Electronic Supplementary Material (ESI) for Energy Advances.

Supporting Information

Atomically Dispersed Co/Ni Dual Sites Embedded in Nitrogen-Doped Graphene for Boosting Oxygen Evolution

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Materials characterization

The morphologies of the samples as-synthesized were examined by a transmission electron microscopy (TEM, TecnaiG220, FEI), and an aberration-corrected high-angle annular dark-field scanning transmission electron microscopy (AC-HAADF-STEM, JEM-ARM300F, 300 kV, Japan). X-ray diffractions (XRD) were performed on a X'Pert-Pro MPD diffractometer (Netherlands PANalytical) with a Cu Kα X-ray source (λ = 1.540598 Å). X-ray photoelectron spectroscopy (XPS, Escalab250Xi, UK) was conducted with a hemispherical electron energy analyzer. Raman tests were performed on a Bruker RAM II microscoped system at 532 nm. The N_2 adsorption/desorption isotherms of samples were investigated using a gas adsorption analyzer (Jingwei Gaobo JWBK200B, China).

Electrochemical measurement

Electrochemical measurements were performed by a CHI760E electrochemical workstation (Chenhua, Shanghai) using a three-electrode cell. A graphite rod and a Ag/AgCl (KCl saturated) electrode served as the counter electrode and the reference electrode, respectively. The working electrode was a glass carbon (GC) disk (5 mm in diameter, 0.196 cm^2 in area) electrode. The catalyst ink was prepared by ultrasonically mixing 5 mg of a catalyst and 5 mg of carbon powder with 970 μL of isopropanol and 30 μL of Nafion (0.5 wt.%) for 1 h. After that, 21 μL of the ink was deposited on the GC electrode dried at room temperature. Potentials were referenced to a reversible hydrogen electrode (RHE): $E_{\mathrm{RHE}} = E_{\mathrm{Ag/AgCl}} + 0.197 + 0.059 \times \mathrm{pH}$. Polarization curves were plotted by linear sweep voltammetry (LSV) at a scan rate of 5 mV s^{-1} and corrected by *iR* compensation. All electrochemical tests were performed in 1.0 M KOH at room temperature.

The EIS measurements were performed at an applied potential with the scanning frequency values ranging from 10^6 Hz to 0.1 Hz, the EIS data were then fitted by an appropriate equivalent circuit through Zview software to obtain the resistance values.

Electrochemically active surface area (ECSA) measurements. Based on literature, the electrochemically active surface area (ECSA) could be estimated from the electrochemical double layer capacitance (C_{dl}) of the catalytic surface. The C_{dl} can be determined by cyclic voltammetry in a non-faradaic potential range at various scan rates; capacitive current I_{dl} (A cm^{-2}) is directly proportional to the scan rate ν (mV s^{-1}), as indicated by the following equation:

$$C_{\mathrm{dl}} = I_{\mathrm{dl}}/\nu \ (\mathrm{mF\ cm^{-2}})$$

Accordingly, a series of CV measurements were conducted with 10 mV s^{-1}, 20 mV s^{-1}, 40 mV s^{-1}, 60 mV s^{-1}, 80 mV s^{-1}, in the range of -0.1 to 0.1 V for OER in alkaline solution. The C_{dl} value can be obtained by plotting the difference in current density (*J*) between the anodic and cathodic sweeps (J_{anodic}- $J_{cathodic}$) at the middle potential versus scan rate, and the linear slope is twice that of the double-layer capacitance C_{dl}.

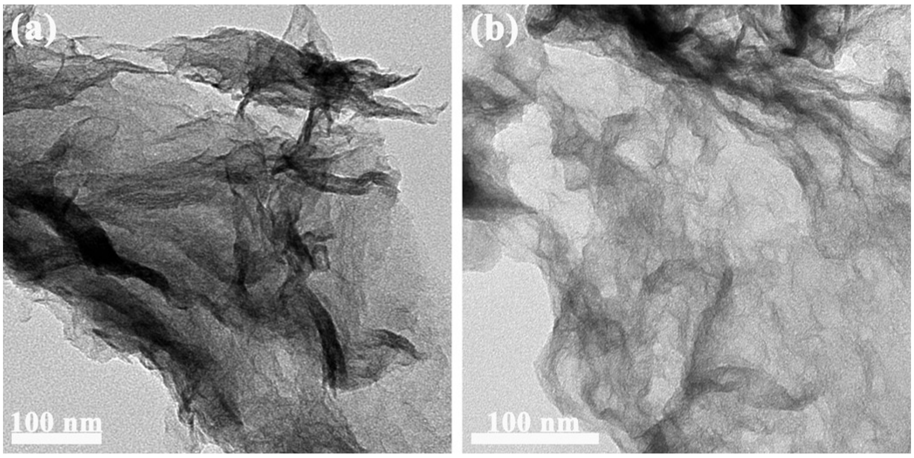

Figure S1 TEM images of (a) Co-SA/NG and (b) Ni-SA/NG.

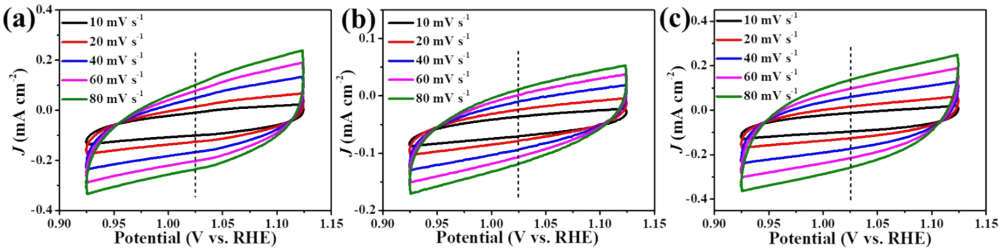

Figure S2 CV measurements of (a) Co-SA/NG, (b) Ni-SA/NG, and (c) CoNi-DSA/NG in 1.0 M KOH solution at different scan rates from 10 to 80 mV s^{-1}.

Table S1. The compositions of CoNi-DSA/NG, Co-SA/NG and Ni-SA/NG obtained by ICP-OES.

Sample	Co (wt%)	Ni (wt%)
CoNi-DSA/NG	1.09	1.80
Co-SA/NG	1.23	/
Ni-SA/NG	/	1.66

Table S2. Comparison of OER performance of CoNi-DSA/NG catalyst with recently reported studies

Material	**Electrolyte (KOH)**	**η_{10} (mV)**	**Ref.**
$Ni_{0.65}Fe_{0.35}$@PCN	1.0 M	310	[S1]
NiFe DG	1.0 M	358	[S2]
Ni_7Fe_3-CNG	1.0 M	270	[S3]
CoNi-SAs/NC	1.0 M	340	[S4]
Ru/Co-N-C	1.0 M	247	[S5]
a-NiCo/NC	1.0 M	252	[S6]
IrO_2	1.0 M	330	[S7]
Co-SA/NG	1.0 M	275	This work
Ni-SA/NG	1.0 M	377	This work
CoNi-DAS/NG	1.0 M	214	This work

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