

***Electronic Supplementary Information***

General approach to construct hierarchical-structured porous Co-Ni bimetallic oxide for efficient oxygen evolution

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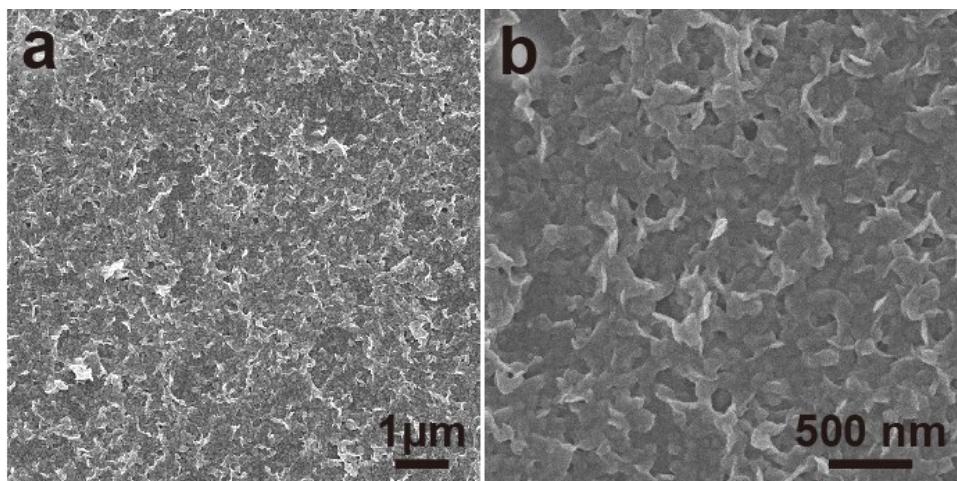
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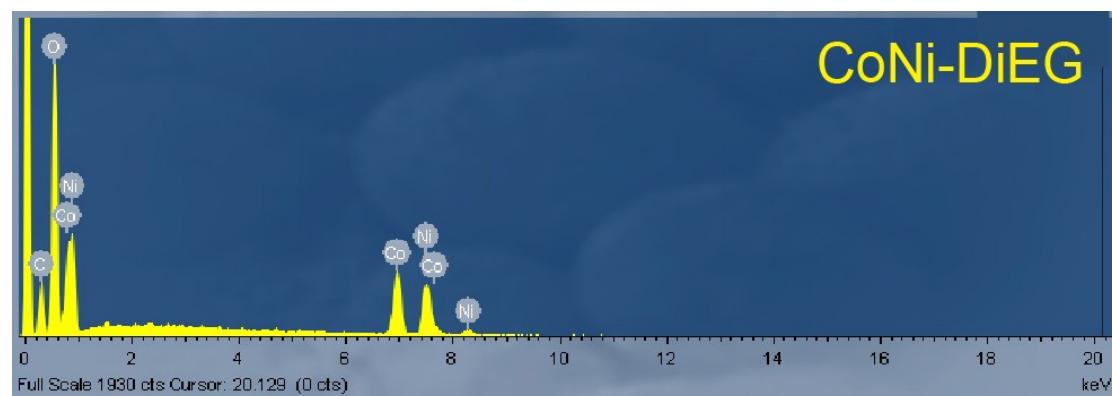
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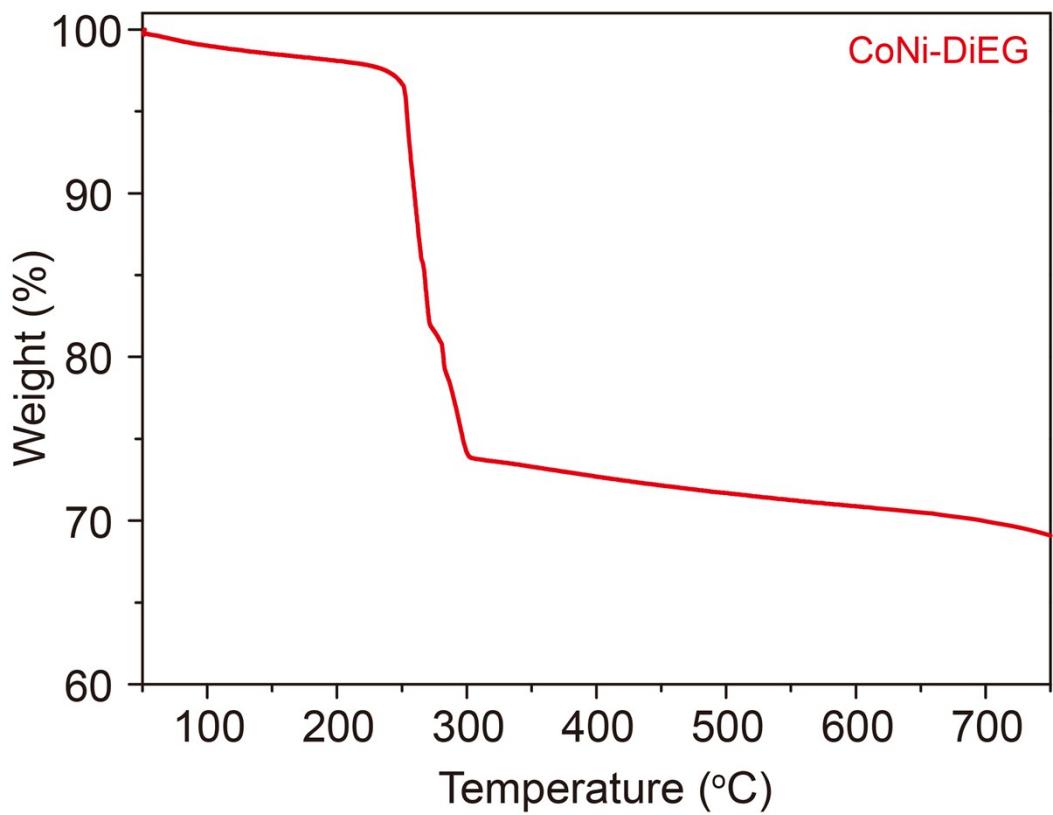
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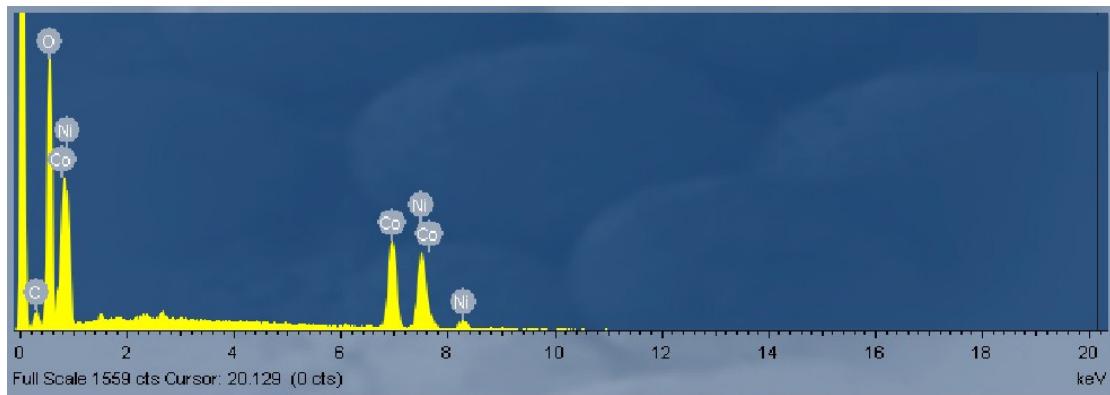
**Figure S1.** (a,b) SEM images of the CoNi-DiEG prepared by using pure DiEG as solvent without adding methanol.



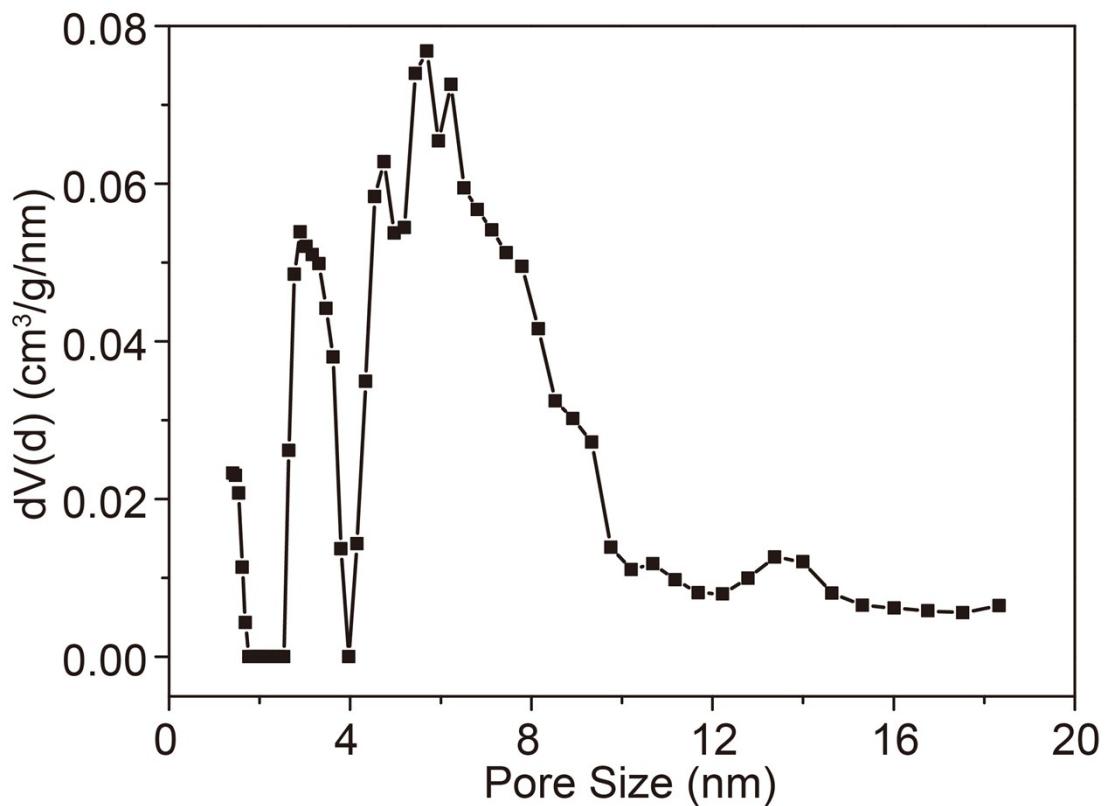
**Figure S2.** The EDX spectrum of the CoNi-DiEG precursor.



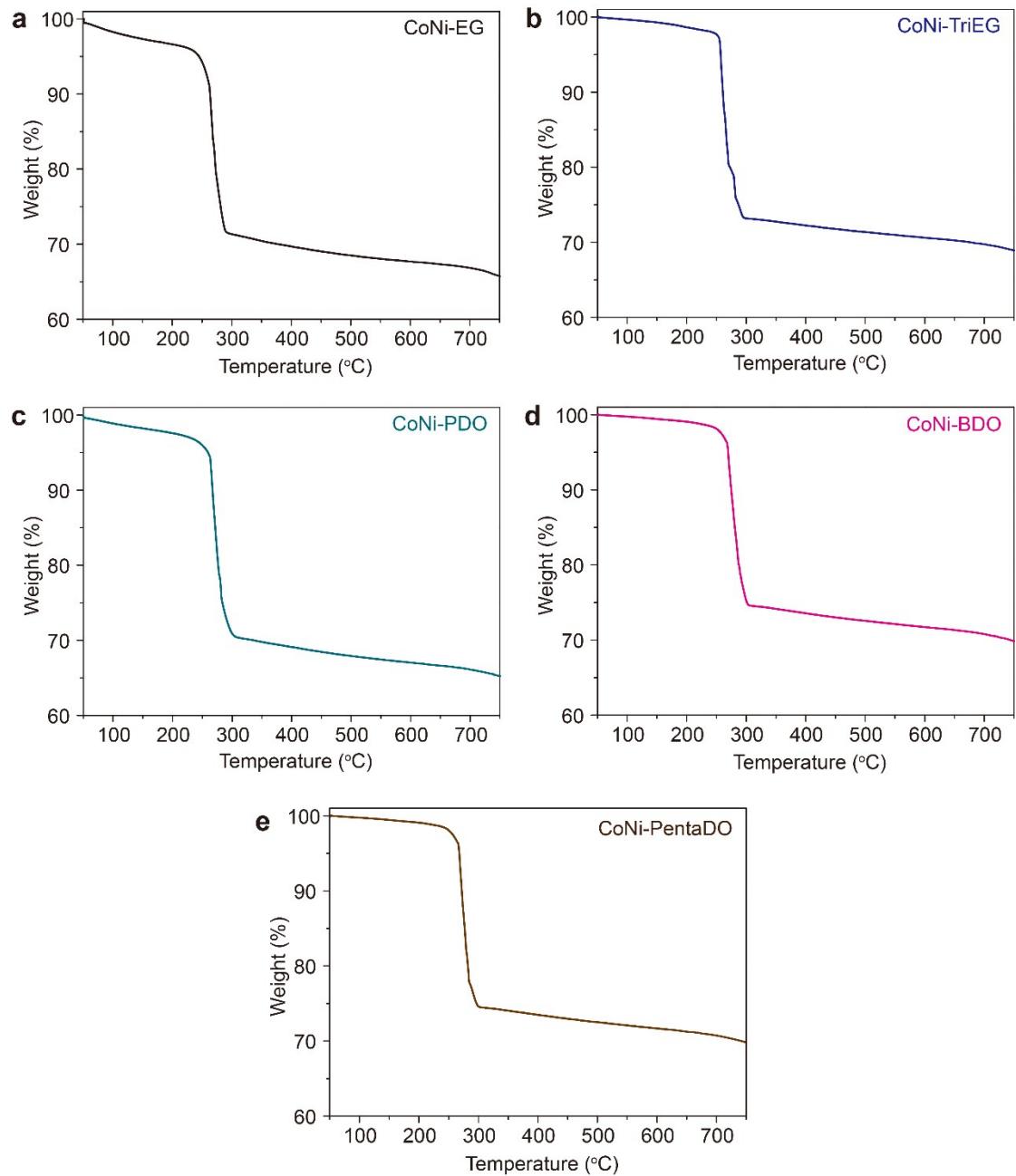
**Figure S3.** The TGA curve of the CoNi-DiEG precursor.



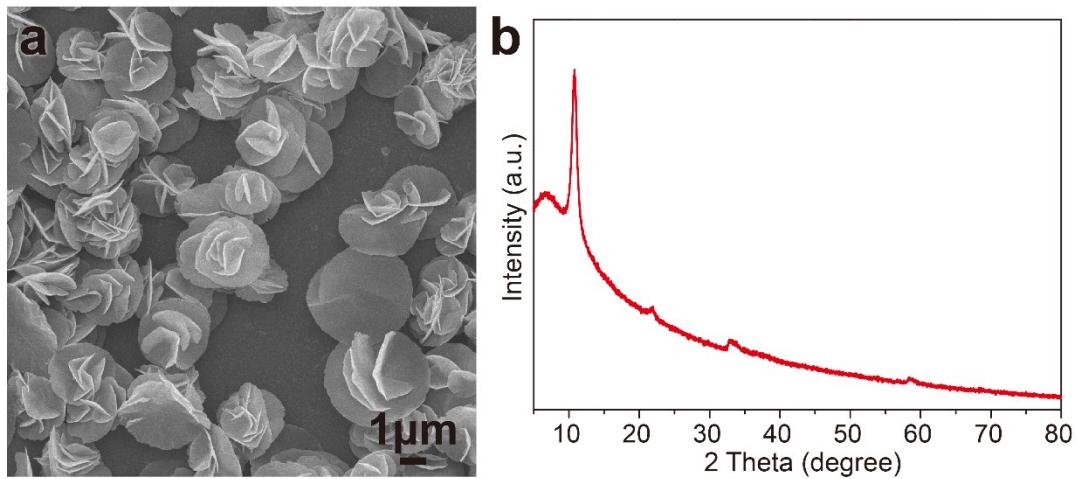
**Figure S4.** The EDX spectrum of the  $\text{CoNiO}_x$ .



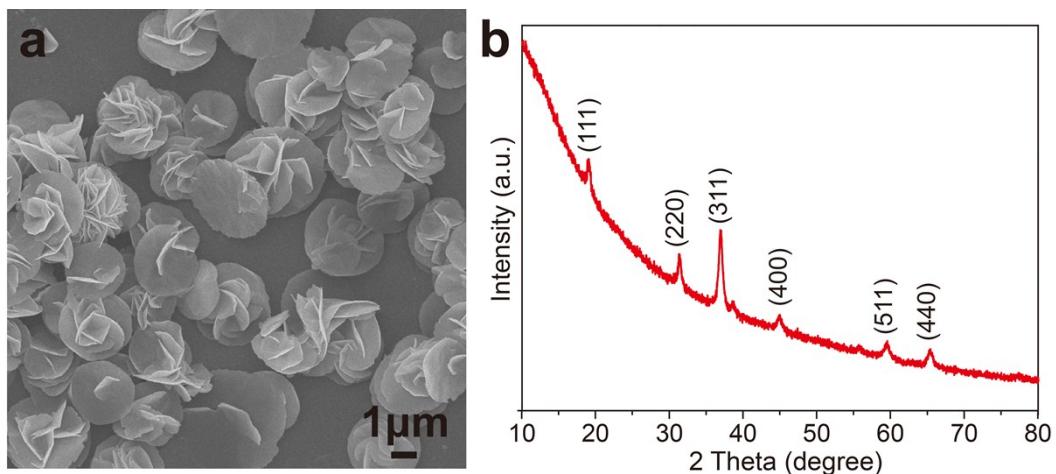
**Figure S5.** The NLDFT pore size distribution curve of the CoNiO<sub>x</sub>.



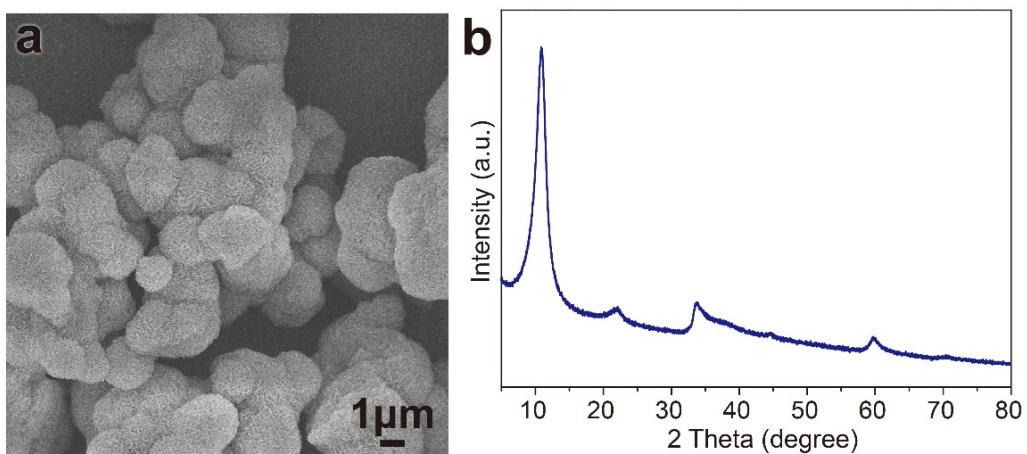
**Figure S6.** The TGA curves of various CoNi-polyol complexes: (a) CoNi-EG, (b) CoNi-TriEG, (c) CoNi-PDO, (d) CoNi-BDO, and (e) CoNi-PentaDO.



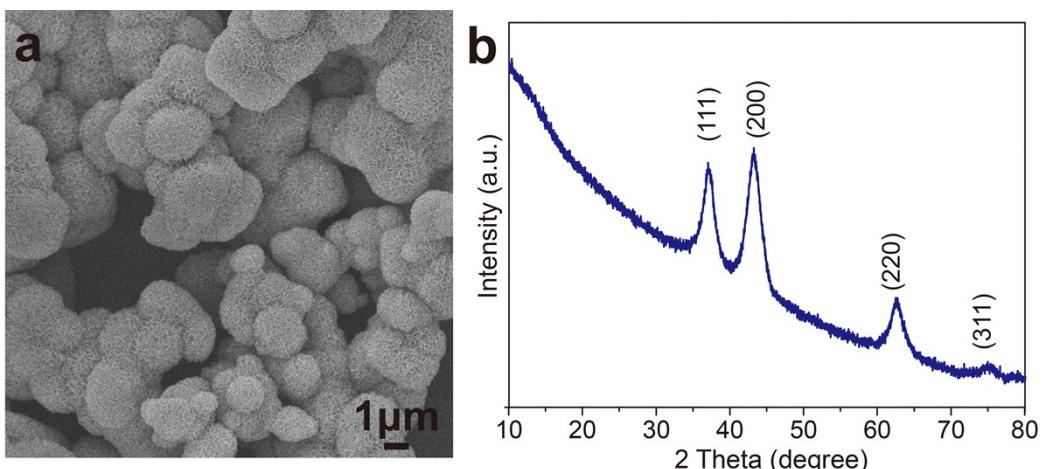
**Figure S7.** Characterizations of Co-DiEG precursor: (a) SEM image and (b) XRD pattern.



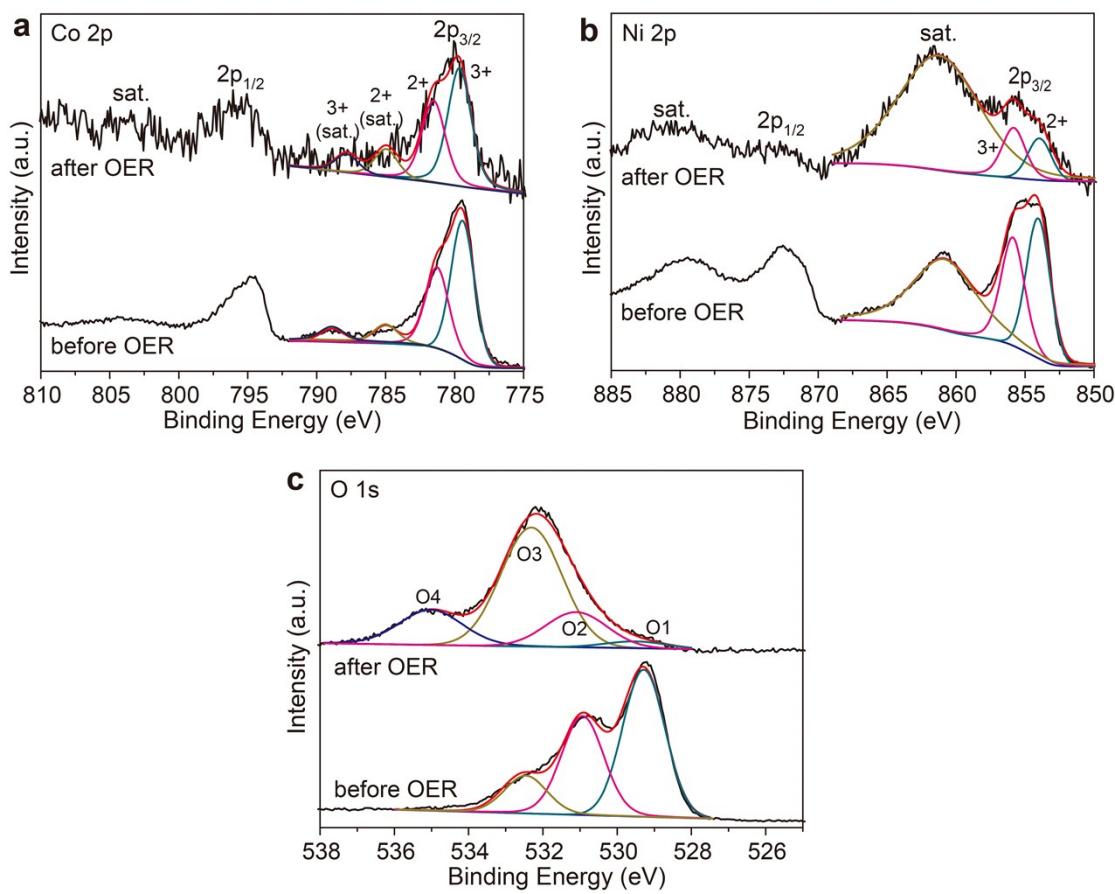
**Figure S8.** Characterizations of  $\text{Co}_3\text{O}_4$ : (a) SEM image and (b) XRD pattern.



**Figure S9.** Characterizations of Ni-DiEG precursor: (a) SEM image and (b) XRD pattern.



**Figure S10.** Characterizations of NiO: (a) SEM image and (b) XRD pattern.



**Figure S11.** (a) Co 2p, (b) Ni 2p, and (c) O 1s XPS spectra of the CoNiO<sub>x</sub> before and after the electrochemical CP stability test.

**Notes:** For the XPS spectra of the CoNiO<sub>x</sub>, the Co 2p (Figure S11a) and Ni 2p spectra (Figure S11b) are split into 2p<sub>3/2</sub> and 2p<sub>1/2</sub> doublets, due to the spin-orbit coupling, together with shakeup satellite peaks. And both Co 2p<sub>3/2</sub> and Ni 2p<sub>3/2</sub> spectra can be deconvoluted into two distinct metal species, i.e., Co<sup>2+</sup> (781.3 eV) and Co<sup>3+</sup> (779.4 eV), Ni<sup>2+</sup> (854.2 eV) and Ni<sup>3+</sup> (855.9 eV), indicating the coexistence of Co<sup>2+</sup>, Co<sup>3+</sup>, Ni<sup>2+</sup> and Ni<sup>3+</sup> in the sample.<sup>1</sup> In the O 1s spectrum (Figure S11c), three peaks can be clearly identified. The peak at ~529.3 eV is attributed to the lattice oxygen in the spinel oxide, the peak at ~530.9 eV is assigned to the surface defect sites with a low oxygen coordination, and the peak at ~532.5 eV can be assigned to the hydroxyl species.<sup>2,3</sup>

**Table S1.** Comparison of the OER catalytic performance of our hierarchical-structured porous CoNiO<sub>x</sub> to other recently reported high-performance OER catalysts in alkaline solution.

Catalyst	Mass loading (mg cm <sup>-2</sup> )	Electrolyte	η@10 mA cm <sup>-2</sup> (mV)	Tafel slope (mV dec <sup>-1</sup> )	Ref.
<b>CoNiO<sub>x</sub></b>	<b>0.2</b>	<b>0.1 M KOH</b>	<b>329 309 (after CP test)</b>	<b>66</b>	<b>this work</b>
α-Ni(OH) <sub>2</sub> spheres	0.2	0.1 M KOH	331	42	4
β-Ni(OH) <sub>2</sub> plates	0.2	0.1 M KOH	444	111	4
Hollow Co <sub>3</sub> S <sub>4</sub> nanosheets	0.283	0.1 M KOH	363	90	5
Mn <sub>3</sub> O <sub>4</sub> /CoSe <sub>2</sub>	0.2	0.1 M KOH	450	49	6
N-graphene-CoSe <sub>2</sub>	0.2	0.1 M KOH	366	40	7
N-Co <sub>9</sub> S <sub>8</sub> /graphene	0.2	0.1 M KOH	409	82.7	8
Co <sub>3</sub> O <sub>4</sub> /C nanowire arrays	0.2	0.1 M KOH	290	70	9
Ordered mesoporous Co <sub>3</sub> O <sub>4</sub>	0.12	0.1 M KOH	496-537	86-96	10
Ba <sub>0.5</sub> Sr <sub>0.5</sub> Co <sub>0.8</sub> Fe <sub>0.2</sub> O <sub>3-d</sub>	0.25	0.1 M KOH	362	48	11
FeNC sheets/NiO	0.24	0.1 M KOH	390	76	12
CoNC sheets/NiO	0.24	0.1 M KOH	410	80	12
Ultrathin NiCo <sub>2</sub> O <sub>4</sub> nanosheets	0.285	0.1 M KOH	415	N.A.	2
rGO@CoNiO <sub>x</sub>	0.2	0.1 M KOH	320	45	3
NiFe-LDH/CNTs	0.2	0.1 M KOH	308	35	13
sea-urchin-like (Co <sub>0.54</sub> Fe <sub>0.46</sub> )P <sub>2</sub>	0.2	0.1 M KOH	370	N.A.	14
Ag-CoSe <sub>2</sub> nanobelts	0.2	0.1 M KOH	320	56	15

N-doped graphitic carbon	0.2	0.1 M KOH	380	75-80	16
P-doped graphitic C <sub>3</sub> N <sub>4</sub>	0.2	0.1 M KOH	400	61.6	17
Graphitic C <sub>3</sub> N <sub>4</sub> nanosheets/carbon nanotubes	0.2	0.1 M KOH	370	83	18
IrO <sub>2</sub> /C	0.2	0.1 M KOH	370	N.A.	16
Rutile RuO <sub>2</sub>	0.05	0.1 M KOH	> 470	N.A.	19
RuO <sub>2</sub> /C	0.2	0.1 M KOH	380	157.5	8
RuO <sub>2</sub>	0.2	0.1 M KOH	387	90	4
Ni <sub>x</sub> Co <sub>3-x</sub> O <sub>4</sub> nanowire arrays	2.3-2.7	1 M NaOH	370	59-64	20
Ni-Co oxide hierarchical nanosheets	N.A.	1 M NaOH	340	51	21
Amorphous NiCo <sub>2.7</sub> (OH) <sub>x</sub> nanocages	0.2	1 M KOH	350	65	22
NiCo LDH nanosheets	0.17	1 M KOH	367	40	23
rGO/CoNi-P	0.1	1 M KOH	314	60.0	24
CoP/NCNHP	N.A.	1 M KOH	310	70	25
CoP/rGO	0.28	1 M KOH	340	66	26

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