Supporting Information

N, S Co-doped Coal-based Activated Carbon as a High-efficiency and Durable Metal-free Catalyst for the Oxygen Reduction Reaction in Zn-air Batteries

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1. Materials Characterization

Powder X-ray diffraction (XRD) measurements were performed on Bruker D8 Advance diffractometer with Cu K_α radiation (λ=0.15406 nm). The morphologies and the structures of the samples were characterized using field-emission scanning electron microscopy (FESEM; Hitachi SU-4800) and transmission electron microscopy (TEM; JEM-2100F). Raman spectra measurements were conducted using a multichannel modular triple Raman system, with confocal microscopy at room temperature and an excitation wavelength of 532 nm (HR Evlution). The Brunauer-Emmett-Teller (BET) specific surface area was determined by nitrogen adsorption-desorption isotherm measurements at 77 K (ASAP 2020). XPS measurements were performed on an X-ray photoelectron spectrometer (Thermo ESCALAB 250). The superficial functional groups of samples were obtained by Fourier transform infrared (VERTEX 70) spectrophotometer. Evaluation of surface wettability of materials employing DCAT 21.

2. Details supplement

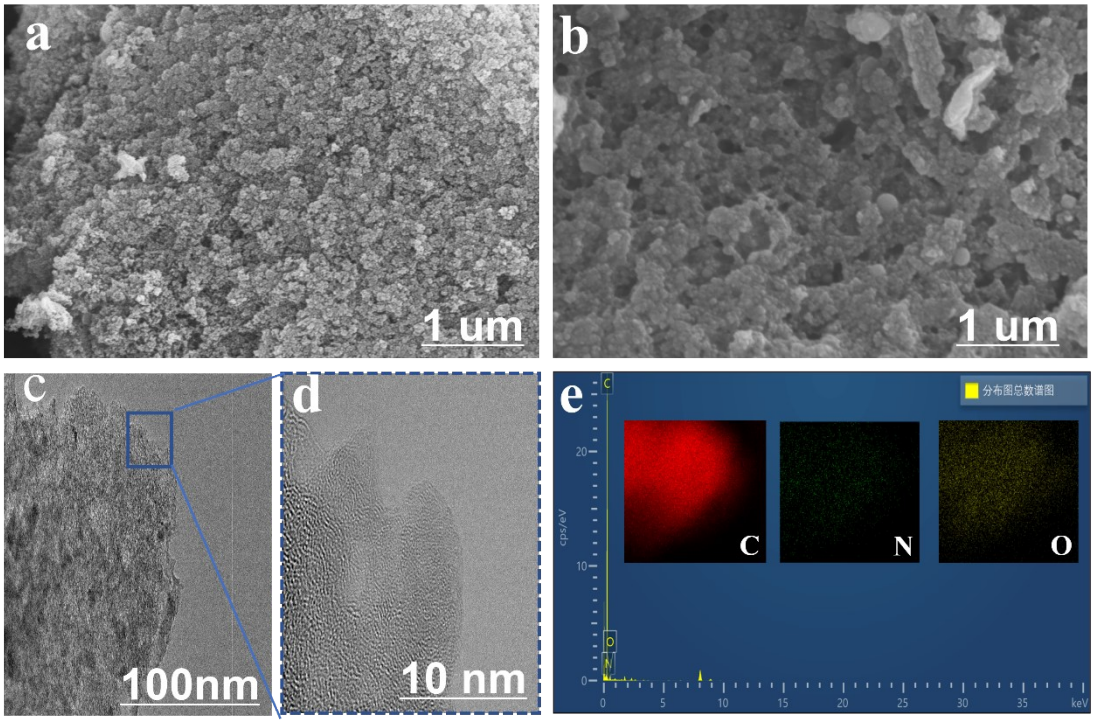

Figure S1: a, b) SEM images of AC and N-AC; c, d) TEM and HRTEM images of N-AC; e) EDS elemental mapping images of N-AC.

Table S1: The samples of BET surface area and pore volume

Samples	BET surface area ($m^2\ g^{-1}$)	pore volume[a] ($cm^3\ g^{-1}$)
AC	1042.5	0.65
N-AC	1146.3	0.71
N,S-AC	1153.9	0.74

[a]: Pore volume is calculated by the Barret-Joyner-Halenda model.

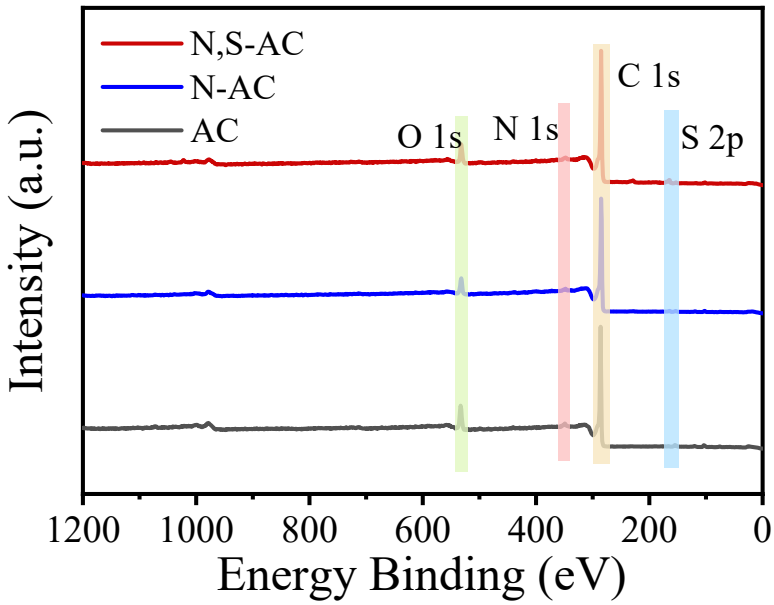

Figure S2: XPS survey spectra of AC; N-AC and N,S-AC.

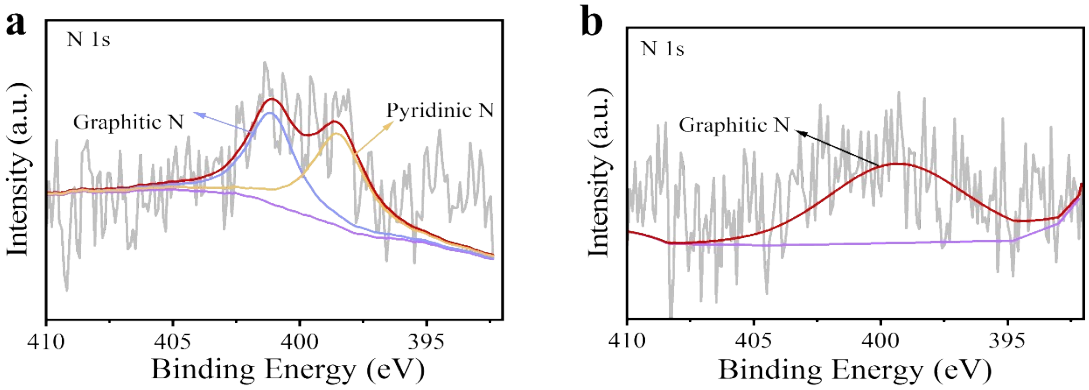

Figure S3: N 1s XPS spectra of (a) N-AC and (b) AC.

Table S2: The atomic percentage of S, N, C, and O elements based on XPS analysis.

	N,S-AC	N-AC	AC
N(at%)	6.58	0.69	0.14
S(at%)	0.65	/	/
C(at%)	82.64	92.80	91.74
O(at%)	10.13	6.51	8.12

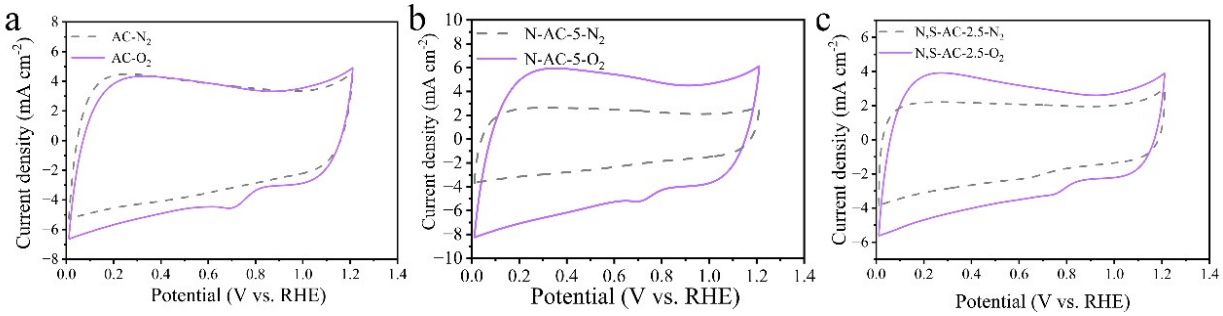

Figure S4: CV curves of a) AC, b) N-AC, and c) N,S-AC in N_2-saturated (dashed line) and O_2-saturated (solid line) 0.1 M KOH solution.

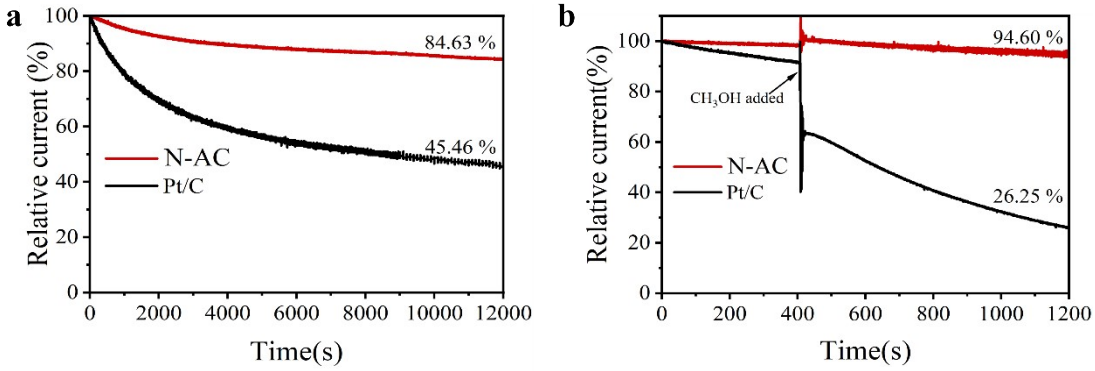

Figure S5: a) I-t test of N-AC and 20 % Pt/C in an O_2-saturated 0.1 M KOH electrolyte; b) Methanol tolerance test of N-AC and 20 % Pt/C catalysts in O_2-saturated 0.1 M KOH electrolyte.

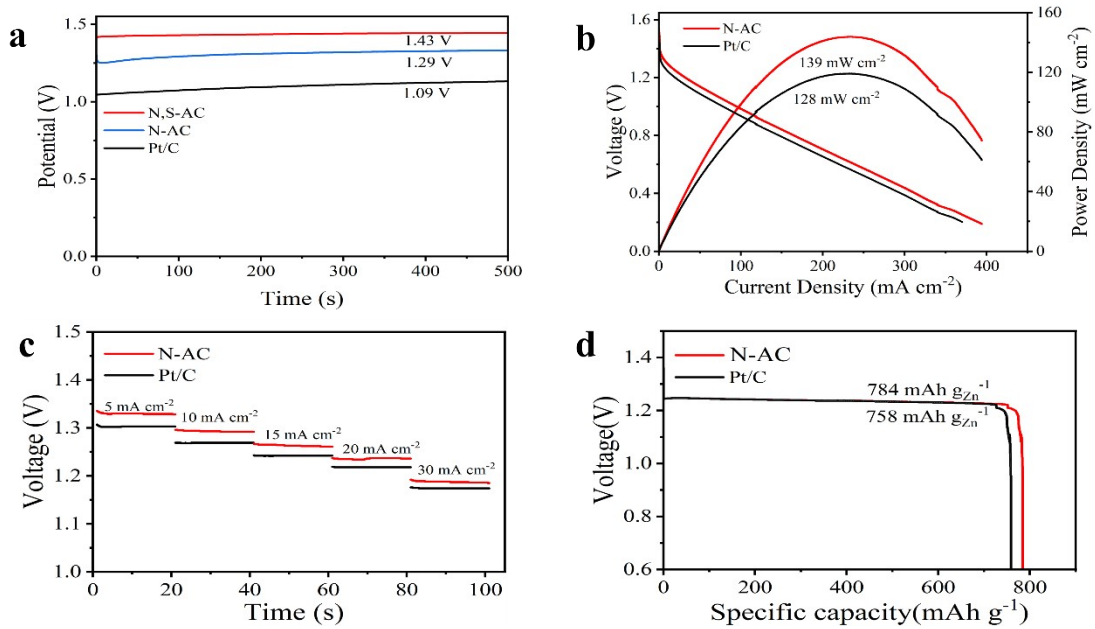

Figure S6: a) Open-circuit voltage plots of the N,S-AC, N-AC, and Pt/C; b) Polarization curves and corresponding power density curves of the N-AC and 20% Pt/C; c) Discharge curves at different current densities from 5 to 30 mA cm^{-2}; d) Voltage-specific capacity curves of N-AC and 20% Pt/C-based ZABs at a current density of 10 mA cm^{-2}.

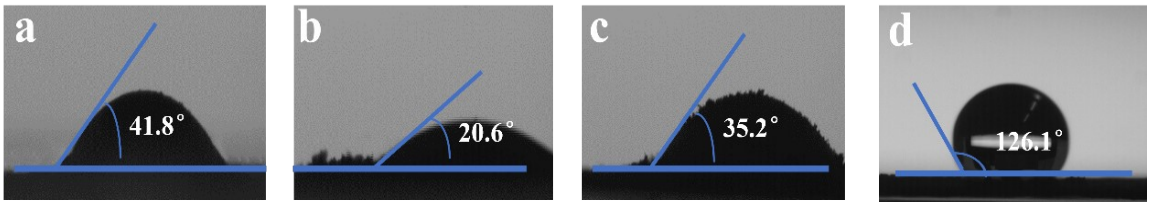

Figure S7: Contact angles for water droplets on pressed pellets of different samples: a) AC; b) N-AC; c) N,S-AC; d)Pt/C.

Table S3: Comparison of electrocatalytic performances between our N, S-AC sample and recently reported electrocatalysts applied in Zn-air battery.

Catalysts	$E_{1/2}$(V)	J_L(mA cm^{-2})	Peak Power Density (mW cm^{-2})	Ref.
N, S-AC	0.79	6.35	142	This Work
N, S@CM-1000	0.76	5.5	90	1
NSG	0.75	4.85	/	2
PSN-G1	0.79	/	/	3
G100-1B	0.77	4.56	/	4
S-POP	0.72	5.10	/	5
N,S-GNR-2s	0.79	5.06	/	6
N-graphene@N-rGO	0.86	/	139	7
PANZ@CNTs75	0.73	4.30	201.9	8
HEO/Co NC	0.65	/	162	9
NF @ CB	0.81	5.26	130	10
CNTs	0.78	4.25	94.8	11
NPNC-2	0.77	/	145	12

References

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