Supporting Information

Ligand-regulated Ni-based coordination compounds to promote self-reconstruction for improved oxygen evolution reaction

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S1. Characterizations

X-ray diffraction (XRD) patterns were conducted on a Bruker D8 Focus equipped with nickelfiltered Cu K α radiation (k = 1.54056 Å) at a scan rate of 5° min⁻¹. Scanning electron microscopy (SEM) was characterized using an Apreo S LoVac field emission scanning electron microscope. Transmission electron microscopy (TEM), high-resolution TEM (HRTEM), and energy dispersive X-ray spectroscopy (EDX) were conducted on field emission transmission electron microscope JEOL JEM-F200. X-ray photoelectron spectroscopy (XPS) measurements were conducted on an ESCALAB250xi electron spectrometer using Al K α source as a radiation source, where the adventitious carbon 1s peak at 284.8 eV was taken as an internal standard. Raman spectra were obtained on Confocal Raman Microscopy (LabRAM HR Evolution) with an excitation wavelength of 532 nm. Fourier transform infrared (FTIR) spectroscopy was constructed on a Bruker VERTEX70 spectrometer.

S2. Electrochemical measurements

All the electrochemical experiments were performed using an IVIUMSTAT workstation (Ivium Technologies BV, Netherlands). A standard three-electrode cell, consisting of a glassy carbon rotating disk electrode (RDE) (diameter of 5 mm, geometric surface area of 0.196 cm², Pine Research Instrumentation) deposited with catalysts as the working electrode, a graphite rod as the auxiliary electrode, a saturated Hg/HgO as the reference electrode was used for all electrochemical experiments. The potentials were converted to the reversible hydrogen electrode (RHE) according to Eq. (1).¹

$$E_{\rm RHE} = E_{\rm Hg/HgO} + 0.059 \times pH + 0.098 \tag{1}$$

Cyclic voltammetry (CV) and linear sweep voltammetry (LSV) curves were measured at scan rate of 10 mV·s⁻¹ and 5 mV·s⁻¹ respectively, in 100 mL of 1.0 M KOH aqueous solution (pH = 13.6).

The LSV calculations were done with 100% iR correction. The diagrams of the Tafel slope were obtained by plotting the relationships between overpotential and the logarithmic current density. The electrochemical impedance spectroscopy (EIS) was conducted under 1.50 V (vs RHE) from 0.1 Hz to 100 KHz (infinitesimal disturbance of 5 mV) to obtain the solution resistance (Rs) for iR compensation and the Nyquist plots. Moreover, to calculate the electrochemically active surface area (ECSA), the double layer capacitance (C_{dl}) was determined by plotting a graph of the difference in current density between the anodic and cathodic sweeps at 0.15-0.25 (V vs Hg/HgO) (non-Faradaic region) against the scan rate. The slope of this linear line corresponds to twice the value of C_{dl} . Subsequently, the ECSA was derived from C_{dl} using Eq. (2).²

$$ECSA = \frac{C_{dl}}{C_s}$$
(2)

Where, C_{dl} is double layer capacitance and C_s is specific capacitance, we have used the value of 0.04 mV·cm^{-2.2} Moreover, the turn over frequency (TOF) was calculated by using Eq. (3).³

$$TOF = \frac{j \times A}{4 \times F \times n} \tag{3}$$

Where j (mA·cm⁻²) is the current density at a particular overpotential, A is the area of the working electrode, F is the Faraday constant (96,500 C·mol⁻¹) and n is the number of moles of the active materials. Notably, the working electrode decorated with a thin catalyst film was obtained as follows: drop-casting 10 μ L catalyst ink evenly in four drops, covering a 10 mL beaker, and then drying at room temperature. The mass loading of catalysts on the working electrode was 0.25 mg·cm⁻². The catalyst ink was prepared by dispersing 5 mg catalyst and 1 mg carbon black powders in 1 mL water-ethanol solution (3:1 by volume) by adequate ultrasonic dispersion for 2 h and adding 30 μ L 5 wt.% Nafion solution followed by another 2 h ultrasonic dispersion. The stability test was performed by pasting 10 μ L of catalyst ink on 0.5 × 0.5 cm nickel foam.



Figure S1. (a) Synthesis of Ni(HNCN)₂. (b) Preparation of Ni(NCN) and Ni(N(CN)₂)₂.



Figure S2. SEM image of Ni(NCN).



Figure S3. (a, b) HRTEM and EDX images of Ni(NCN) respectively. (c) EDX images of Ni(HNCN)₂.



Figure S4. Distorted octahedra of (a) $Ni(HNCN)_2$, (b) Ni(NCN) and (c) $Ni(N(CN)_2)_2$ respectively.



Figure S5. (a, b) N 1s and C 1s spectra of Ni(N(CN)₂)₂, Ni(NCN) and Ni(HNCN)₂ respectively.



Figure S6. FTIR spectra of Ni(HNCN)₂ before and after OER.



Figure S7. Self-reconstruction behavior of N(OOH)-DC during OER.



Figure S8. N₂ adsorption-desorption curves of Ni(N(CN)₂)₂, Ni(NCN) and Ni(HNCN)₂.



Figure S9. C_{dl} and ECSA data (a, b, c) CV curves of Ni(OOH)-DC, Ni(OOH)-CN, and Ni(OOH)-HC respectively, in the non-Faradaic potential region at scan rates from 10 to 100 mV·s⁻¹. (d) C_{dl} measurements of Ni(OOH)-DC, Ni(OOH)-CN, and Ni(OOH)-HC.



Figure S10. (a, b) Intrinsic OER activities of Ni(OOH)-DC, Ni(OOH)-CN and Ni(OOH)-HC normalized by BET and ECSA respectively.



Figure S11. TOF values of Ni(OOH)-DC, Ni(OOH)-CN, Ni(OOH)-HC and IrO₂.



Figure S12. Raman spectra of Ni(N(CN)₂ before and after 100 h stability test.

Catalyst	Ni ²⁺ (%age)		Ni ³⁺ (%age)	
	Before	After	After	
Ni(OOH)-DC	100	0	100	
Ni(OOH)-CN	100	39.82	60.17	
Ni(OOH)-HC	100	55.31	44.68	

 Table S1. Before and after OER percentages of Ni in different catalysts

Electrocatalyst	Substrate	Electrolyte	J	Overpotential	Ref.
			(mA·cm ⁻²)	(mV)	
Ni-MOF	СР	1.0 M KOH	10	346	4
NiCo ₂ O ₄ @CC	GCE	1.0 M KOH	10	340	5
Ni-Co PBA nanocages	GCE	1.0 M KOH	10	380	6
Fe-Ni/NC	GCE	1.0 M KOH	10	350	7
FeNi-substituted ZIF-8	GCE	1.0 M KOH	10	300	8
$Ni_2C_2O_4$	GCE	1.0 M KOH	10	270	9
Ni-bipy-MWNT	GCE	1.0 M KOH	10	290	10
NiFeP/MXene	GCE	1.0 M KOH	10	286	11
Fe/NieCoTe@NCFs	-	1.0 M KOH	10	287	12
NiFe-LDH-0.4M	GCE	1.0 M KOH	10	280	13
Ni(OOH)-DC	GCE	1.0 M KOH	10	264	This work

Table S2. OER activity comparison of different Ni-based catalysts

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