

Supporting Information

Synthesis of $\text{Co}_{2-x}\text{Ni}_x\text{O}_2$ ($0 < x < 1.0$) Hexagonal Nanostructures as an Efficient Bifunctional Electrocatalysts for Overall Water Splitting

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Experiment section

Material Characterization

The crystal phases of several different $\text{Co}_{2-x}\text{Ni}_x\text{O}_2$ materials ($x=0,0.2,0.4,0.6,0.8$) synthesized were recorded by X-ray powder diffractometry (XRD), wherein Cu $K\alpha$ radiation ($\lambda = 0.15406 \text{ nm}$) 2θ ranged from 20 to 80. The Brunauer-Emmett-Teller (BET) experimental equipment (BET; Micromeritics, ASAP 2020, America) was used to analyze the BJH pore structure and BET specific surface area, which was achieved by nitrogen adsorption-desorption at a temperature of 77.35 K. The surface morphology and microstructure of nanocrystals with different Co : Ni molar ratios were observed by scanning electron microscopy (SEM; LEO1430VP, Germany) and transmission electron microscopy (TEM; FEI, Tecnai G2 F20 S-Twin, America).

Electrochemical measurements

The electrochemical test for oxygen evolution reaction (OER) and hydrogen evolution reaction (HER) was performed in a CHI760e electrochemical workstation (CHI 760E, CH Instruments Inc., Shanghai, China) in a three-electrode system with platinum wire counter electrode, Ag/AgCl reference electrode, and the working electrode of the $\text{Co}_{2-x}\text{Ni}_x\text{O}_2$ nanosheet samples in 1.0 M KOH electrolyte for OER(HER measurement is the same as OER).

We evaluated the OER performance of the material on nickel foam (NF) according to previous reports with some modifications ¹. Prior to the OER performance test, 2 mg of $\text{Co}_{2-x}\text{Ni}_x\text{O}_2$ powder was uniformly dispersed in a mixed solution of 500 μL of absolute ethanol and 500 μL of DI water by ultrasonic treatment for 10 minutes. Then,

15 μL of 60 wt% polytetrafluoroethylene (PTFE) solution was ultrasonically added to the mixed solution for 20 minutes to obtain an ink solution. Then 100 μL of the ink solution prepared in the above step was uniformly applied to the surface of the NF($\sim 1 \times 1 \text{ cm}^2$). In order to obtain a series of electrochemical properties of the materials synthesized by the above experimental methods:

The LSV measurements were taken at a scan rate of 5 mV/s. The polarization curves for the OER reaction were recorded on a voltage window of -0.2 to 0.8 V and the HER was recorded at -1 to -1.6 V. Recording the curve of electrochemical impedance spectroscopy (EIS) at an open circuit potential in the frequency range of 10 mHz to 100 kHz. The electrochemically active surface area (ECSA) is evaluated by the idea of measuring the electrochemical double-layer capacitance (C_{dl}), which is tested by cyclic voltammetry (CV) in a voltage range without the Faraday effect in 1.0 M KOH electrolyte. The voltage window is selected from 0 to 0.1 V and the scan rate is increased from 10 mV/s to 100 mV/s. In order to test the long-term stability of the above synthetic material, the measurement was performed at a voltage corresponding to the current density of 10 mA/cm² for a measurement of 30,000 seconds.

By performing RHE correction in the above-mentioned electrolyte, all measured potentials were corrected to reversible hydrogen electrodes (RHE), and the formula was as follows ² :

$$E_{\text{RHE}} = E_{\text{Ag/AgCl}} + 0.197 + 0.059 \text{ pH} \quad (1)$$

The iR compensation of ohmic potential drop losses caused by solution resistance is performed by this formula ^{3, 4}:

$$E_{iR \text{ compensation}} = E_{RHE} - iR \quad (2)$$

The overpotential of each material is obtained by the following formula²:

$$E_{\text{overpotential}} = E_{iR \text{ compensation}} - 1.23V \quad (3)$$

In addition, the reaction kinetics of the corresponding electrocatalysts were further detected by the Tafel slope. The linear portion of the Tafel image is fitted to the following equation ⁵:

$$\eta = a + b \log j \quad (4)$$

where " η " represents an overpotential, " b " represents the Tafel slope, and " j " represents the current density.

Overall water splitting is performed in the two-electrode system using $\text{Co}_{1.4}\text{Ni}_{0.6}\text{O}_2$ on both anode and cathode. To measure the performance of the overall water splitting, LSV polarization curves were recorded from 1.0 to 2.25 V at a scan rate of 5 mV/s. Chronoamperometric measurement was tested on corresponding potential to support a current density of around 10 mA/cm² for 30,000 seconds.

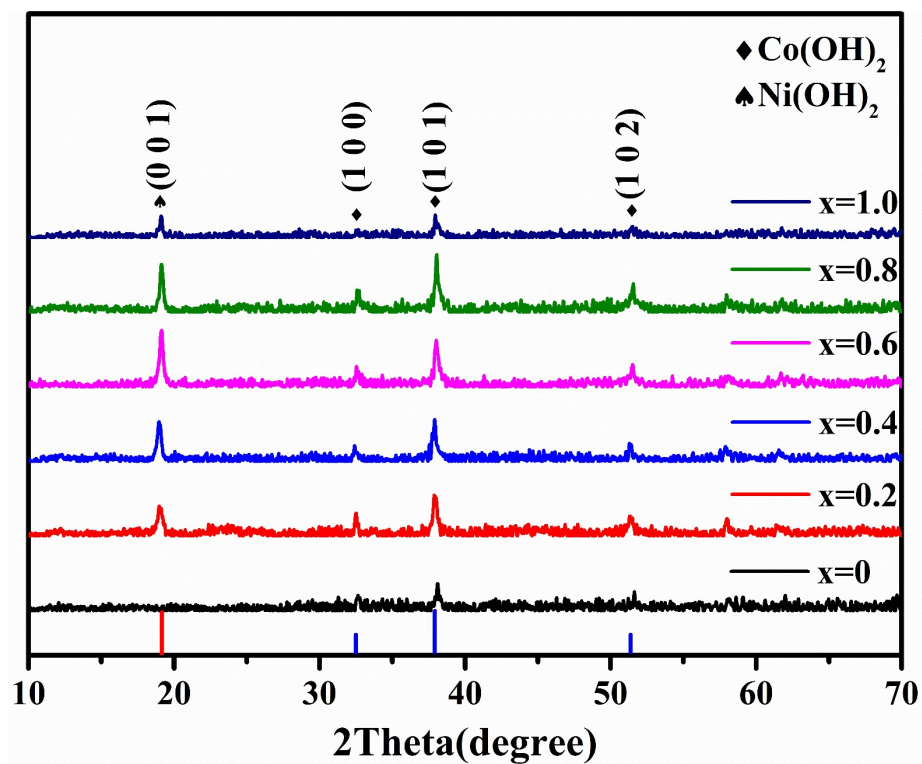


Fig.S1 XRD pattern of $\text{Co}_{2-x}\text{Ni}_x\text{O}_2$ precursor with x from 0 to 1.0.

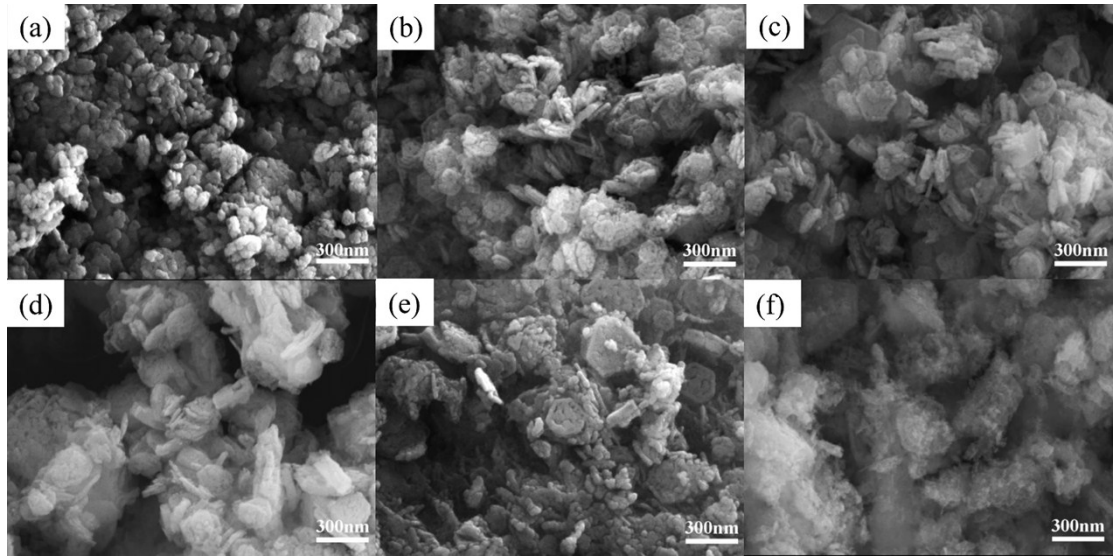


Fig.S2 SEM images of $\text{Co}_{2-x}\text{Ni}_x\text{O}_2$ nanosheets: (a) CoO ; (b) $\text{Co}_{1.8}\text{Ni}_{0.2}\text{O}_2$; (c) $\text{Co}_{1.6}\text{Ni}_{0.4}\text{O}_2$; (d) $\text{Co}_{1.4}\text{Ni}_{0.6}\text{O}_2$; (e) $\text{Co}_{1.2}\text{Ni}_{0.8}\text{O}_2$; (f) $\text{Co}_{1.0}\text{Ni}_{1.0}\text{O}_2$.

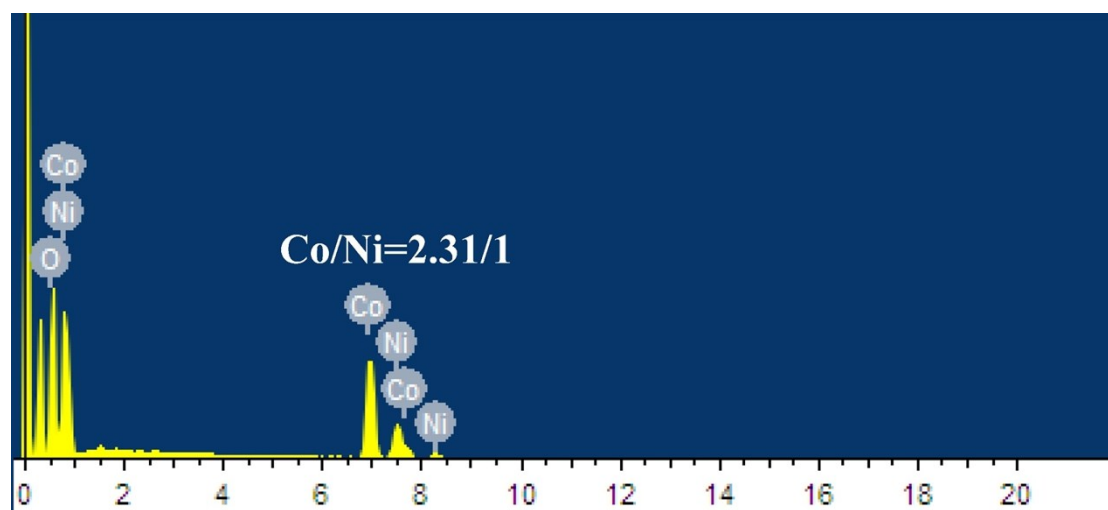


Figure S3. Energy-dispersive X-ray spectroscopy of $\text{Co}_{1.4}\text{Ni}_{0.6}\text{O}_2$.

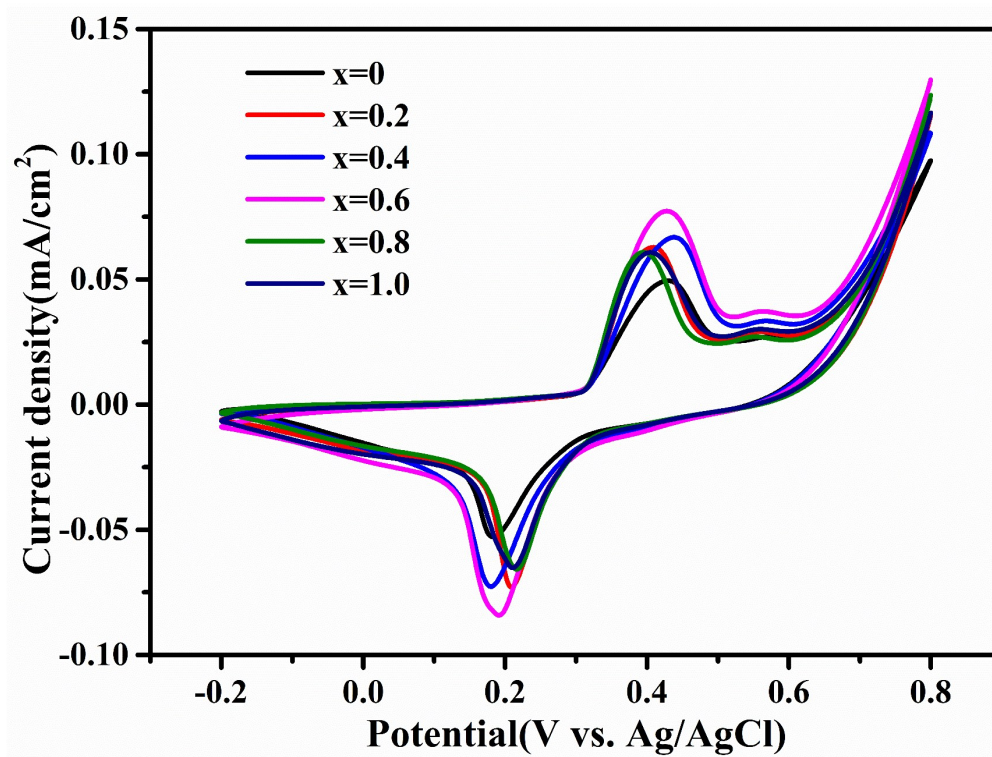


Fig. S4 CV curves of $\text{Co}_{2-x}\text{Ni}_x\text{O}_2$ with x from 0 to 1.0 at scan rate of 100 mV/s.

Table S1 Lattice parameters of synthetic Co_{2-x}Ni_xO₂ measured by XRD analysis

Catalyst	Lattice parameters						Vol. (Å ³)
	a (Å)	b (Å)	c (Å)	α	β	γ	
CoO	4.264	4.264	4.264	90	90	90	77.527
Co _{1.8} Ni _{0.2} O ₂	4.235	4.235	4.235	90	90	90	75.972
Co _{1.6} Ni _{0.4} O ₂	4.236	4.236	4.236	90	90	90	76.031
Co _{1.4} Ni _{0.6} O ₂	4.238	4.238	4.238	90	90	90	76.127
Co _{1.2} Ni _{0.8} O ₂	4.240	4.240	4.240	90	90	90	76.226
Co _{1.0} Ni _{1.0} O ₂	4.247	4.247	4.247	90	90	90	76.603

References

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