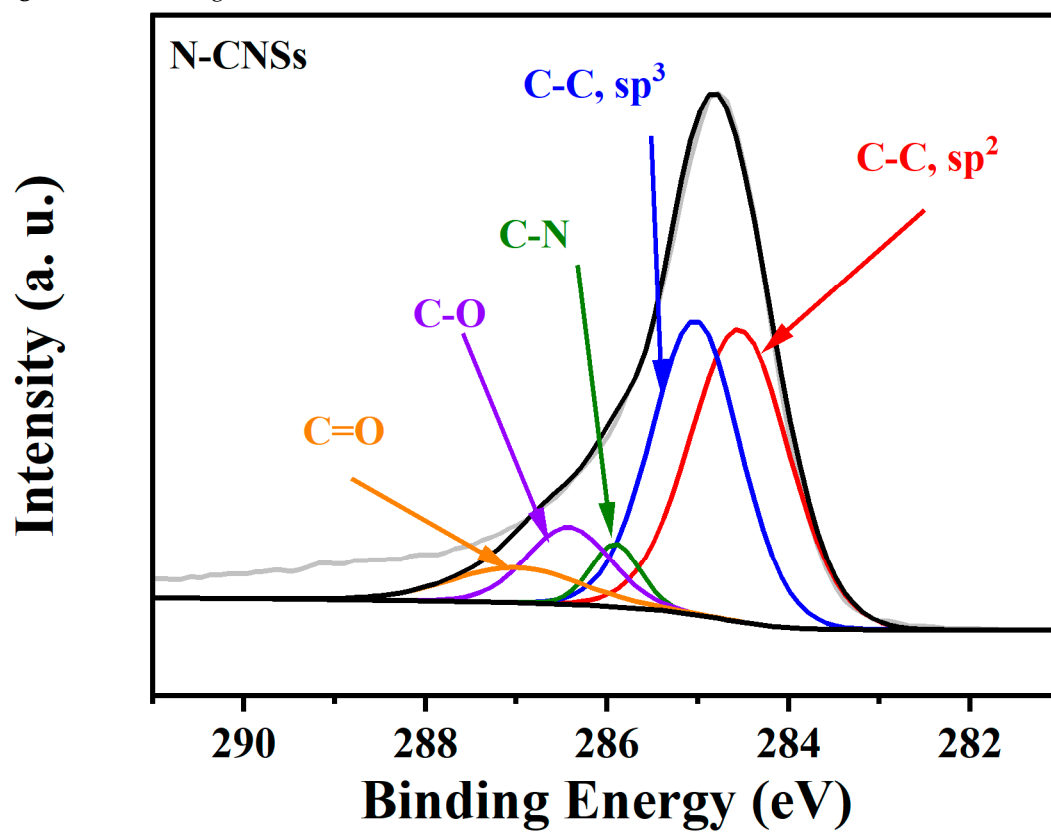**Figure S7.** TEM images of Fe-N-CNSs.



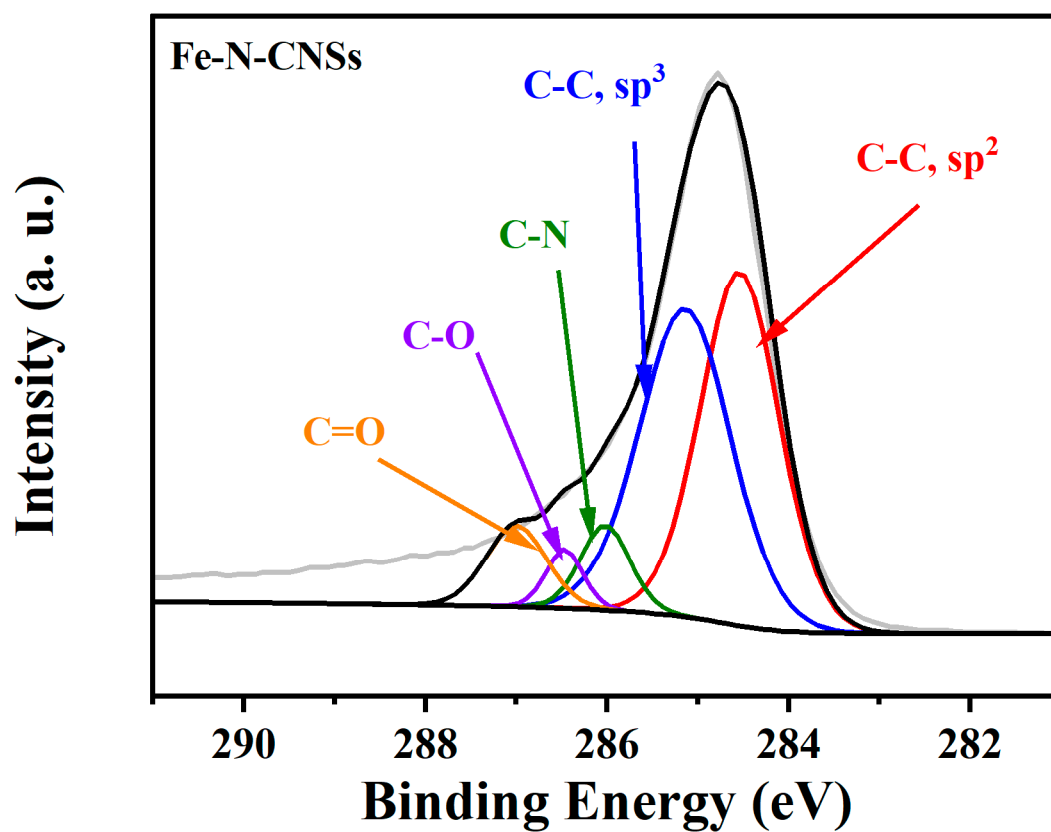**Figure S8.** High-resolution C 1s spectrum of N-CNSs.

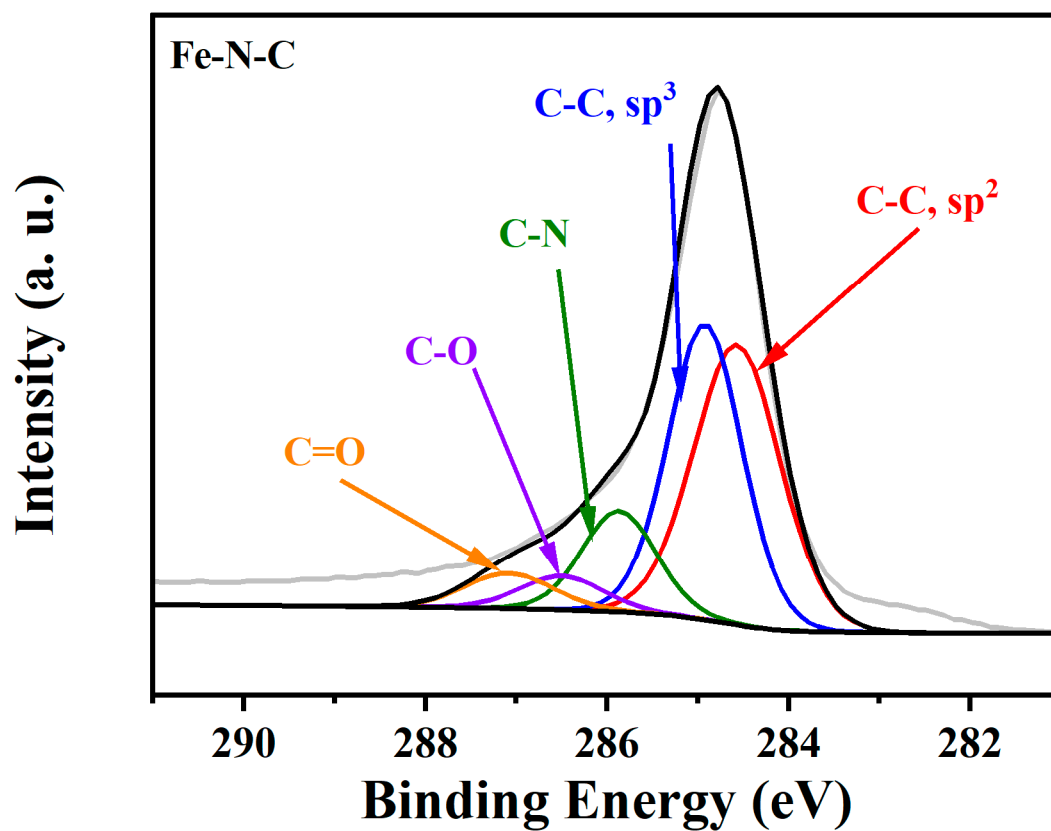**Figure S9.** High-resolution C 1s spectrum of Fe-N-CNSs.



**Figure S10**. High-resolution C 1s spectrum of Fe-N-C.
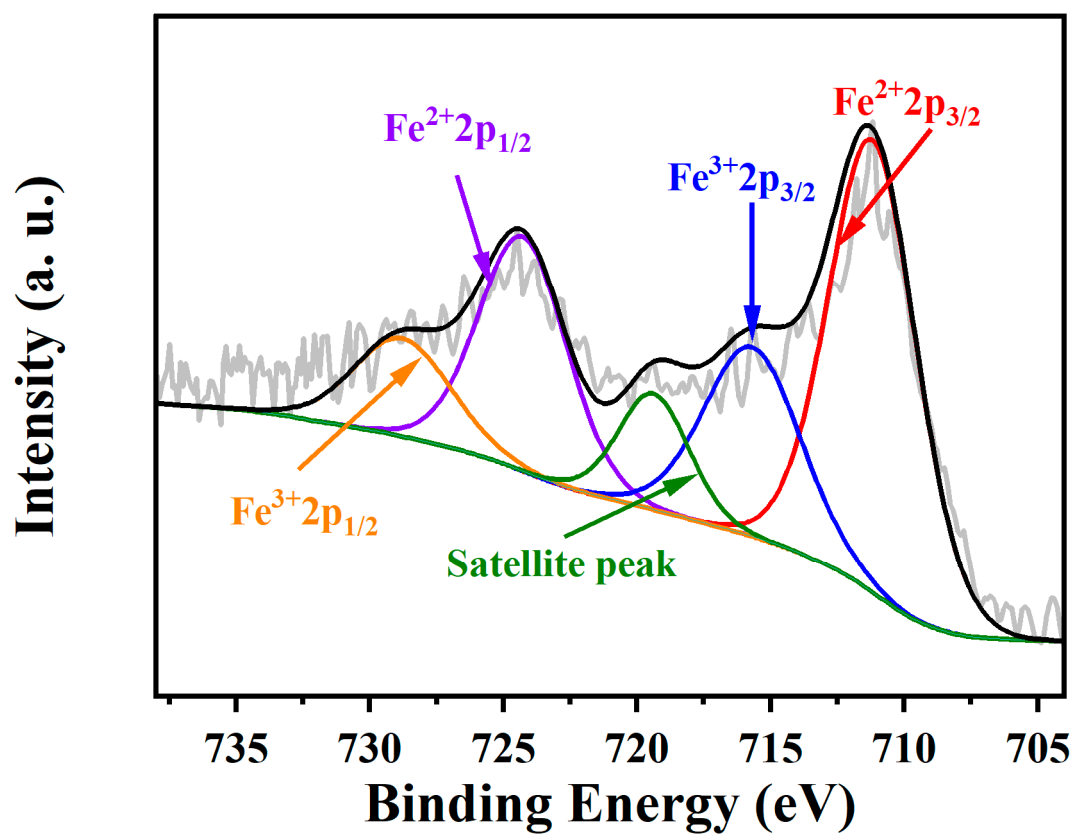
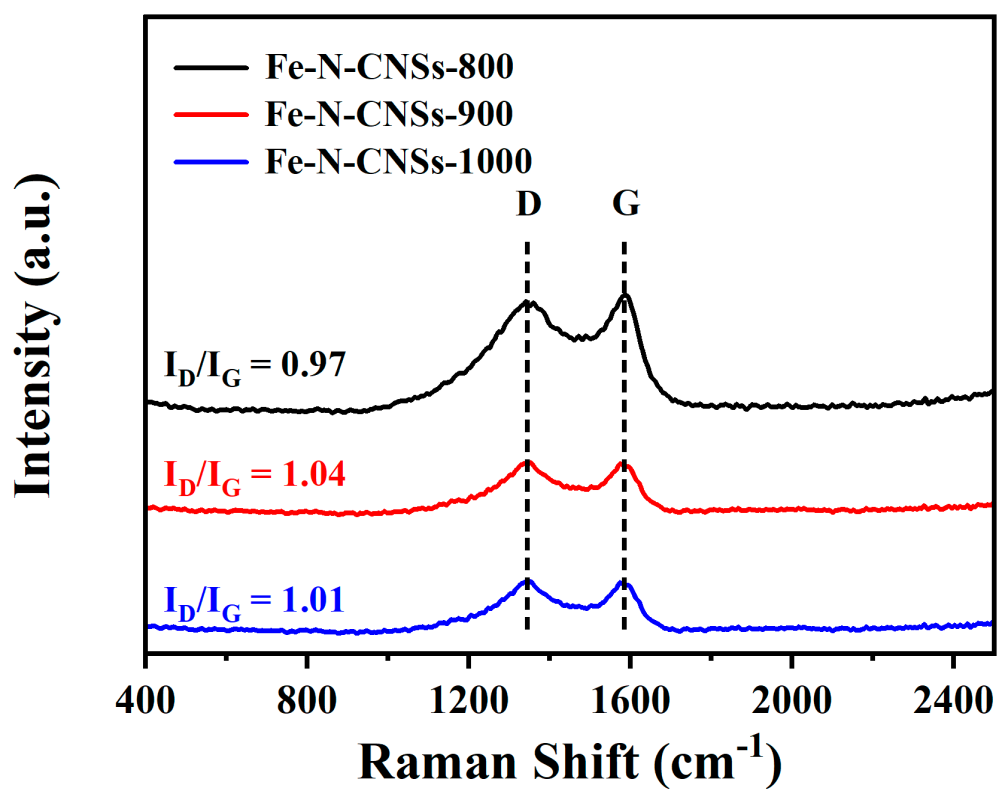**Figure S11**. High-resolution Fe 2p spectrum of Fe-N-CNSs.



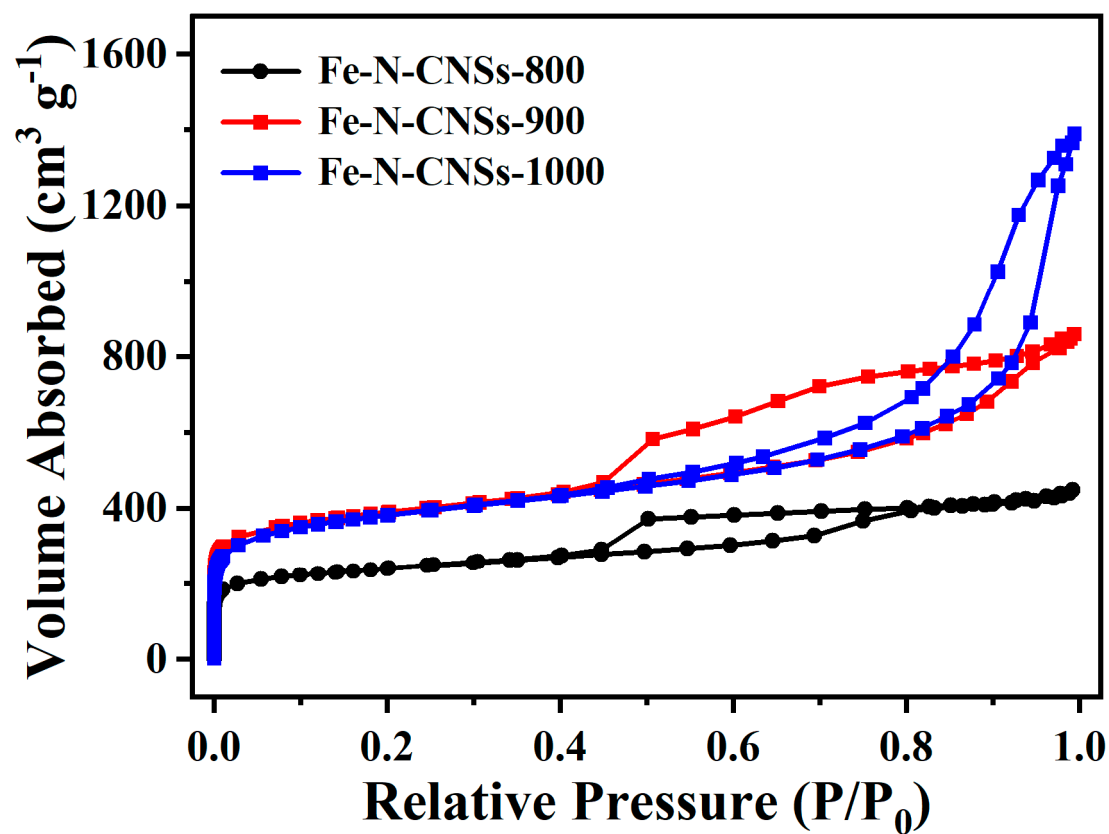**Figure S12**. Raman spectra of the synthesized materials.

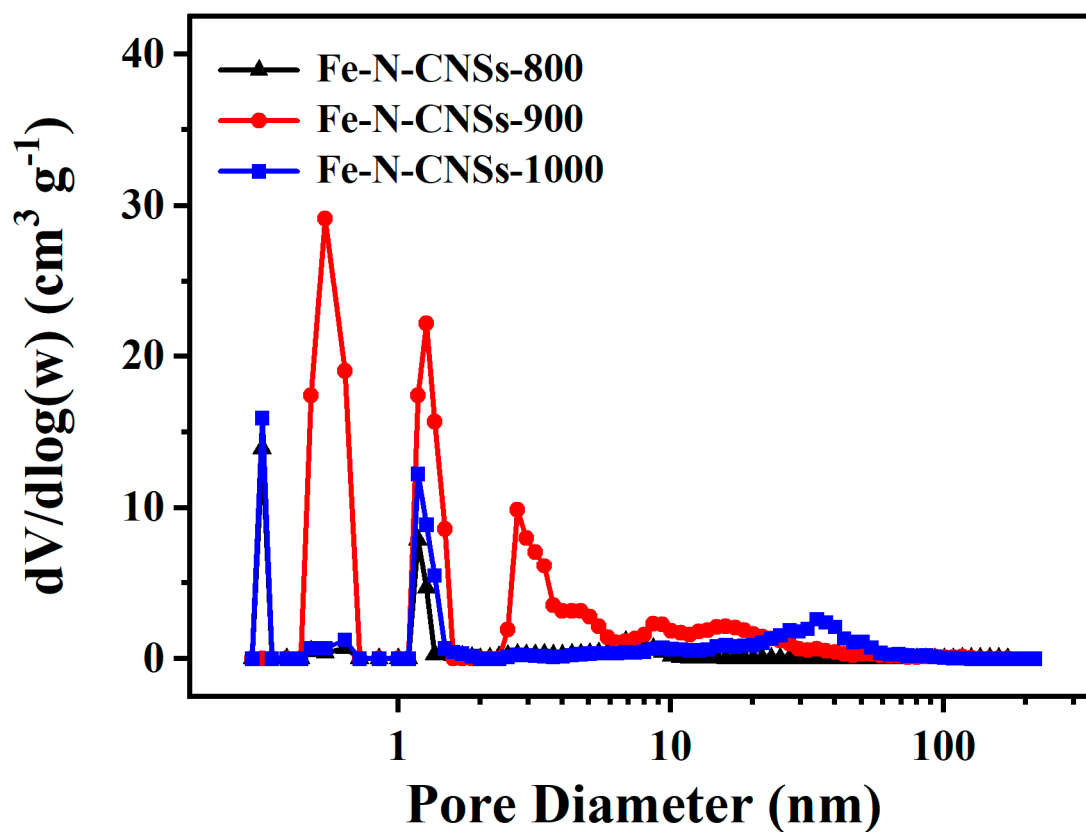Figure **S13**. N2 adsorption-desorption isotherms of Fe-N-CNSs-800, Fe-N-CNSs-900 and Fe-N-CNSs-1000.

Figure **S14**. The pore size distribution of Fe-N-CNSs-800, Fe-N-CNSs-900 and Fe-N-CNSs-1000.

**Table S1.** Comparison of ORR activity of various Fe-N-C catalysts.

| Catalysts | Electrolyte | $E_{1/2}$(V vs. RHE) | Reference |
|-----------|-------------|---------------------|-----------|
| **Fe-N-CNSs** | **0.1 M KOH** | **0.87** | **This work** |
| Fe-SAC/NC | 0.1 M KOH | 0.84 | *Nano Energy* **2020,** 72, 104670. |
| Fe-N-C/NG | 0.1 M KOH | 0.84 | *J. Alloy. Compd.* **2022,** 901, 163763. |
| Fe-AC-2 | 0.1 M KOH | 0.87 | *J. Mater. Chem. A* **2021,** 9, 7137–7142. |
| Fe-N-C/HPC-NH₃ | 0.1 M KOH | 0.80 | *Nano Res.* **2019,** 12, 1651–1657. |
| AC-N-Fe | 0.1 M KOH | 0.76 | *Chem. Eng. J.* **2022,** 446, 137256. |
| Fe-mela-mPD-N-C | 0.1 M KOH | 0.86 | *J. Alloy. Compd.* **2023,** 935, 168069. |
| FeN4-O-NCR | 0.1 M KOH | 0.94 | *Adv. Mater.* **2022,** 34, 2202544. |
| Fe-N-C/MXene | 0.1 M KOH | 0.84 | *ACS Nano* **2020,** 14, 2436–2444. |
| FeNSC-ZM | 0.1 M KOH | 0.87 | *Chem. Eng. J.* **2022,** 444, 136433. |

| | | | |
|---|---|---|---|
| Fe/Meso-NC-1000 | 0.1 M KOH | 0.88 | *Adv. Mater.* **2022,** 34, 2107291. |
| SA Fe@ZrO$_2$/NC | 0.1 M KOH | 0.86 | *Chem. Eng. J.* **2021,** 420, 129938. |

**Table S2.** Comparison of the performance of the primary zinc-air batteries with the present work.

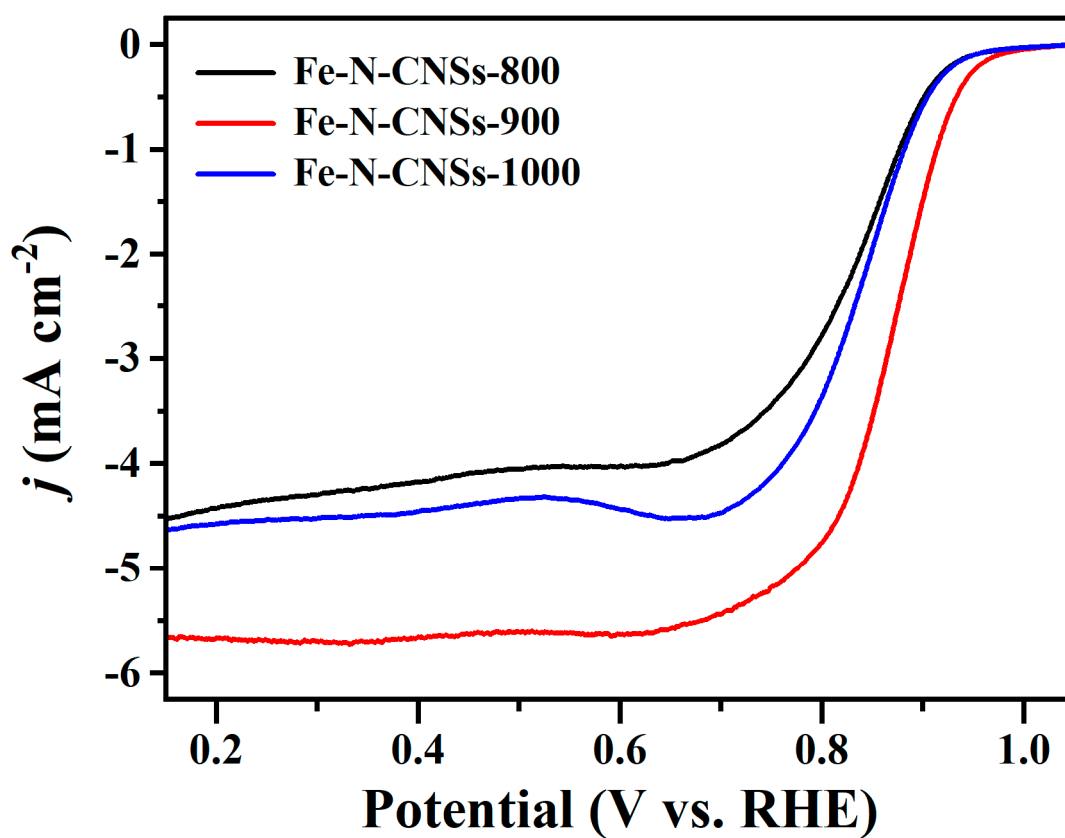| Catalysis | Peak power density (mW cm$^{-2}$) | Reference |
|---|---|---|
| **Fe-N-CNSs** | **128.20** | **This work** |
| IrFe-N-C | 113.9 | *ACS Catal.* **2022,** 12, 9397-9409 |
| M-Fe-NCNSs | 110.5 | *Chem. Eng. J.* **2020,** 393, 124702 |
| FeN$_x$/S-NC | 83 | *Appl. Surf. Sci.* **2022,** 580, 152255 |
| Fe-N-C | 167 | *Nano Energy* **2022,** 104, 107869 |
| FeNC | 118.05 | *Materials Today Energy* **2020,** 18, 100500 |
| AC-N-Fe | 124.2 | *Chem. Eng. J.* **2022,** 446, 137256 |
| P-Fe-N-CNTs | 145 | *Appl. Catal. B: Environ.* **2023,** 327, 122469 |
| Fe$_3$-N-C-800 | 80 | *New J. Chem.* **2022,** 46,989-994 |



Figure **S15.** LSV curves of Fe-N-CNSs-800, Fe-N-CNSs-900, Fe-N-CNSs-1000.
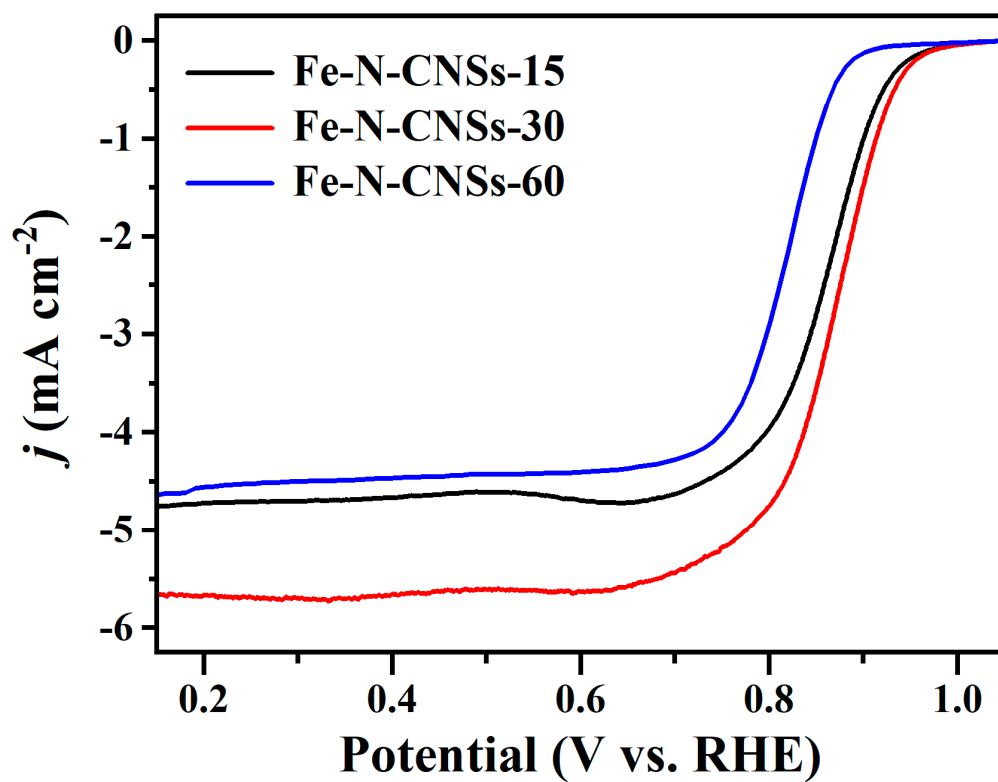
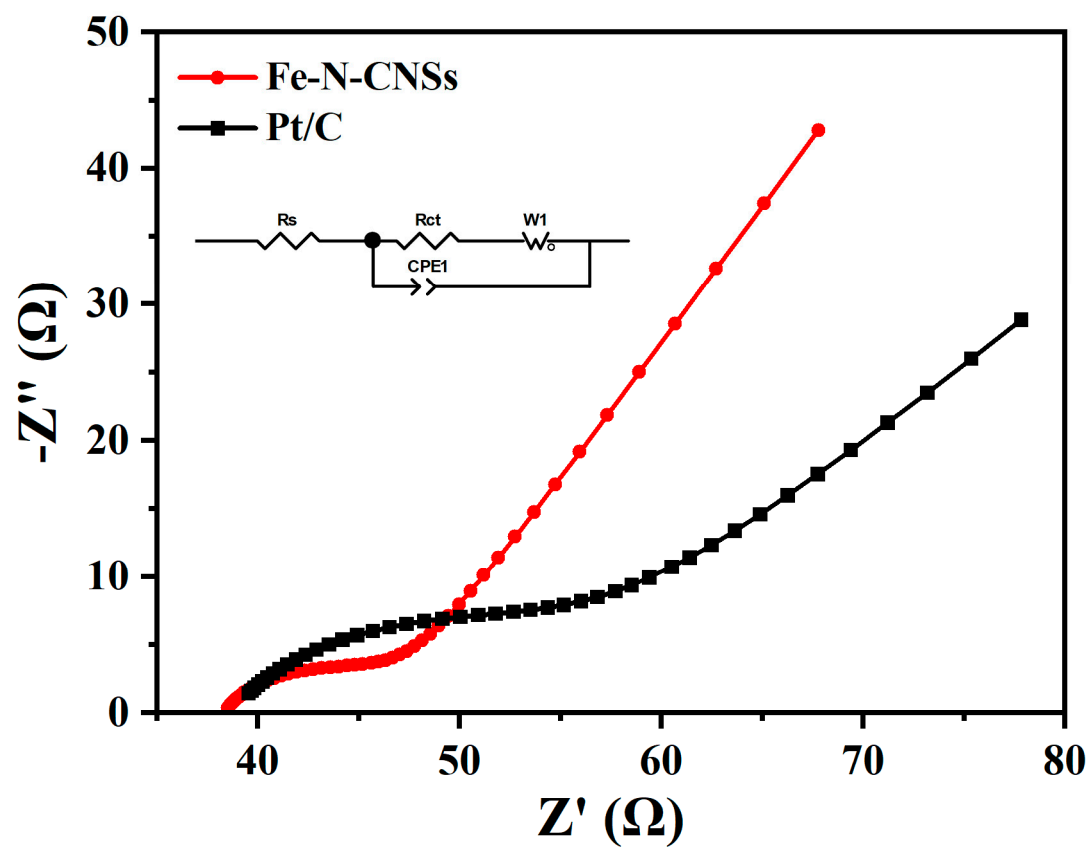**Figure S16.** LSV curves of Fe-N-CNSs-15, Fe-N-CNSs-30, Fe-N-CNSs-60.



**Figure S17.** Nyquist plots of electrochemical impedance spectroscopy (EIS) for Fe-N-CNSs and Pt/C.
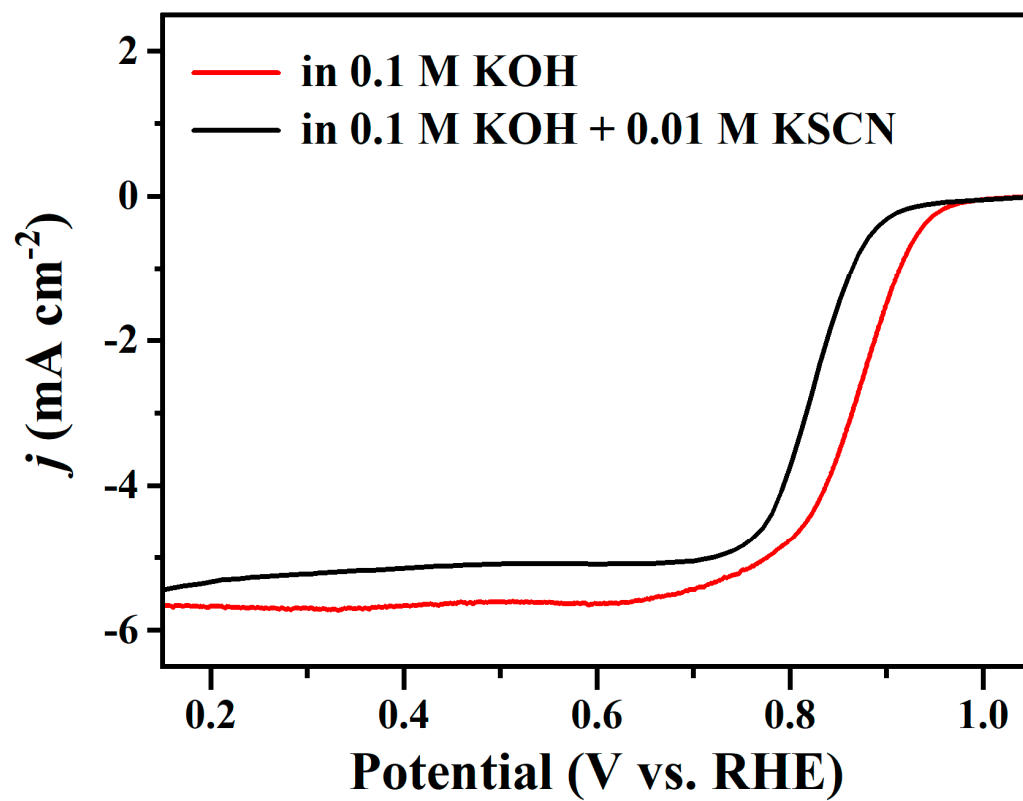
Figure **S18**. Poison experiment by KSCN.