Supporting Information

A Ni single-atom catalyst for Advanced Environmental Disinfection Based on Electrochemical Production of Hydrogen Peroxide

Zhijian Liu^a, Bingqian Zhang^a, Yan Liu^a, Chongbo Wang^a, Chenliang Ye^{a,*}, Weijie Yang^{a,*}

^a School of Energy and Power Engineering, North China Electric Power University Baoding, Hebei 071003, P.R. China

E-mail: chenliangye@ncepu.edu.cn; yangwj@ncepu.edu.cn

Experimental methods

1. Materials. Vulcan XC72 carbon black, anion exchange membrane (FAB-PK-130), hydrophobic carbon paper (TGPH060) were purchased from Materials Science Station. Na fion 117 binder was obtained from Suzhou Shengernuo Technology Co., Ltd. $Fe(NO_3)_3 \cdot 9H_2O_1$ $Ni(NO_3)_2 \cdot 6H_2O_1$ $Co(NO_3)_2 \cdot 6H_2O_1$ $Cu(NO_3)_2 \cdot 3H_2O_1$ 2,5pyridinedicarboxylic acid, potassium titanium oxalate, potassium ferrocyanide, potassium chloride, potassium hydroxide, anhydrous ethanol, PBS buffer solution, isopropanol were purchased from Macklin Reagent. Concentrated sulfuric acid, hydrochloric acid, nitric acid, 2.5 mol L⁻¹ H₂O₂ were purchased from Damao Chemical Reagent Co., Ltd. Agar medium, viscous Serratia marcescens cultured to a concentration of approximately 2x10⁹ cfu mL⁻¹ (colony-forming units). All materials and chemicals were used as received without further purification. Deionized water was used throughout the entire study.

2. Preparation of oxidized carbon black (OCB). 4 g of carbon black (XC-72, Fuel Cell Store) was mixed with 100 mL of concentrated nitric acid (7 M) and heated under reflux at 120 °C for 3 hours in an oil bath. The resulting slurry was cooled to room temperature, centrifuged to collect the solid, washed with ethanol and deionized water until reaching neutral pH, and dried overnight in a vacuum oven (at least 12 hours) before use, yielding oxidized carbon black.

3. Characterization of catalysts. The surface morphology and microstructure of the samples were studied using Scanning Electron Microscopy (SEM) (Thermo Scientific Apreo 2C) and Transmission Electron Microscopy (TEM) (Talos F200S G2). The crystal structure of the samples was determined using X-ray Diffraction (XRD). Fourier Transform Infrared Spectroscopy (FT-IR) measurements were conducted using an instrument (INVENIO R, Bruker Optik GmbH). Raman spectroscopy to assess the degree of disorder in the samples was performed using equipment (Thermo Scientific DXR2xi). Inductively Coupled Plasma - Optical Emission Spectroscopy (ICP-OES) was employed to quantify the content of metal elements in the samples. X-ray Photoelectron Spectroscopy (XPS) was conducted using a scanning X-ray microprobe (ThermoFisher, ESCALAB 250Xi, USA), with calibration performed using the binding

energy (BE) of the C 1s peak at 284.8 eV. Extended X-ray Absorption Fine Structure (EXAFS) measurements were conducted at the Synchrotron Radiation Facility of the Institute of High Energy Physics, Chinese Academy of Sciences. Ni foil was used as a reference for EXAFS data collection. The collected EXAFS data were extracted and processed using Athena in the IFEFFIT software package following standard procedures.

2.4 Measurement of H_2O_2 concentration. In this study, the concentration of H_2O_2 was determined using the potassium titanium oxalate spectrophotometric method. Under acidic conditions, potassium titanium oxalate reacts with H2O2 to form a stable yellowcolored complex. The maximum absorption wavelength is at 400 nm, and the H_2O_2 concentration can be calculated by measuring the solution's absorbance and referencing a standard curve. The specific steps for measuring H₂O₂ concentration are as follows: 0.4 mL of the H₂O₂ sample to be tested, 1 mL of 0.05 mol L⁻¹ potassium titanium oxalate solution, and 3 mol L⁻¹ sulfuric acid solution were added to a disposable centrifuge tube and thoroughly mixed. To ensure sufficient reaction between the sample solution and potassium titanium oxalate solution, the mixed solution was allowed to stand for 10 minutes. The wavelength of the visible spectrophotometer was set to 400 nm, and the absorbance of the solution was measured. The concentration of H₂O₂ in the sample was calculated based on the standard curve relating H₂O₂ concentration to absorbance. Additionally, a series of known concentrations of standard H₂O₂ solutions were prepared and their absorbance measured using the same method as described above to establish a standard curve correlating H₂O₂ concentration with absorbance (Fig. S4).



Fig. S1 (a) Linear sweep voltammetry was performed on a rotating ring-disk electrode (RRDE) immersed in a solution containing 0.01 M potassium ferricyanide and 0.1 M KCl at various rotation speeds. The scan rate is set at 10 mV s⁻¹. (b) The limiting diffusion current of the ring electrode and the disk electrode at different rotation speeds 0were fitted using linear regression. The experimental determination yielded a collection efficiency N of 36.9%, which closely approximates the theoretical value of 37%.



Fig. S2 (a-g) Cyclic voltammetry (CV) curves of 3%Ni-N₄-O, 5%Ni-N₄-O, 10%Ni-N₄-O, Fe-N₄-O, Co-N₄-O, and Cu-N₄-O in 0.1 M KOH solution at different scan rates (10 to 120 mV s⁻¹). (h) Corresponding capacitance current plots as a function of scan rate for the respective catalysts.



Fig. S3 Cyclic voltammetry (CV) tests of different catalysts in 0.1 M KOH saturated with N_2 and O_2 (scan rate: 100 mV s⁻¹).



Fig. S4 Standard curve of H_2O_2 concentration versus absorbance. A series of known concentrations of H_2O_2 solutions were prepared. To 0.4 mL and 1 mL of these solutions, 0.05 mol L⁻¹ potassium titanium oxalate and 3 mol L⁻¹ concentrated sulfuric acid were added, thoroughly mixed, and absorbance spectra were obtained.



Fig.S5 Volcano plots of (a)2e⁻ ORR and (b)4e⁻ ORR for Ni-N₄-O and Ni-N₄-OH configurations.



Fig.S6 Volcano plots of (a) 2e⁻ORR and (b) 4e⁻ORR for the M(Fe, Co, Ni, Cu)-N₄-O configuration.



Fig.S7 Liquid State Voltammetry (LSV) curves for different Ni elemental content.



Fig. S8 The electron transfer numbers for 3%, 5%, and 10% Ni-N₄-O.



Fig. S9 LSV curves, H_2O_2 selectivity, and electron transfer numbers for Fe, Co, Ni, Cu, and OCB in 0.1 M KOH.



Fig. S10 Morphological characteristics of Ni-OCB. (a, b, c, d) SEM images of Ni-OCB at different magnifications.



Fig. S11 Morphological characteristics of Ni-OCB. TEM images of Ni-OCB at different magnifications.



Fig. S12 XRD patterns of OCB, Fe-N₄-O, Co-N₄-O, Ni-N₄-O, Cu-N₄-O.



Fig. S13 Raman spectra of OCB, Fe-N₄-O, Co-N₄-O, Ni-N₄-O, Cu-N₄-O.



Fig. S14 FT-IR spectra of OCB and Ni-N₄-O.



Fig. S15 (a) XPS survey spectra of Ni-N₄-O, showing the absence of Ni peak. (b) Ni 2p (c) C 1s (d) N 1s and (e) O 1s spectrum of Ni-N₄-O.



Fig. S16 Ni L_3 edge EXAFS fitting of Ni foil.



Fig. S17 (a) Schematic demonstration of the H-type electrolytic cell. (b) The ultraviolet–visible–near-infrared absorption spectroscopic method.



Fig. S18 Polarization curves of hydrophobic carbon paper and Ni-OCB in an H-type electrolysis cell.



Fig. S19 Diagram of the experimental setup for disinfection and sterilization in a realworld scenario: one biological aerosol generator, one nebulizer, and four Andersen six-stage samplers.

Sample	Constant	Test Test the elemental Dilution		Dilution	Elemental
weight	Volume(mL)	element	concentration of	factor	content of
(g)			the solution	f	the sample
			(mg/L)		(mg/kg)
0.01576	10	Ni	3.7823	1	1.18%

Table. S1 ICP-OES analysis of Ni-N₄-O

Sample	Cdl $(mF cm^{-2})$	ECSA(cm ² cm ⁻² electrode)
OCB	3.47	86.75
3%Ni-N ₄ -O	4.87	121.75
5%Ni-N ₄ -O	4.42	110.5
10%Ni-N ₄ -O	3.92	98
Fe-N ₄ -O	3.16	79
Co-N ₄ -O	3.25	81.25
Cu-N ₄ -O	1.76	44

Table. S2 Capacitive double-layer capacitance (Cdl) and corresponding electrochemically active surface area (ECSA) of OCB, 3% Ni-N₄-O, 5% Ni-N4-O, and 10% Ni-N4-O.

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samples	$I_{\rm D}/I_{\rm G}$	Onest potential E (0.1mA cm ⁻²)	Diffusion-limitting current density/j (mA cm ⁻² @0.2V)	H ₂ O ₂ % (@0.6V)
Fe-N ₄ -O	1.007	0.857	4.447	46
Co-N ₄ -O	1.01	0.874	3.076	21
Ni-N ₄ -O	1.04	0.85	2.953	91
Cu-N ₄ -O	1.03	0.882	4.102	29
OCB	1.009	0.877	2.956	58

Table. S3 Comparison of Raman and electrochemical performance (including onsetpotential, diffusion-limiting current density, and H_2O_2 selectivity) of catalyst.

Table. S4 The content percentages (%) from the decomposed N 1s spectra of the Ni-OCB.

Sample	pyridinic-N	Ni-Nx	pyrrolic-N	graphitic-N	Oxidized-N
Ni-N ₄ -O	17.2	15	27.7	20.5	19.5

Sample	Scattering path	Distance (Å)	C.N.	σ ² (Å ²)	$\Delta \mathrm{E}_{0}\left(\mathrm{eV} ight)$	R-factor
Ni foil	Ni-Ni	2.48	12	0.006	-4.6	0.005
Ni sample	Ni-N	2.04	3.8	0.005	-6.7	0.002
	Ni-O	2.11	1.1	0.005	-6.7	0.002

Table. S5 Fitting results of the sample

Calatyst	Electrolyte	H ₂ O ₂ productivity (mmol g _{cat} ⁻¹ h ⁻¹)	Reference
Co-NC	0.1M HClO ₄	275	[1]
oxo-G/NH ₃ H ₂ O	0.1M KOH	225	[2]
F-mrGo	0.1M KOH	431	[3]
PCMNS	0.1M K ₂ SO4	1103	[4]
Co _{SA} -N-CNTS	0.5M H ₂ SO4	974	[5]
Ni-OCB	0.1M KOH	98.5	[6]
Co-N-KB	0.1M HClO ₄	100	[7]
CoSe ₂ NS/CC	$0.05M H_2SO_4$	65.74	[8]
CoS_2	$0.05M H_2SO_4$	148	[9]
N-GA	$0.1M H_2SO_4$	107.8	[10]
Ni MOF NSs	0.1M KOH	80	[11]
Pt/HSC	1M HClO4	48.75	[12]
Co_1 -NG(O)	0.1M KOH	418	[13]
Co-POC-O	0.1M KOH	478	[14]
Ni-N ₄ -O	0.1M PBS	316.8	This Work

Table. S6 The reported yield rate of H_2O_2

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