Supplementary Materials

Flower-like Co3Ni1B nanosheets based on reduced graphene oxide (rGO) as efficient electrocatalyst for oxygen evolution reaction

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Experimental section.

1. Materials

Cobalt (II) nitrate hexahydrate [Co(NO$_3$)$_2$·6H$_2$O], nickel (II) nitrate nonahydrate [Ni(NO$_3$)$_2$·6H$_2$O], sodium borohydride (NaBH$_4$), Sulfuric acid (H$_2$SO$_4$), phosphoric acid (H$_3$PO$_4$), potassium permanganate (KMnO$_4$), hydrogen peroxide (H$_2$O$_2$, 30 wt.%), hydrochloric acid (HCl) and potassium hydroxide (KOH) were purchased from Sinopharm Chemical Reagent Co., Ltd. Commercial RuO$_2$ (99.9%) was purchased from Aladdin Ltd. All reagents were purchased and used as received without any further purification.

2. Characterization

The overall morphology of the as-prepared catalysts was characterized by Field-Emission Scanning Electron Microscope (FESEM, JEOL JSM6510LV) and Transmission Electron Microscope (TEM, Hitachi, HITACHI H-8100). The EDS with JEOL JSM6510LV (15 kV) was used to measure the elemental distributions and compositions of optimal material. The phase and crystal structure were researched by the powder X-ray diffraction (PXRD, D/MAX UItimate IV) with Cu Kα radiation (λ = 0.15418 nm). The surface chemical properties of as-prepared material, including elemental compositions, elemental chemical states and electron valence states were inspected by X-ray Photoelectron Spectroscopy (XPS, Thermo Scientific K-Alpha USA). The carbon structure in the as-prepared materials was identified semi-quantitatively via plus laser Raman spectrometer. An automatic volumetric sorption apparatus (AutoSor-IQ-MP) was used to measure the specific surface area of the as-

prepared material.

3.   Electrochemical measurements

All electrochemical characterizations were tested on a CHI660E Electrochemical Workstation (CHI 660E, CH Instruments, Inc. Shanghai). OER performance was tested in 1.0 M KOH using a standard three-electrode system with platinum foil, Hg/HgO electrode and glassy carbon electrode (GCE, 3 mm of diam) as counter electrodes, the reference and working electrodes, respectively. First and foremost, 5.00 mg of the as-prepared catalysts were distributed in the mixed solvent containing 580 µL deionized water, 400 µL ethanol and 20 µL of 5 wt.% Nafion solution, respectively, and sonicated until a homogeneous ink was formed. Furthermore, 5 µL of the ink was dropped on the GCE to obtain a working electrode with a total catalyst loading of ~0.36 mg cm^{-2}.

In this work, all tests were carried out in an electrolyte of 1.0 M KOH (pH = 14), and the electrolyte was saturated with high-purity O_2 for 30 minutes continuously. Magnetic stirring system was used to throw away bubbles on the electrode surface during the test. All measured potential curves in the work were converted to reversible hydrogen potential (RHE) according to this following formula: E(RHE) = E(Hg/HgO) + 0.059pH + 0.098[1] and corrected for 95% iR to compensate for the loss of ohmic potential[2]. Cyclic voltammetry (CVs) tests from 0 V to 1.0 V with a scan rate of 50 mV s^{-1} were used to activate and stabilize the working electrode. Polarization curves and Tafel plots were recorded using linear sweep voltammetry (LSV, 0-0.8 V) at a scan rate of 5 mV s^{-1}. The OER kinetics of the prepared catalysts were evaluated according to the Tafel equation: $\eta = \log j + b$[3] (where η is the overpotential, j is the electric current

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density, and b is the Tafel slope). The CV curves at different scan speeds (20-100 mV s^{-1}) were tested in the range of 0.08-0.25 V (vs RHE) to obtain the electric double-layer capacitance (C_{dl}) and electrochemically active specific surface area (ECSA). Electrochemical impedance spectroscopy (EIS) measurements were measured from 0.001 kHz to 100 kHz of the frequency range with an initial voltage of 0.505 V. The stability of the catalyst at 10 mA cm^{-2} was evaluated by chronopotentiometry in O_2-saturated electrolyte.
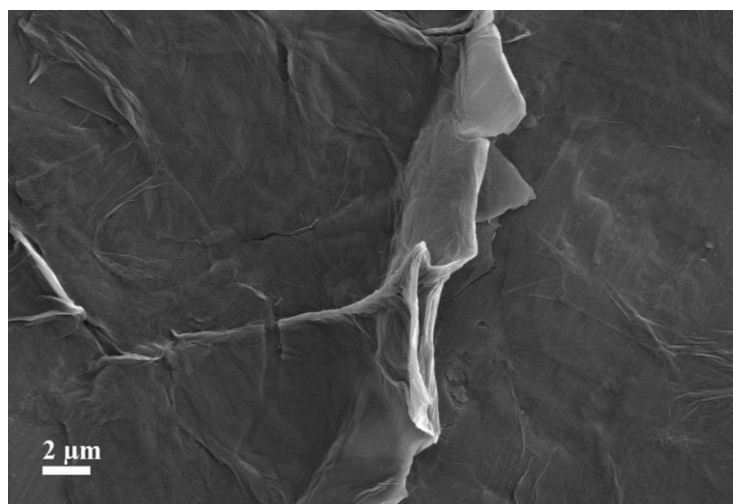
Fig. S1. SEM image of graphene oxide.
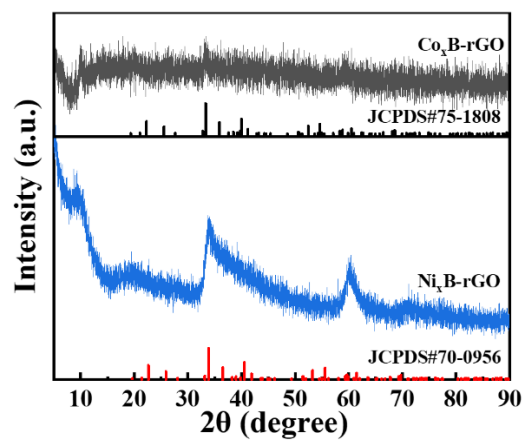
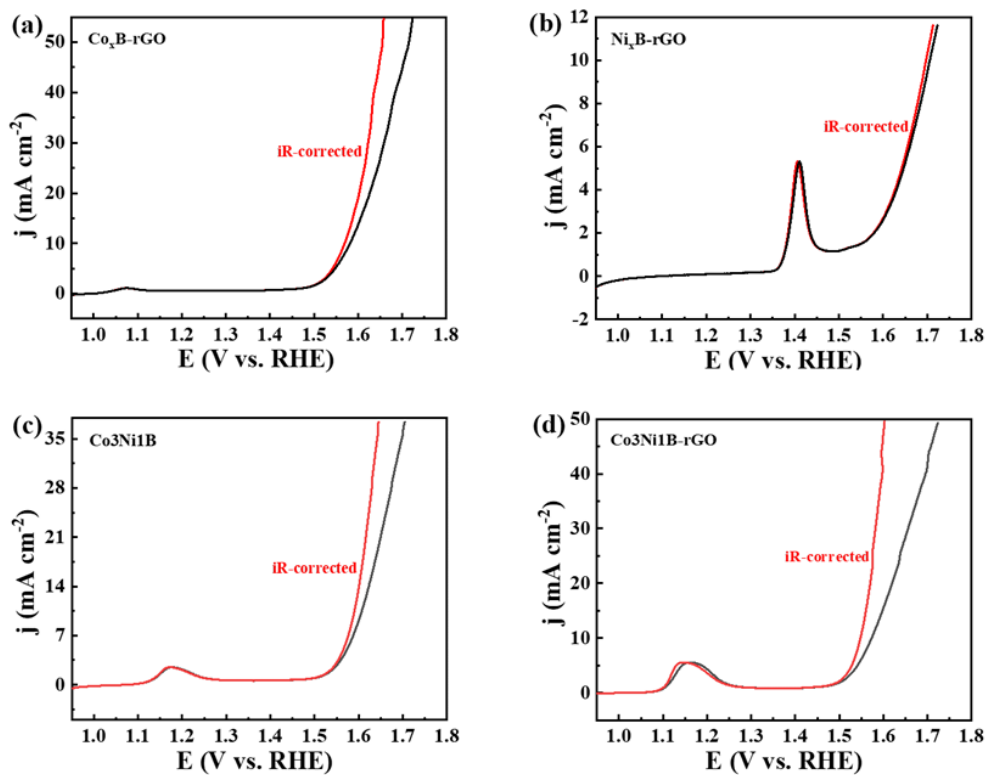Fig. S2. XRD patterns of Co_xB-rGO and Ni_xB-rGO sample.

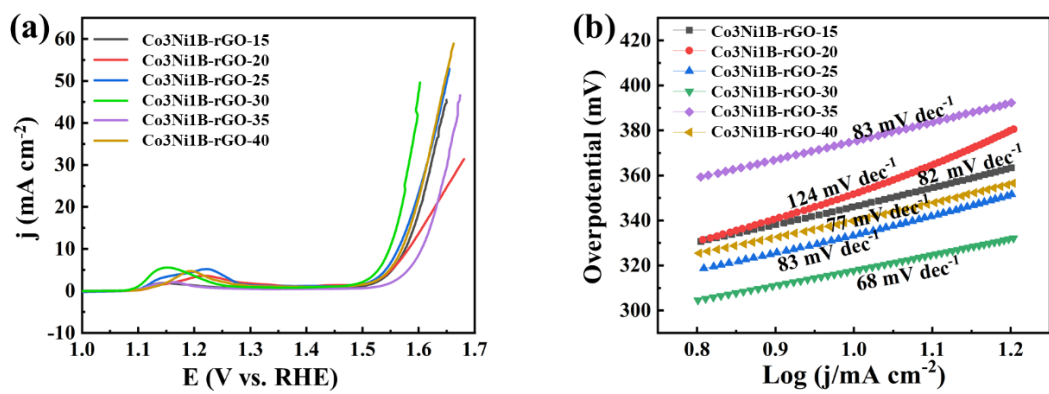Fig. S3. Polarization curves of catalysts with and without iR-compensation on GCE in 1.0 M KOH.

Fig. S4. The LSV curves with a scan rate of 5 mV s^{-1} in 1.0 M KOH solution and (b)

corresponding Tafel slope spectra of those catalysts with different GO contents.

Table S1. Catalyst parameters for preparing different GO contents

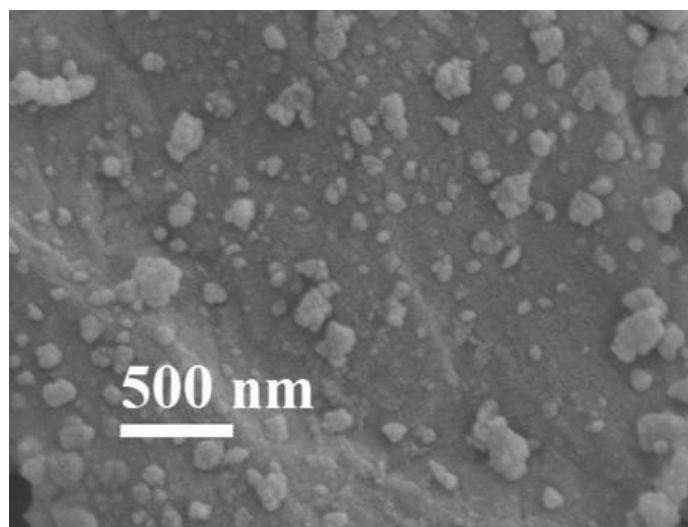| Materials	Co(NO₃)₂·6H₂O	Ni(NO₃)₂·6H₂O	NaBH₄	GO	η10	Tafel slope
	(mg)	(mg)	(mg)	(mg)	(mV)	(mV dec^{-1})
Co3Ni1B-rGO-15	291.03	96.93	227.00	15.00	246	82
Co3Ni1B- rGO-20	291.03	96.93	227.00	20.00	352	124
Co3Ni1B- rGO-25	291.03	96.93	227.00	25.00	334	83
Co3Ni1B- rGO-30	**291.03**	**96.93**	**227.00**	**30.00**	**318**	**68**
Co3Ni1B- rGO-35	291.03	96.93	227.00	35.00	375	83
Co3Ni1B- rGO-40	291.03	96.93	227.00	40.00	340	77

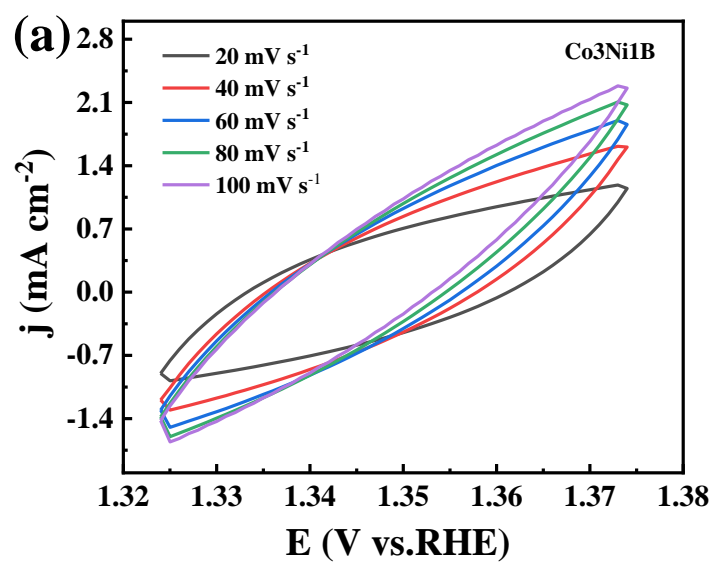Fig. S5. SEM image of Co3Ni1B-rGO catalyst after stability test.

Fig. S6. The CV curves at different scan rates for Co3Ni1B (a) sample.

References

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