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Electronic Supplementary Information

Cobalt coordinated 4, 4'-bipyridine derived ordered mesoporous N doped carbon as efficient electrocatalyst for oxygen reduction

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Preparation of working electrodes

For electrochemical measurements, a glassy carbon electrode (GCE, Φ 5 mm) was used as the working electrode substrate. Before every measurement, the GCE was cleaned by ethanol in an ultrasonic bath, polished with α-Al_2O_3 slurry (50 nm) on a micro cloth, and rinsed with DI water.

For catalysts ink preparation, 5.0 mg catalyst and 1 mL Nafion ethanol solution (0.25 wt%) were thoroughly mixed under ultrasound for about 30 min. And then, 20 μL slurry was coated onto the GCE and dried naturally. The catalyst loading was calculated to be 0.5 mg cm^{-2}. For comparisons, a commercial Pt/C (20 wt%, Johnson Matthey Corp.) catalyst was also used for ORR.

Preparation of air electrode

For air electrode preparation, a Toray carbon paper (TGP-H-060) was used as the substrate. A diffusion layer was formed by painting the suspension of carbon black (XR-72R) and polytetrafluoroethylene (PTFE) onto one side of the carbon paper (the mass ratio of carbon black and PTFE is 3:2).

For catalyst layer fabrication, a catalyst ink was first prepared through the same procedure described in the working electrode preparation. The ink obtained was painted onto the other side of carbon paper and dried under an infrared lamp. The catalyst loading for NC-S900 is calculated to be 1.0 mg cm^{-2}. For comparisons, Pt/C was also used to fabricate an air electrode through the same procedures.

Characterization

The scanning electron microscopy (SEM) images were obtained on a JSM-7100F field emission scanning electron microscope (JEOL, Japan), with an acceleration voltage of 5 kV. The transmission electron microscopy (TEM) was operated on a JEM-2100 transmission electron microscope (JEOL, Japan) at an acceleration voltage of 200 kV. The X-ray photoelectron spectroscopy (XPS) was conducted on an ESCALAB 250 X-ray photoelectron spectrometer (Thermo-VG Scientific, USA). The X-ray difraction (XRD) is conducted on a TD-3500 powder difractometer (Tongda, China) at a scan rate of $5°min^{-1}$. The

Co content in various catalysts were measured by using a PerkinElmer Optima 8300 inductively coupled plasma emission spectrometer (ICP-AES).

Electrochemical measurements

Electrochemical measurements were carried out at room temperature by using a three-electrode glass cell on an Interface 1010B electrochemical workstation (Gamry, USA), coupled with a rotating ring-disk electrode (RRDE) system (PINE, USA). An Hg/HgO/NaOH (1M) and graphite stick was used as the reference and counter electrodes, respectively. All the potentials initially measured versus Hg/HgO/NaOH (1M) were converted to the ones versus reversible hydrogen electrode (RHE) by adding 0.92 V.

Before every measurement, the KOH solution (0.1 M) was saturated with O_2 (99.999%) for at least 30 min. All the current densities were normalized to the GCE's geometric area (0.1964 cm^2).

The linear sweep voltammetry (LSV) measurements were conducted at a scan rate of 5 mV s^{-1} in an O_2 or N_2 saturated 0.1 M KOH solution.

The rotating ring-disk electrode (RRDE) measurements were conducted by using a glassy carbon disk with a Pt ring which was biased at 1.42 V (*vs.* RHE). The peroxide yields and the electron transfer number (n) per oxygen molecule were calculated based on the following equations[1]:

$$\eta = 200 I_r (\mathrm{N} I_d + I_r)^{-1}$$

$$n = 4 I_d (I_d + I_r \mathrm{N}^{-1})^{-1}$$

where I_r and I_d refer to the ring and disk currents, respectively, and N is the collection efficiency, which was confirmed to be 0.36 by the reduction of $K_3Fe(CN)_6$.

The turnover frequency (TOF) was calculated by the equation[2]:

$$\mathrm{TOF} = \mathrm{JS}/(4\mathrm{Fn})$$

J (mA cm^{-2}) is the measured current density at 1.5 V (vs. RHE); F is Faraday constant; S (0.1964 cm^{2}) is the surface area of glassy carbon electrode; n is the molar number of Co in the catalyst, calculated based on the ICP-AES results.

The electrochemical impedance spectra (EIS) were recorded at open circle potential with an amplitude of 10 mV within the frequency range of 10^{-1} to 10^{5} Hz.

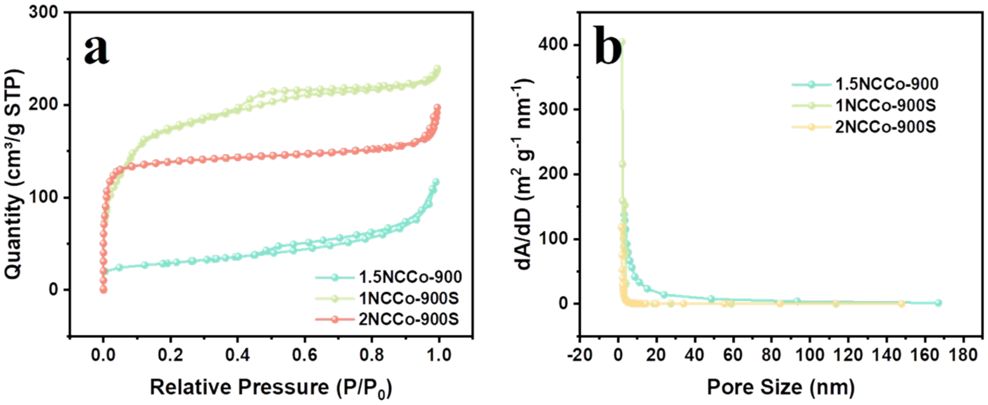

Fig. S1 (a) N_2 adsorption-desorption isotherms of different catalysts; (b) pore-size distribution of obtained catalysts.

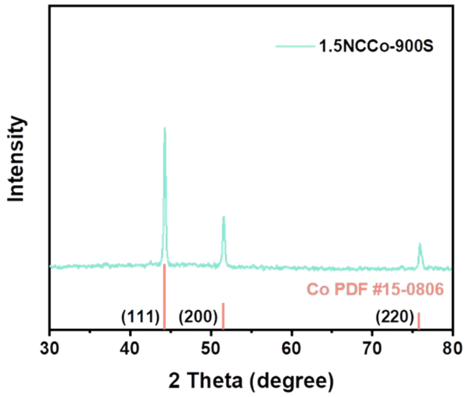

Fig. S2 XRD pattern of 1.5NCCo-900S

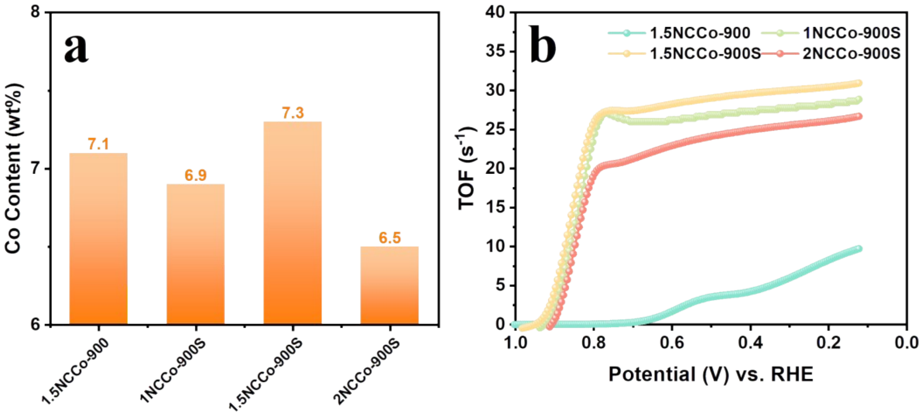

Fig. S3 (a) Co contents in various catalyst, based on the ICP-AES measurements. (b) time of turnover (TOF) for the obtained catalysts, based on the LSV and ICP-AES results.

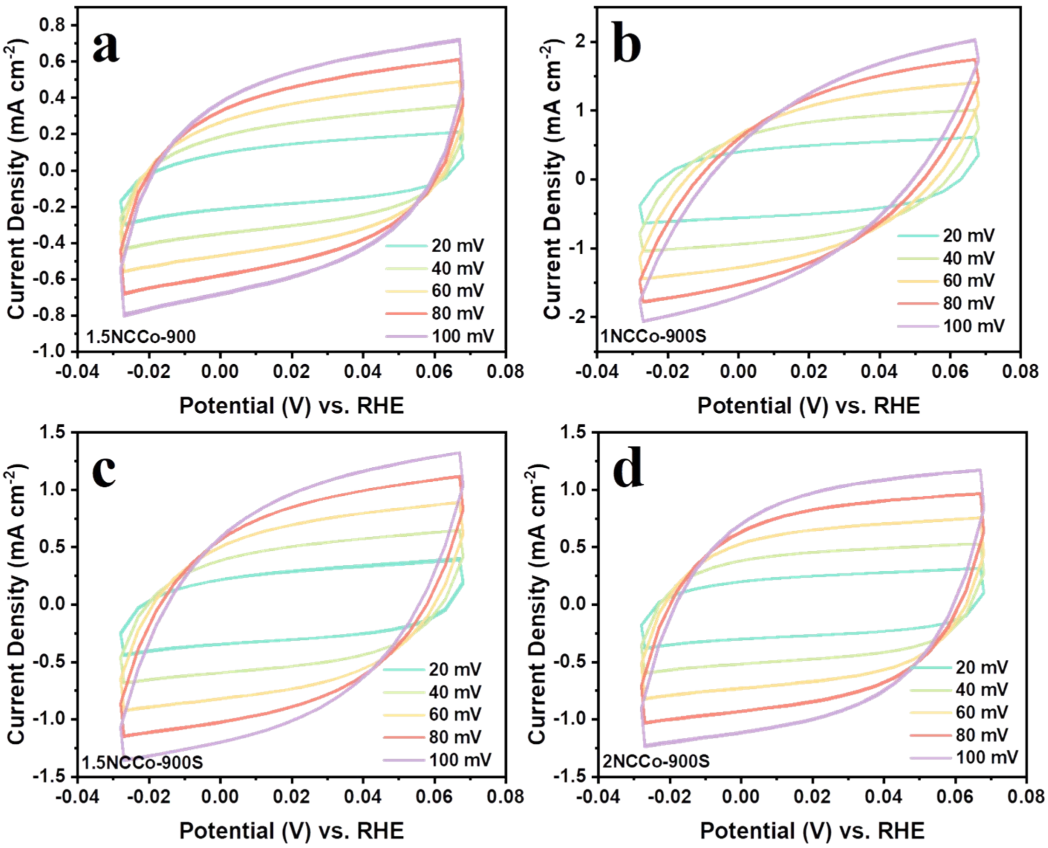

Fig. S4 CV curves under different scanning rates: (a) 1.5NCCo-900; (b) 1NCCo-900S; (c) 1.5NCCo-900S; (d) 2NCCo-900S;

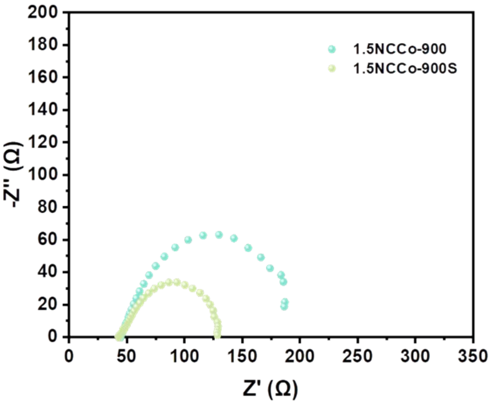

Fig. S5 Nyquist plots of 1.5NCCo-900 and 1.5NCCo-900S

Table S1 Atomic compositions of different catalysts (at%), derived from the XPS results.

	1NCCo-900S	1.5NCCo-900S	2NCCo-900S	1.5NCCo-900
C1s	94.56	93.8	91.72	90.56
O1s	2.1	2.91	3.7	4.31
N1s	2.81	2.89	4.3	4.59
Co2p	0.53	0.41	0.29	0.54

Table S2 comparison of CNSCo-600 and ORR catalysts recently reported

No.	Catalyst	Electrolyte	$E_{1/2}$	Reference electrode	Reference
1	Fe SAC-MOF-5	0.1 M KOH	0.83	RHE	[2]
2	$Fe_{0.5}Co$@HOMNCP	0.1 M KOH	0.903	RHE	[3]
3	Se@NC-1000	0.1 M KOH	0.85	RHE	[4]
4	Fe-ACSA@NC	0.1 M KOH	0.90	RHE	[5]
5	O-Zr-N-C	0.1 M KOH	0.90	RHE	[6]
6	Cu-N-C/GC	0.1 M KOH	0.84	RHE	[7]
7	Azo-COF	0.1 M KOH	0.68	RHE	[8]
8	FeNC-CVD-750	0.1 M KOH	0.85	RHE	[9]
9	Fe_3C@NCNTs	0.1 M KOH	0.84	RHE	[10]
10	meso-Fe–N–C	0.1 M KOH	0.846	RHE	[11]
	1.5NCCo-900S	**0.1 M KOH**	**0.87**	**RHE**	**Our work**

Table S3 The Rp. R and Rs. R values derived from the EIS results

	Rp. R (Ω)	Rs. R (Ω)
1.5NCCo-900	154.19	44.09
1.5NCCo-900S	90.82	42.74

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