Supporting Information

Chromium-Modified Ultrathin CoFe LDH as High-Efficiency Electrode for Hydrogen Evolution Reaction

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1. Experimental details

Preparation of Pt/C

Prior to the synthesis, NFs were sonicated in 3 M HCl, ethanol and ultrapure water for 15min to clean the surface. 1.2mg Pt/C was dispersed into a solution of water, ethanol and Nafion with a volume ratio of 60/120/15 via sonication. Coat a third of the total well-dispersed solution to the cleaned NF surface.

2. Supplementary Figures:

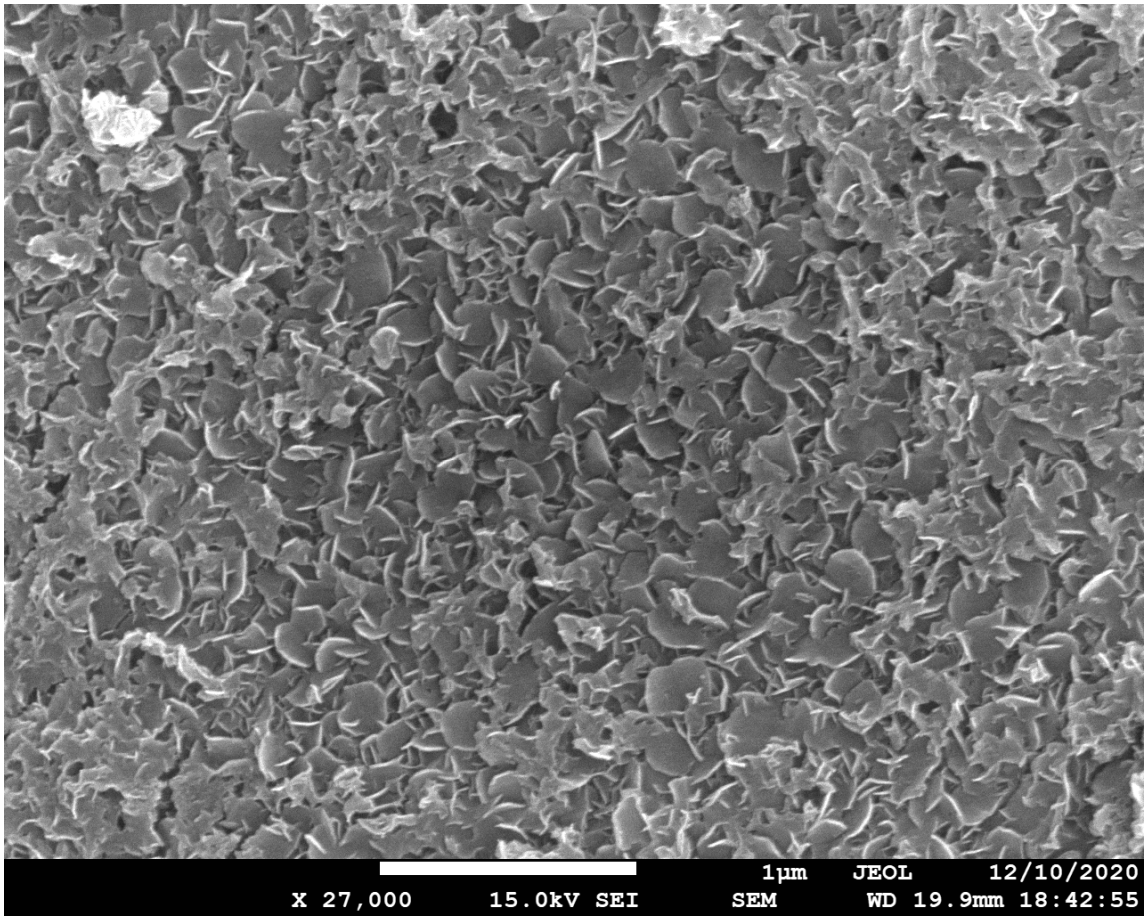

Figure S1. The SEM of CoFeCr LDH electrode. The results show that the electrode has obvious array structure, which is beneficial to mass transfer in the catalytic process.

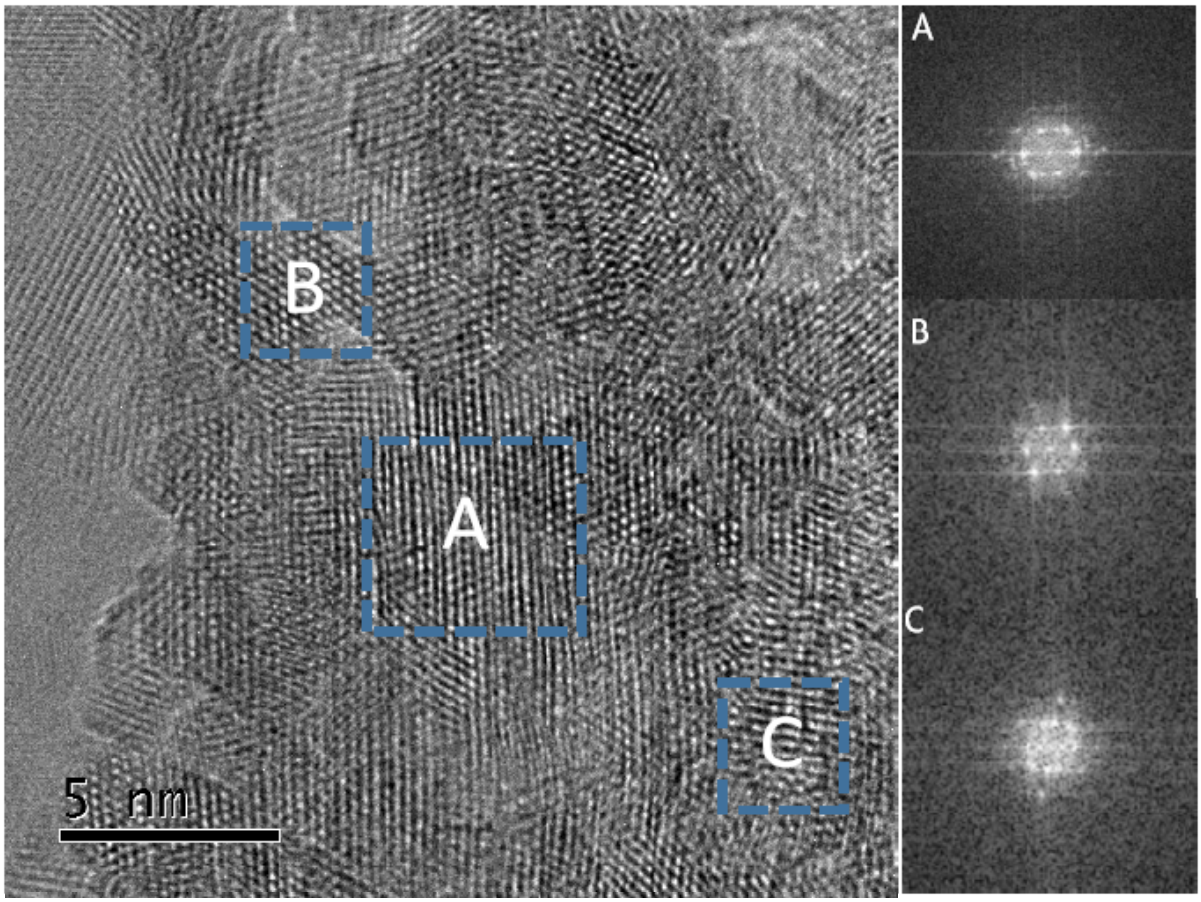

Figure S2. The HRTEM and corresponding fast-Fourier-transform (FFT) of the as-obtained CoFeCr LDH electrode. The results show that the sample is polycrystalline in nature.

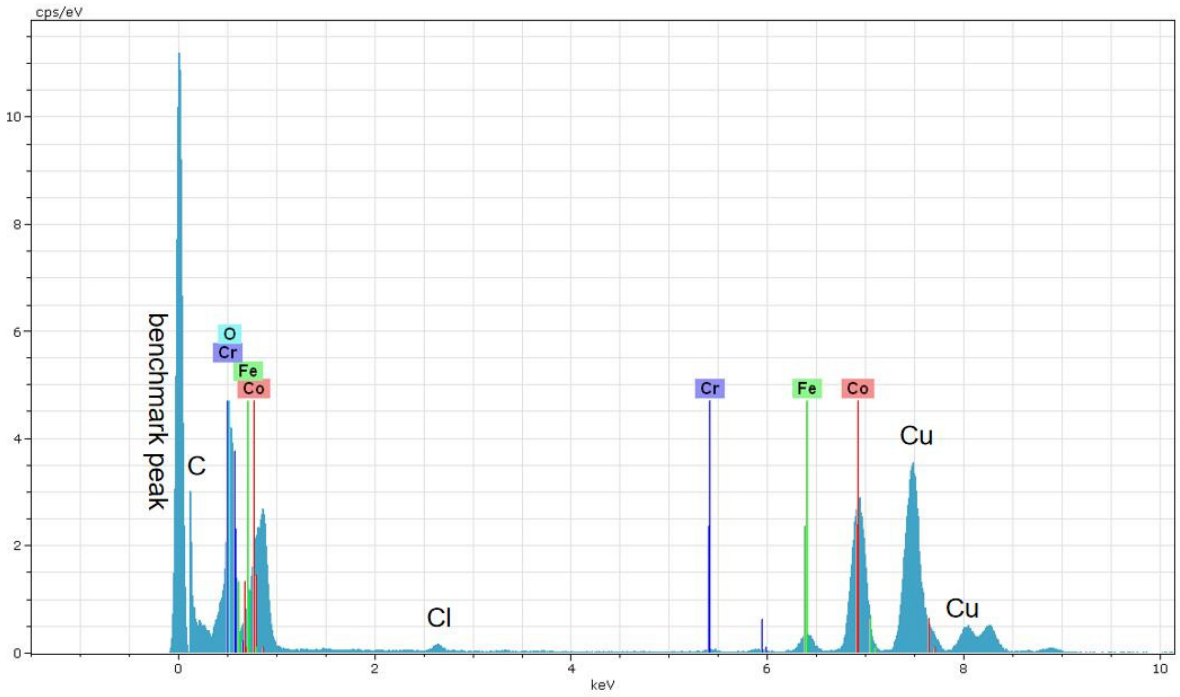

Figure S3. The energy-dispersive X-ray (EDX) of CoFeCr LDH electrode.

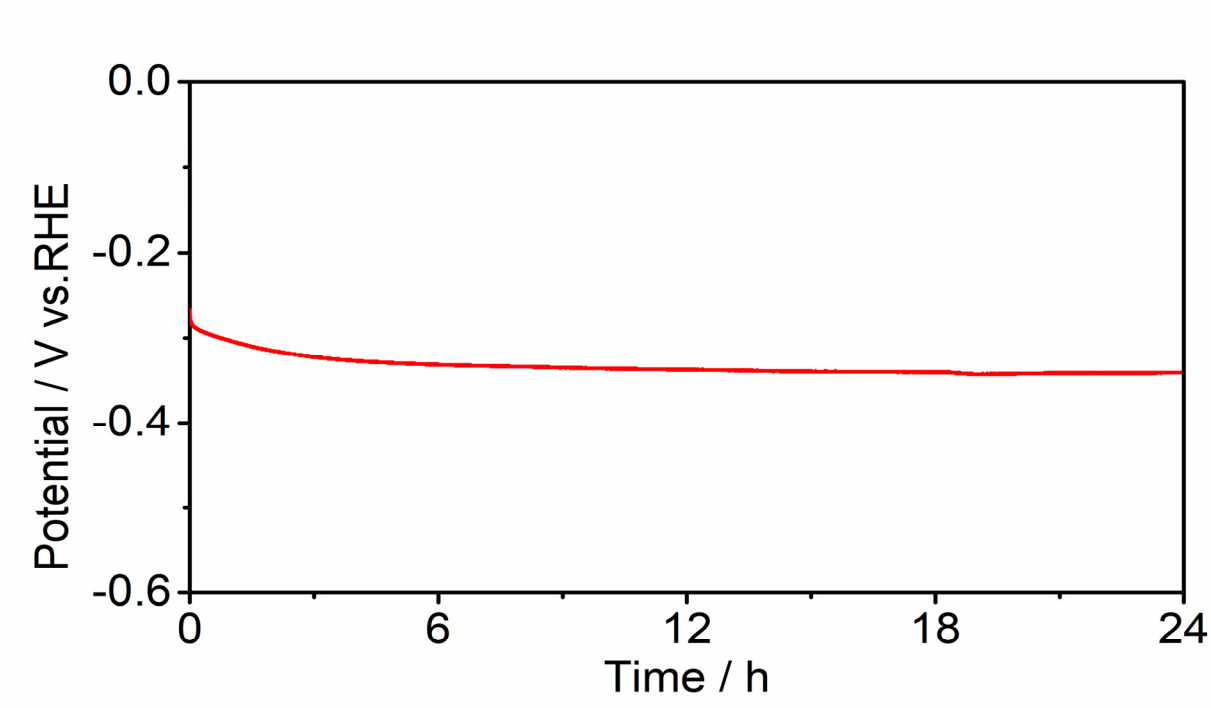

Figure S4. The chronopotentiometry measurement of CoFeCr LDH electrode. The current density used for the chronopotentiometry measurement is 10 mA cm^{-2}. The electrolyte used for the test is 1.0 M KOH.

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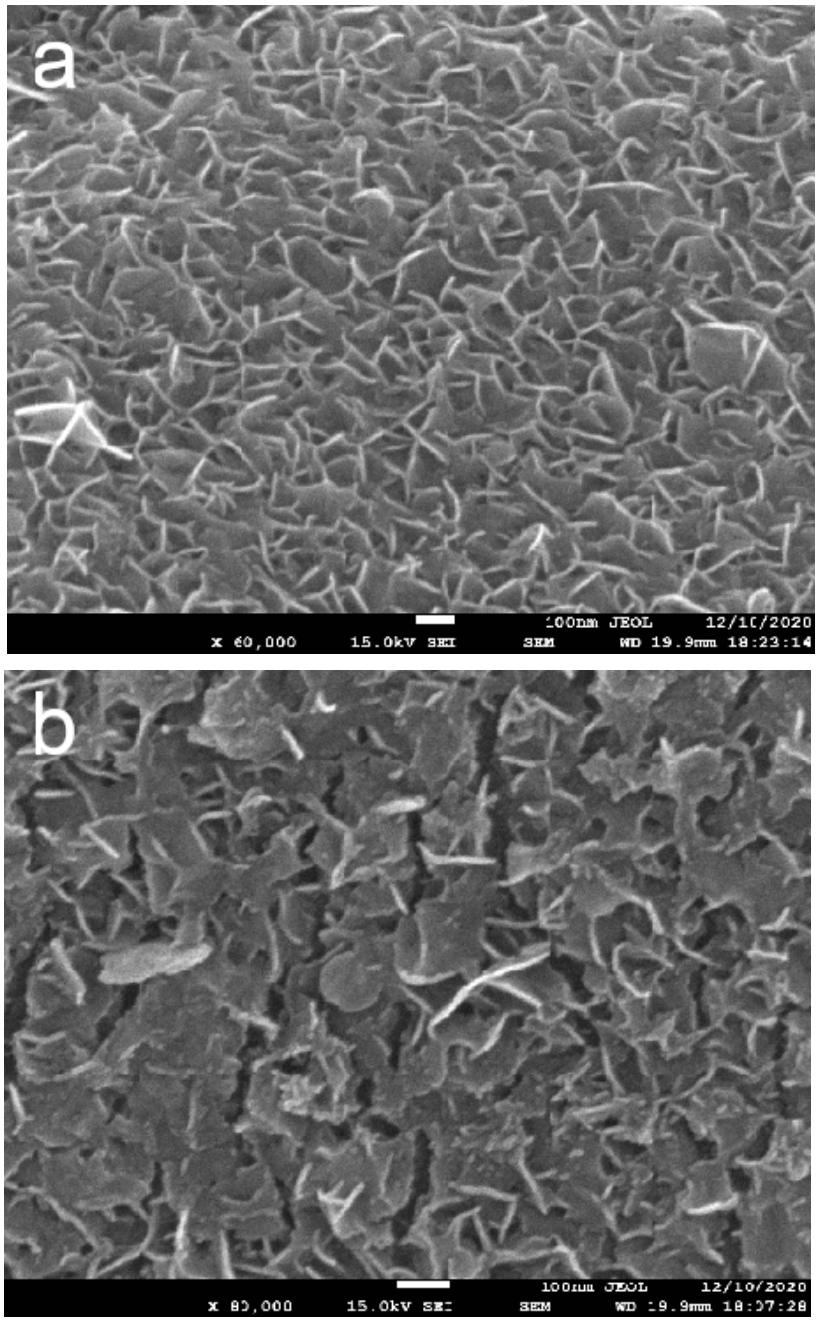

Figure S5. The SEM images of CoFeCr LDH electrode after long-time stability test.

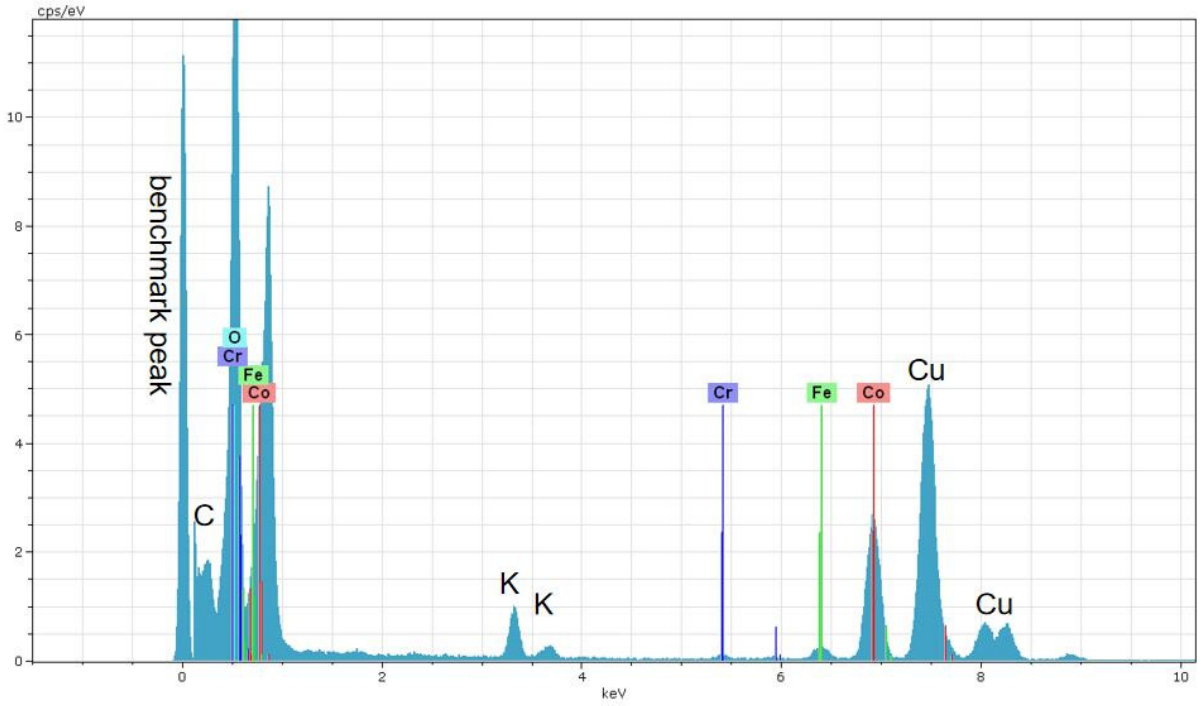

Figure S6. The energy-dispersive X-ray (EDX) of CoFeCr LDH electrode after long-time stability test.

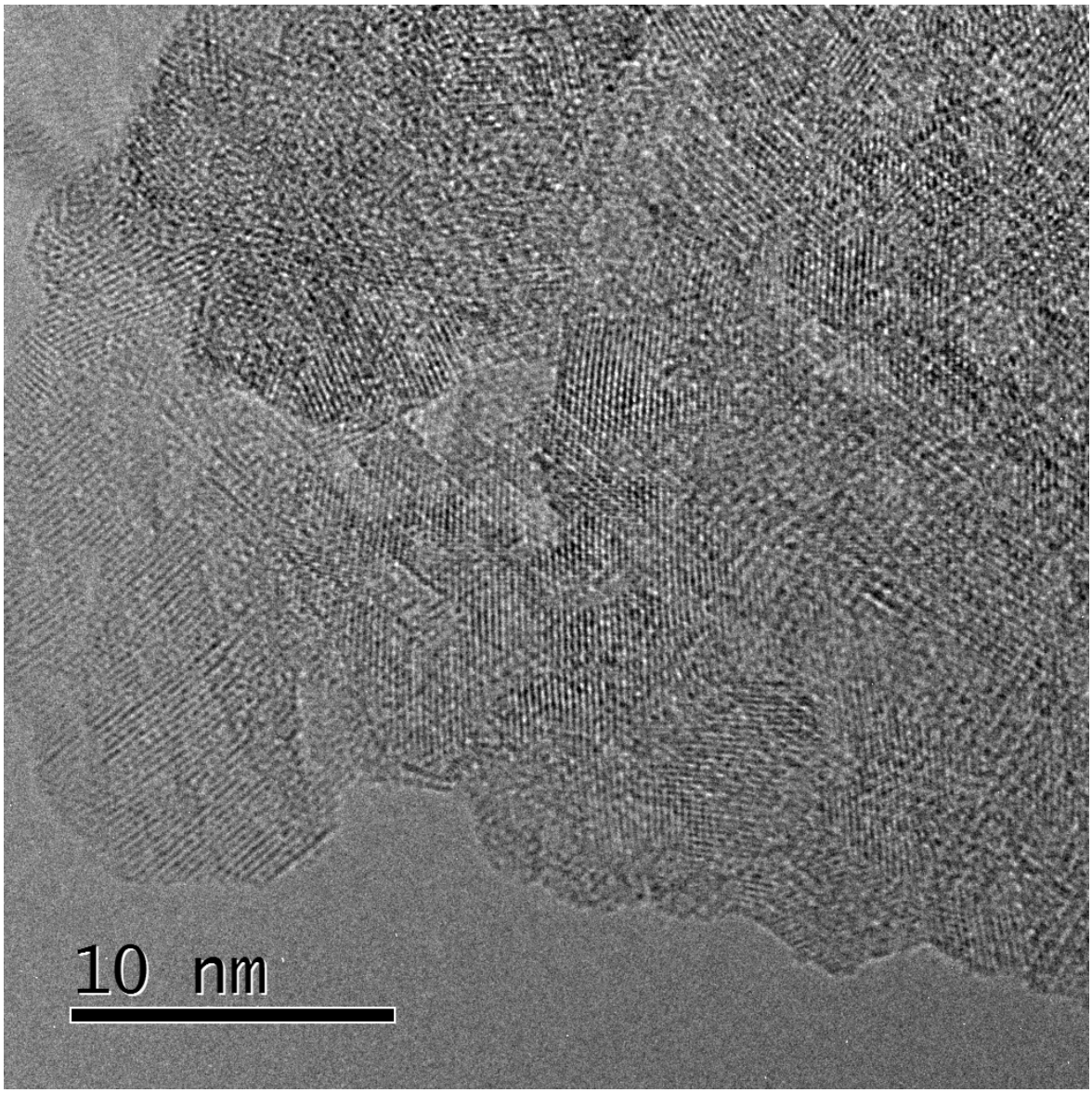

Figure S7. The HRTEM image of CoFeCr LDH electrode after long- time stability test.

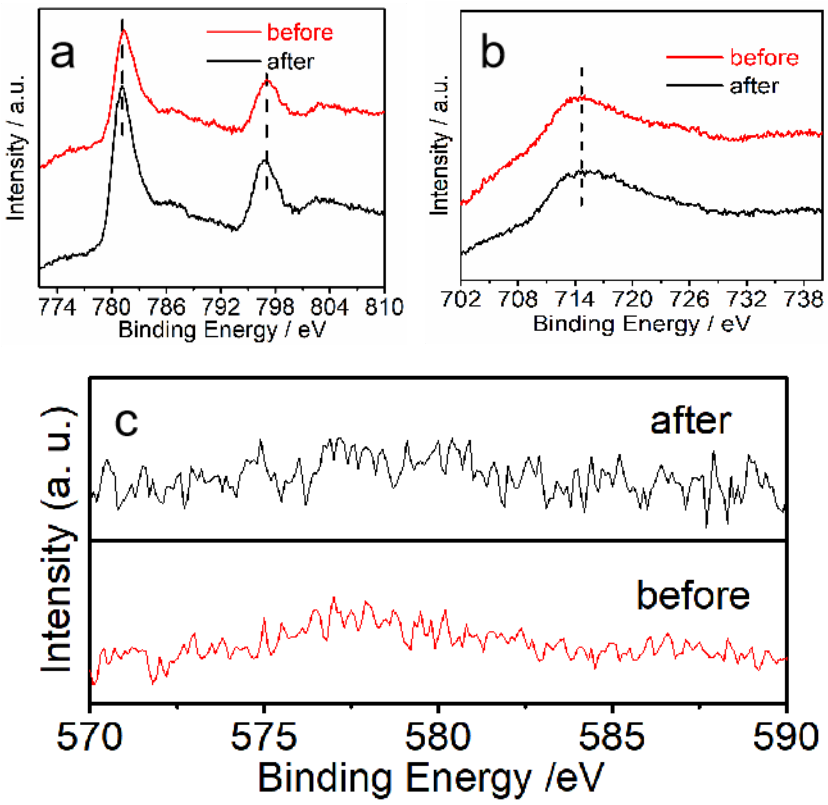

Figure S8. High-resolution XPS (a) Co 2p, (b) Fe 2p and (c) Cr 2p spectra of CoFeCr LDH electrode before and after long-time stability test.

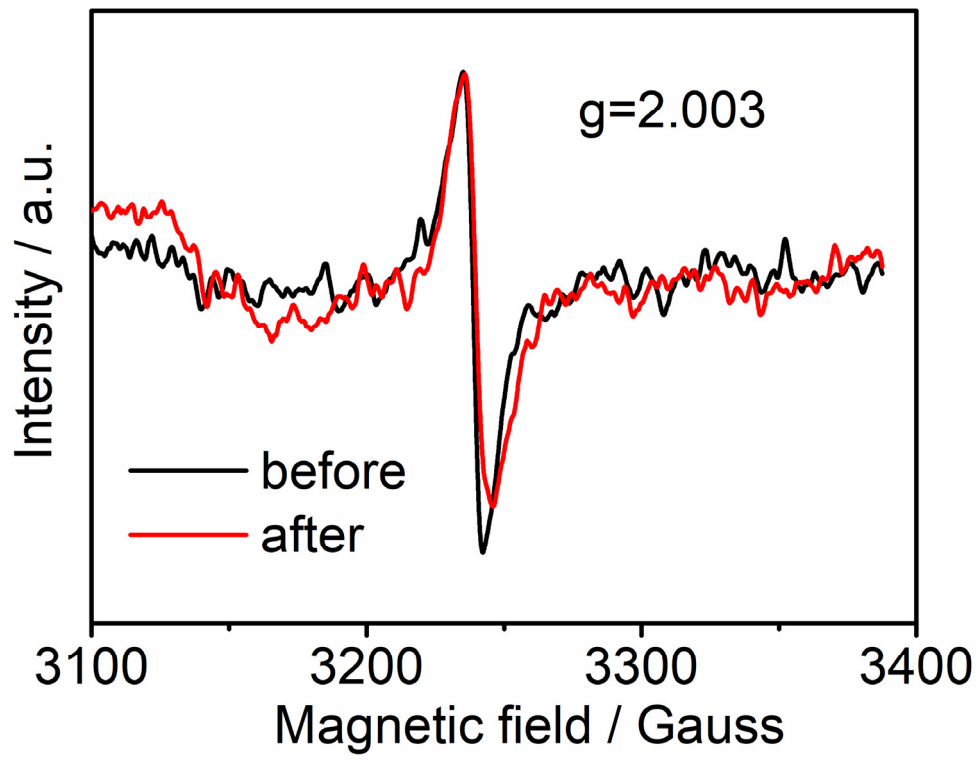

Figure S9. The electron paramagnetic resonance (EPR) spectra of CoFeCr LDH electrode before and after long time stability test.

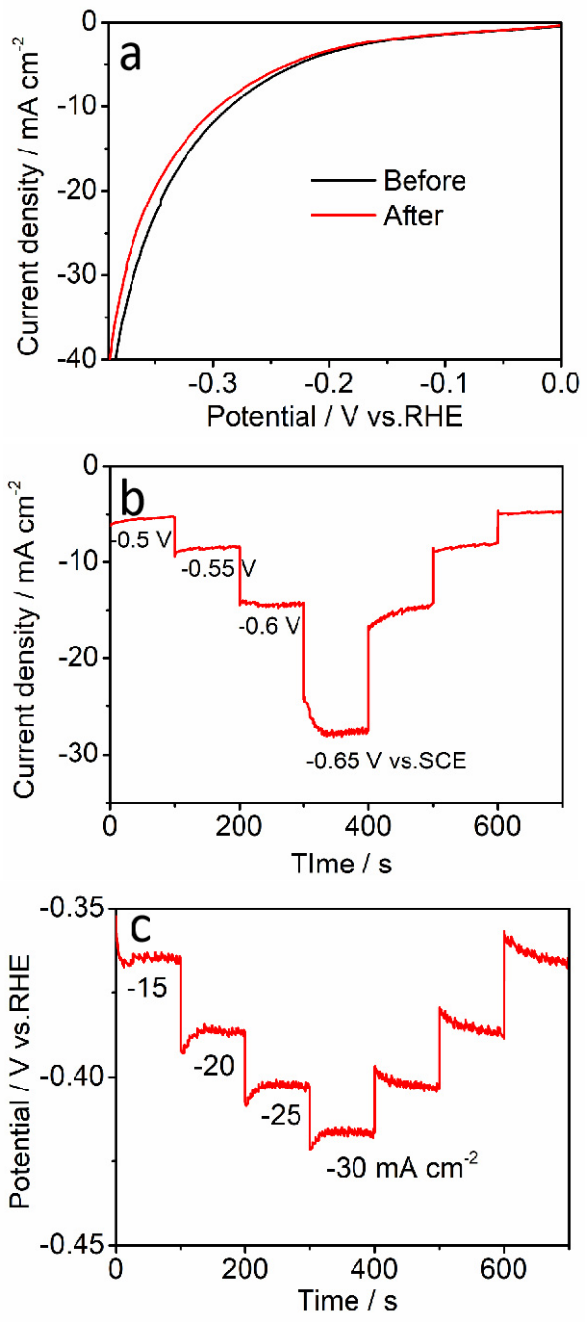

Figure S10. (a)the polarization curves of the electrode before and after 500 cycles. (b) the multi-potential steps and (c) the multi-current steps results.

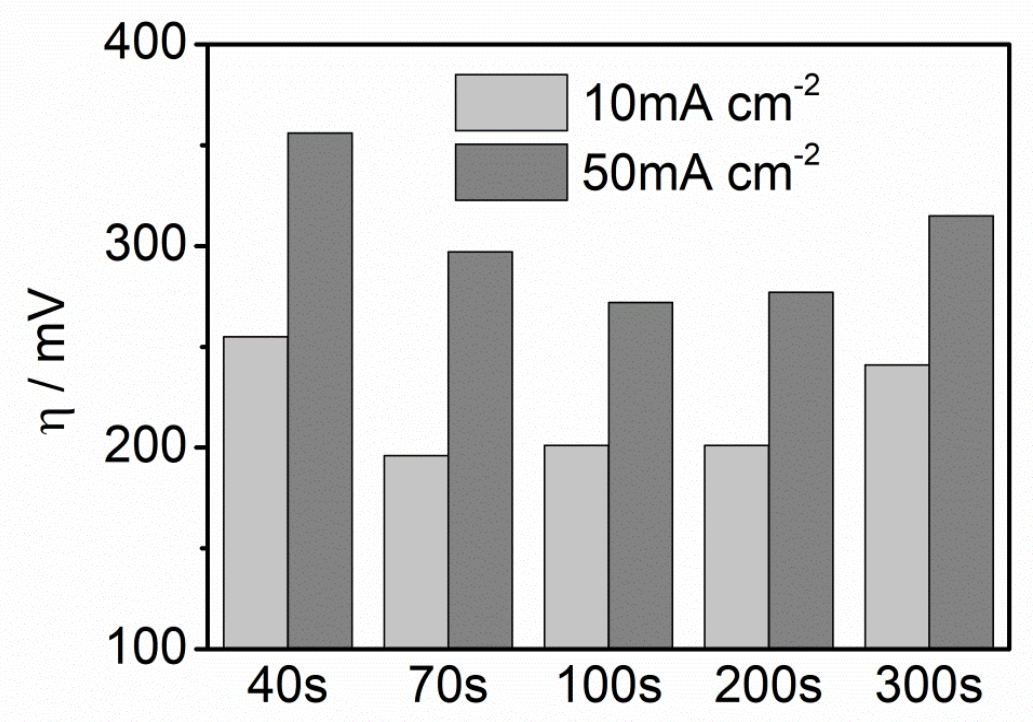

Figure S11. The comparison of the corresponding overpotentials at 10 mA cm^{-2} and 50 mA cm^{-2} current outputs based on different deposition time electrodes.

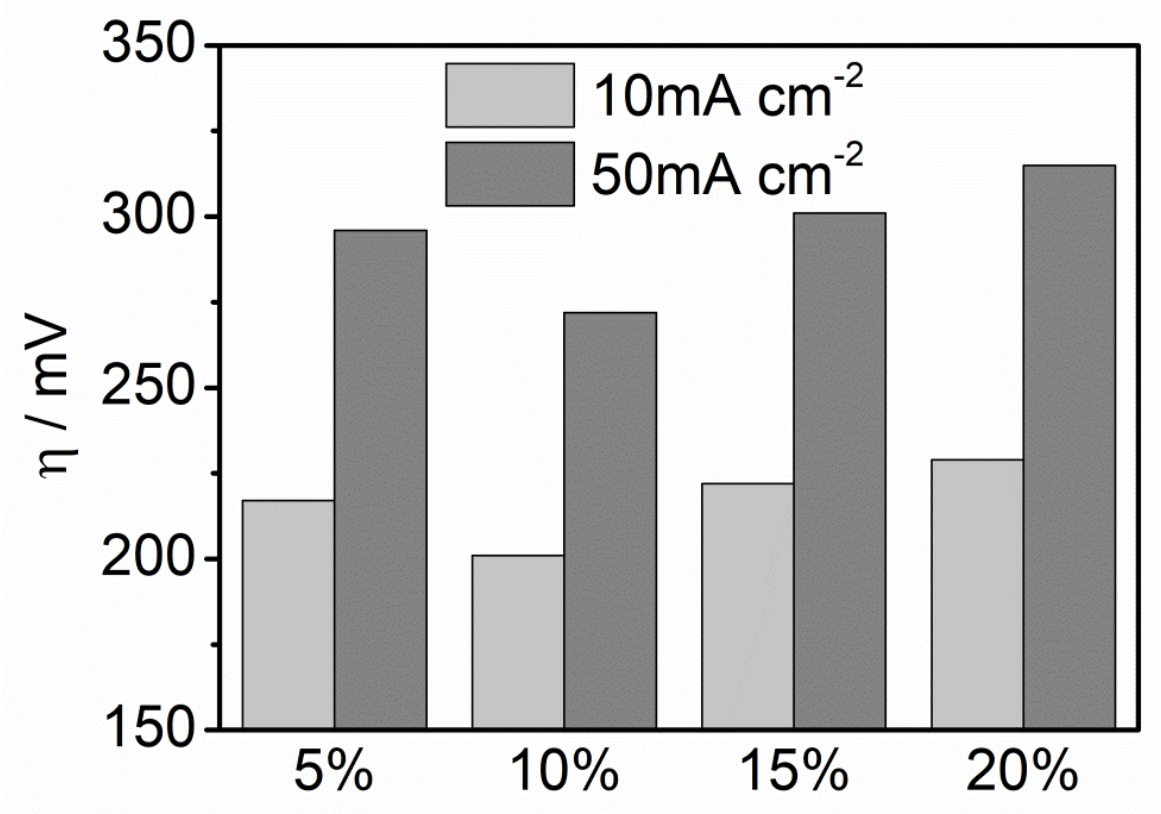

Figure S12. The comparison of the corresponding overpotentials at 10 mA cm^{-2} and 50 mA cm^{-2} current outputs based on different doping concentrations electrodes.

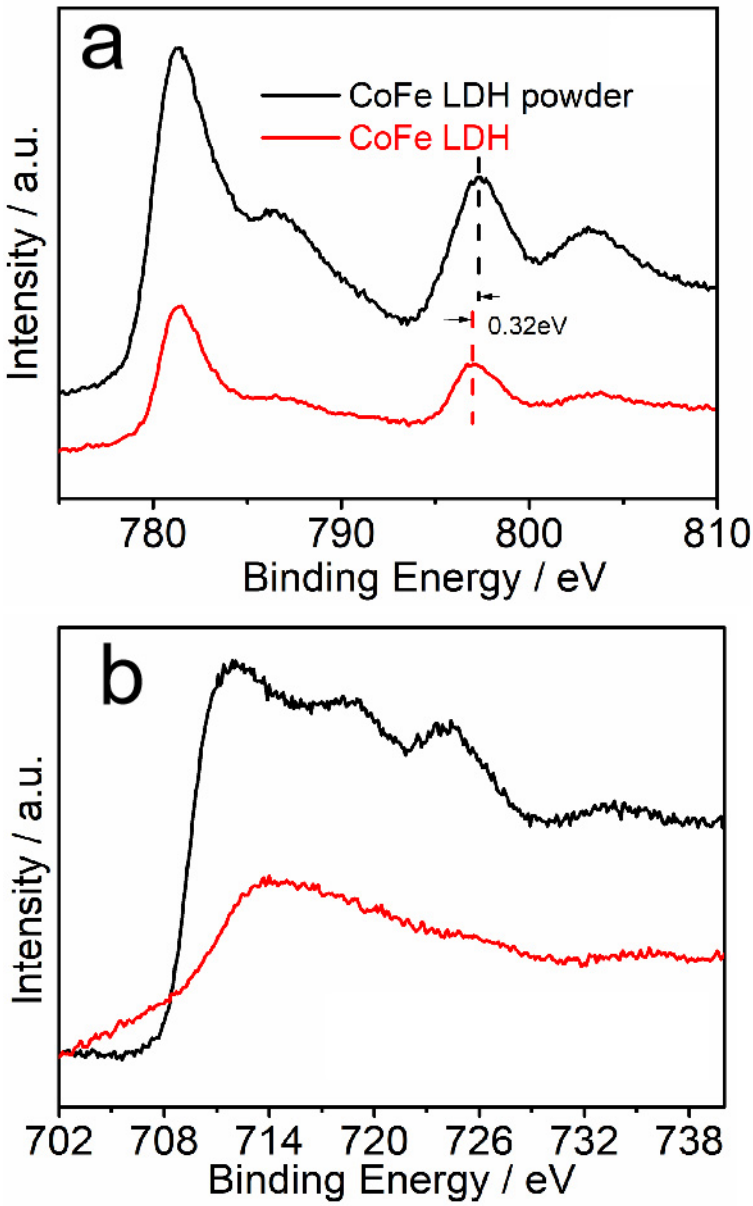

Figure.S13 The high-resolution (a) Co 2p and (b) Fe 2p spectra of CoFe LDH electrode and CoFe LDH powder. CoFe LDH powder sample was prepared according to our previously published article[16].

3. Supplementary Tables

Table S1. Summary of parameters for sample preparation.

Doping concentration	$Cr(NO_3)_3 \cdot 9H_2O$ (mmol)	$FeCl_2 \cdot 4H_2O$ (mmol)	$Co(NO_3)_2 \cdot 6H_2O$ (mmol)
0	0	1.25	2.5
5%	0.1875	1.0625	
10%	0.375	0.875	
15%	0.5625	0.6875	
20%	0.75	0.5	

Table S2. Comparison of the HER performance of CoFeCr LDH catalyst with other reported OER catalysts

Catalyst	Tafel slope ($mV\ dec^{-1}$)	Overpotential (η/mV) Vs. 10mA cm^{-2}	Electrolyte	Reference
Cr-CoFe LDH	**105**	**201**	**1 M KOH**	**This work**
Co/CoP	73.8	253	1 M KOH	*Advanced Energy Materials, 2017, 7, 1602355.*
Mesoporous Thin-Film NiS2	N/A	250	1 M KOH	*ACS Catalysis, 2020, 10, 15114–15122.*
NiFe-LDH/NF	58.9	210	1 M KOH	*Science, 2014, 345, 1593–1596.*
1T-MoS_2	56	270		*Int. J. Hydrogen Energy, 2021,46, 8377-8390*
$Zn_{1-x}Fe_x$–LDH/NF	110	221	1 M KOH	*Small, 2018, 14, 1803638.*
EG/$Co_{0.85}$Se/NiFe-LDH	160	265	1 M KOH	*Energy Environ. Sci., 2016, 9, 478-483.*
Co_9S_8@NOSC	105	320	1 M KOH	*Advanced Science, 2018, 5, 1700464*
$Ni_{1-x}Fe_x$-LDH	110	242	1 M KOH	*ACS Applied Materials & Interfaces, 2018, 10, 42453−42468.*
$NdBaMn_2O_{5.5}$	87	290	1 M KOH	*ACS Catalysis, 2017, 8, 364-371.*
CoMoV LDH/NF	182	270	1 M KOH	*Chemical Communications, 2019, 55, 3521- 3524.*
NiFe-LDH NS/DG10	210	300	1 M KOH	*Advanced Materials, 2017, 29, 1700017.*
CoP@BCN	52	215	1 M KOH	*Advanced Science, 2018, 5, 1700464.*
CoFe LDH-F	95	255	1 M KOH	*ACS Applied Materials & Interfaces, 2016, 8, 34474−34481.*
CoFeNiMo@NCNT	67.4	209.9	0.5 M H_2SO_4	*Applied Catalysis B: Environmental, 2021, 280, 119421.*
NiFe-LDH/$FeCoS_2$/CFC	157	308	1 M KOH	*Journal of Materials Science, 2020, 55, 16625–16640.*
Ni/NiS	123.3	230	1 M KOH	*Advanced Functional Materials, 2016, 26, 3314–3323.*

4. Notes and references.

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