Supplementary Information (SI) for Catalysis Science & Technology.
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Supporting information

Manipulating anion intercalation into layered double hydroxide for alkaline seawater oxidation at high current density†

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Experimental

1.1 Synthesis of NiFe LDH-CO_3^{2-}, NiFe LDH-F^-, and NiFe LDH-Cl^-

1 mmol $Ni(NO_3)_2 \cdot 6H_2O$ and 1 mmol $Fe(NO_3)_3 \cdot 9H_2O$ were dissolved in 35 mL of deionized (DI) water, add 10 mmol $CO(NH_2)_2$, NH_4F, and NaCl separately, and then the pretreated nickel foam (NF, 2 × 3 cm^2) was immersed in the solution, and kept at 120 ℃ for 6 h. After drying in air, nickel foam self-supporting NiFe LDH-CO_3^{2-}, NiFe LDH-F^-, and NiFe LDH-Cl^- nanosheets were obtained.

1.2 Materials characterization

Scanning electron microscopy (SEM, GeminiSEM 460), and transmission electron microscopy (TEM, JEM-2100) were conducted to characterize the morphologies and elements distribution of catalysts. X-ray diffraction (XRD) data was recorded on an X-ray diffractometer (Rigaku TTR-III) with a Cu Kα source. X-Ray photoelectron spectroscopy (XPS, K-Alpha) tests were conducted using Al source radiation.

1.3. Electrochemical measurements

An EnergyLab XM potentiostat (Solartron Analytical) was utilized for electrochemical measurements in a conventional three-electrode system. The working electrode, reference electrode, and counter electrode were the as-synthesized sample, Hg/HgO electrode, and carbon rod, respectively. For uniformity, all measurements were calibrated to the RHE using the formula $E_{RHE} = E_{Hg/HgO} + 0.098\ V + 0.059 \times pH$. Linear sweep voltammetry (LSV) curves with 95% iR compensation were documented at a scan rate of 2 mV s^{-1}. The electrochemical double-layer capacitance (C_{dl}) by implementing CV measurements with varying scan rates ranging from 20 to 100 mV/s in the non-faradaic region. The electrochemical impedance spectroscopy (EIS) was performed with a frequency from 100 kHz to 0.01 Hz.

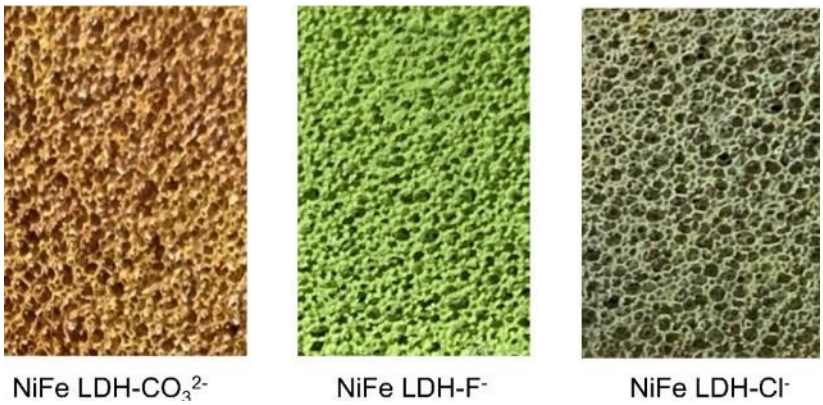

Fig. S1. The digital photographs of NiFe LDH-CO_3^{2-}, NiFe LDH-F^-, NiFe LDH-Cl^-.

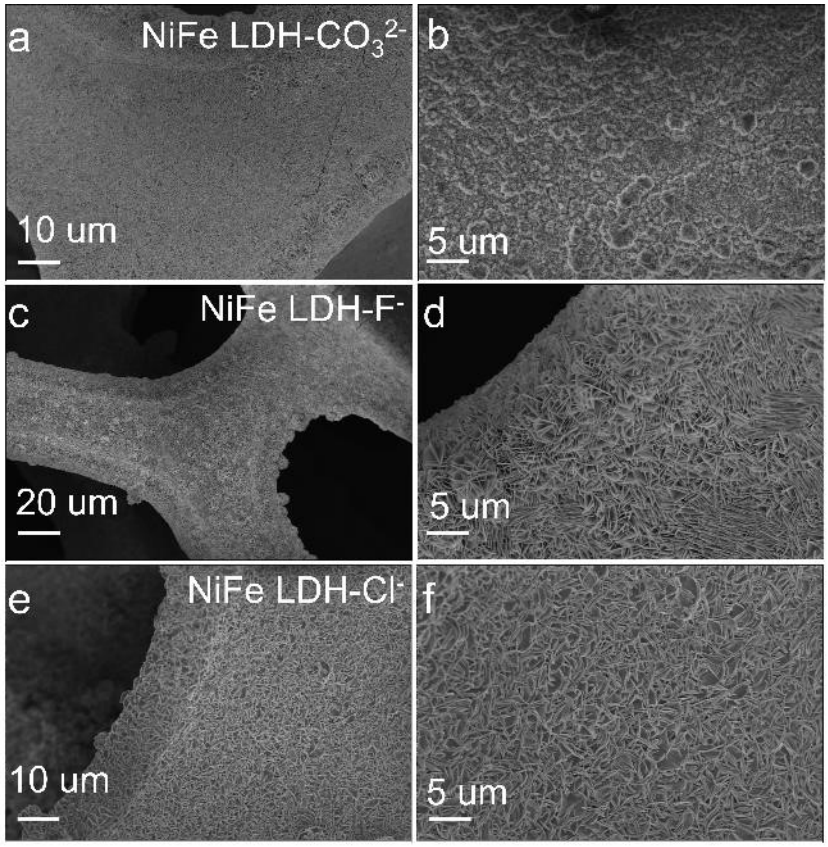

Fig. S2. Morphological characteristics of NiFe LDH on NF at low magnification: (a, b) NiFe LDH-CO_3^{2-}, (c, d) NiFe LDH-F^-, (e, f) NiFe LDH-Cl^-.

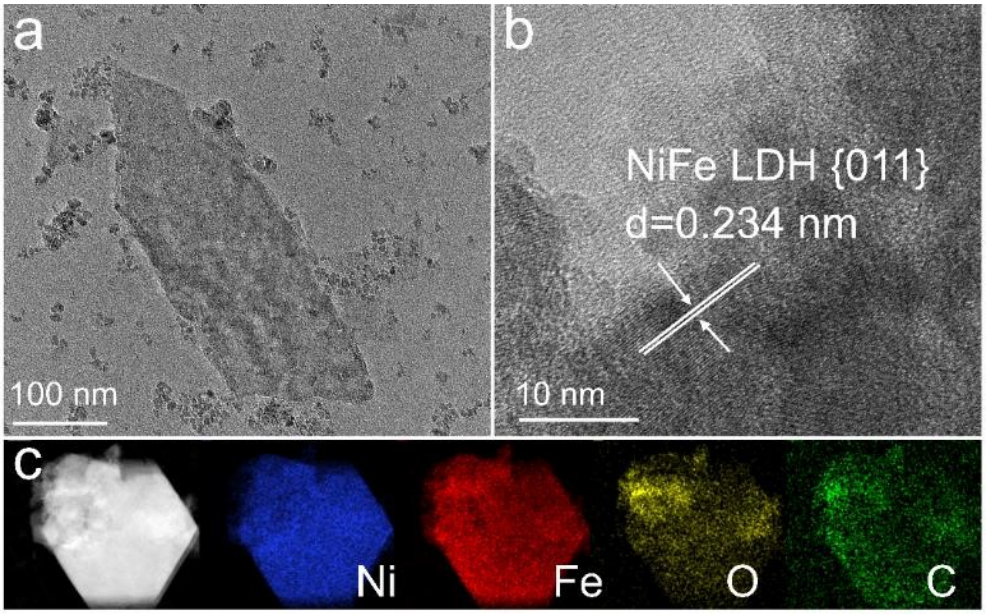

Fig. S3. a) TEM, b) HRTEM, and c) elemental mapping images of pristine NiFe LDH.

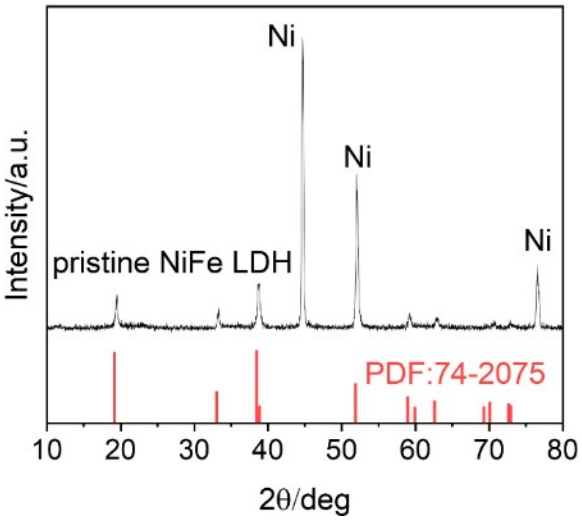

Fig. S4. XRD pattern of the pristine NiFe LDH.

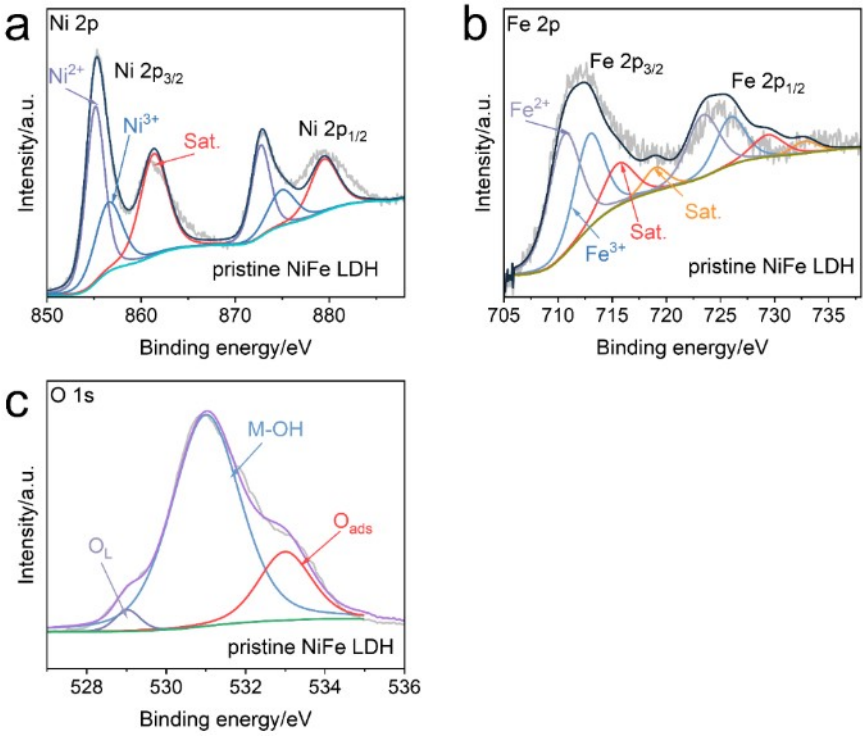

Fig. S5. High−resolution XPS spectra of (a) Ni 2p (b) Fe 2p (c) O 1s for pristine NiFe LDH.

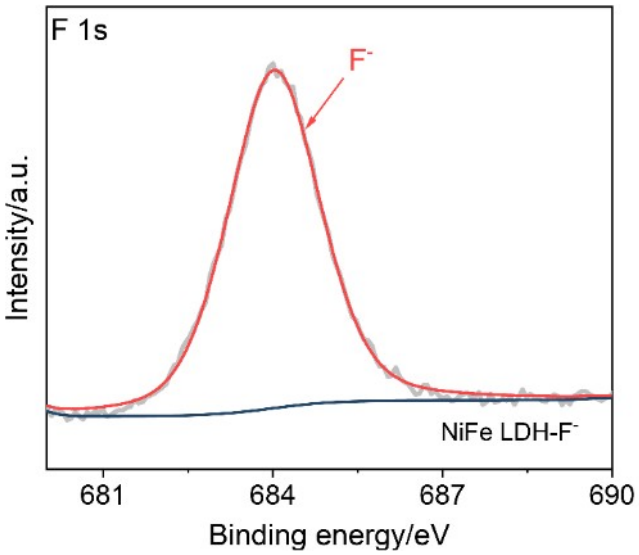

Fig. S6. F 1s XPS spectra for NiFe LDH-F^-.

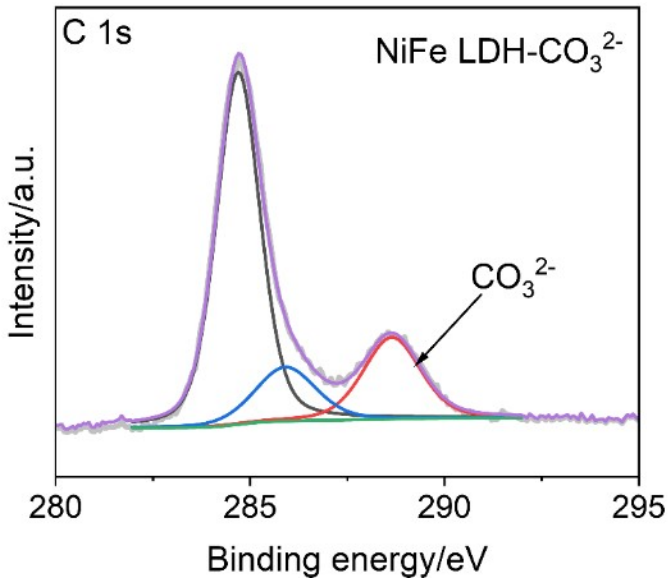

Fig. S7. C 1s XPS spectra for NiFe LDH-CO_3^{2-}.

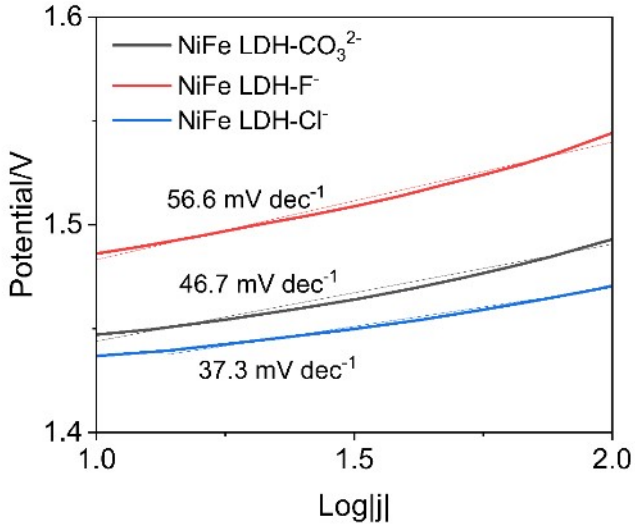

Fig. S8. Tafel plots of NiFe LDH-A (A=CO_3^{2-}, F^-, Cl^-).

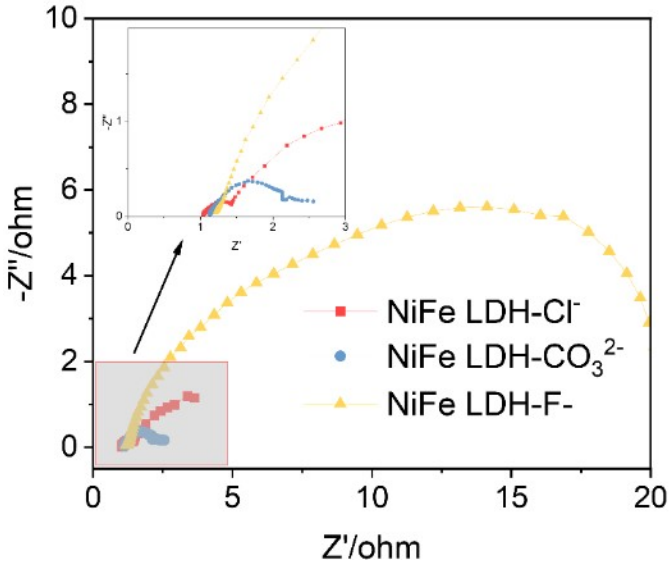

Fig. S9. Nyquist plots of of NiFe LDH-A (A=CO_3^{2-}, F^-, Cl^-).

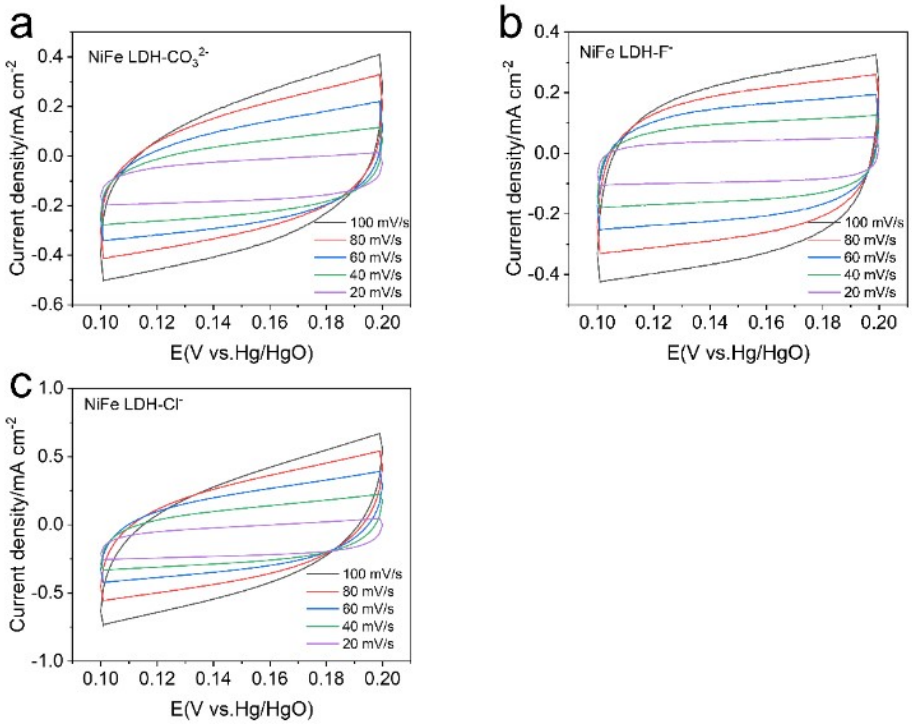

Fig. S10. CV curves in the non-Faradaic region (0.1~0.2 V vs Hg/HgO) with various scan rates (20-100 mV s^{-1}) for (a) NiFe LDH-CO_3^{2-}, (b) NiFe LDH-F^-, (c) NiFe LDH-Cl^-.

Table S1. Comparison of representative OER in alkaline simulated seawater or alkaline seawater.

Catalyst	j/mA cm^{-2}	η/mV	Stability/h	Electrolyte	Refs
NiFe LDH(NaCl)	100	240	N/A	1 M KOH	This work
	200	255	200		
	500	300	N/A		
	500	330	N/A	0.5 M NaCl+1 M KOH	
	500	325	N/A	1 M NaCl+1 M KOH	
	500	310	N/A	2 M NaCl+1 M KOH	
	200	350	100	seawater+1 M KOH	
NiFe LDH_CO_3^{2-}	100	237	200	1 M KOH +0.5 M NaCl	1
	200	264			
	500	302			
	1000	351			
	500	N/A	1000	1 M KOH + 0.5 M NaCl + 1 M Na_2CO_3	
	1000	N/A			
NiIr-LDH	100	286	N/A	1M KOH + 0.5 M NaCl	2
$Fe(OH)_3$-$Ni(SO_4)_{0.3}(OH)_{1.4}$-$Ni(OH)_2$	100	268	N/A	1 M KOH + 0.5 M NaCl	3
	200	283	500		

	400	290	N/A		
$CoFe\text{-}Ni_2P$	100	246	140	1 M KOH + 0.5 M NaCl	4
	500	360	510	1 M KOH + Seawater	
B-$MnFe_2O_4$@MFOC	100	333	100	1 M KOH + 0.5 M NaCl	5
	500	N/A			
	100	405		1 M KOH + Seawater	
NiTe@FeOOH	100	280	100	Alkaline Seawater	6
	500	328	N/A		
Co_xP_v@NC	500	323	N/A	1 M KOH + Seawater	7
	800	N/A	100		
Ni_xCr_yO	100	370	275	1 M KOH + Seawater	8
	500	460	100		
Cr-Co_xP	100	334	140	1 M KOH + Seawater	9
	500	392	N/A		
	1000	423	N/A		

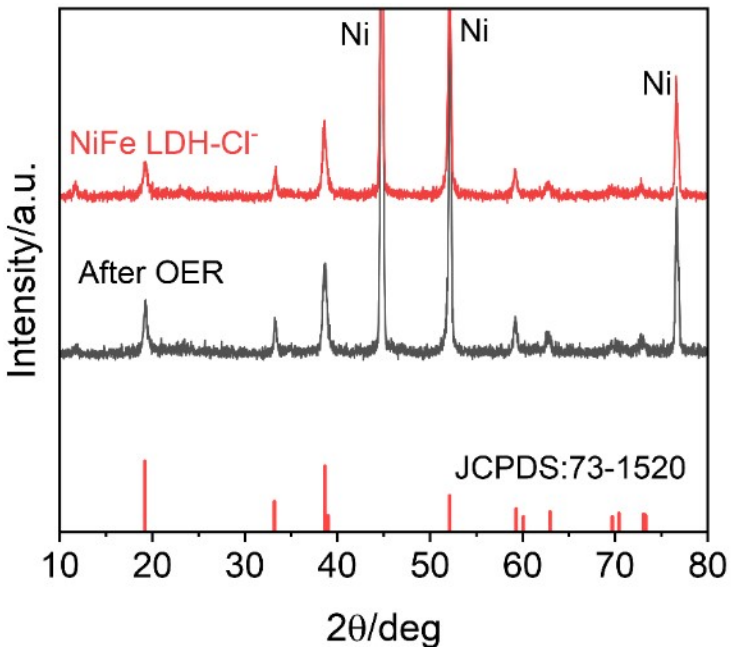

Fig. S11. XRD patterns of NiFe LDH-Cl^- before and after OER test.

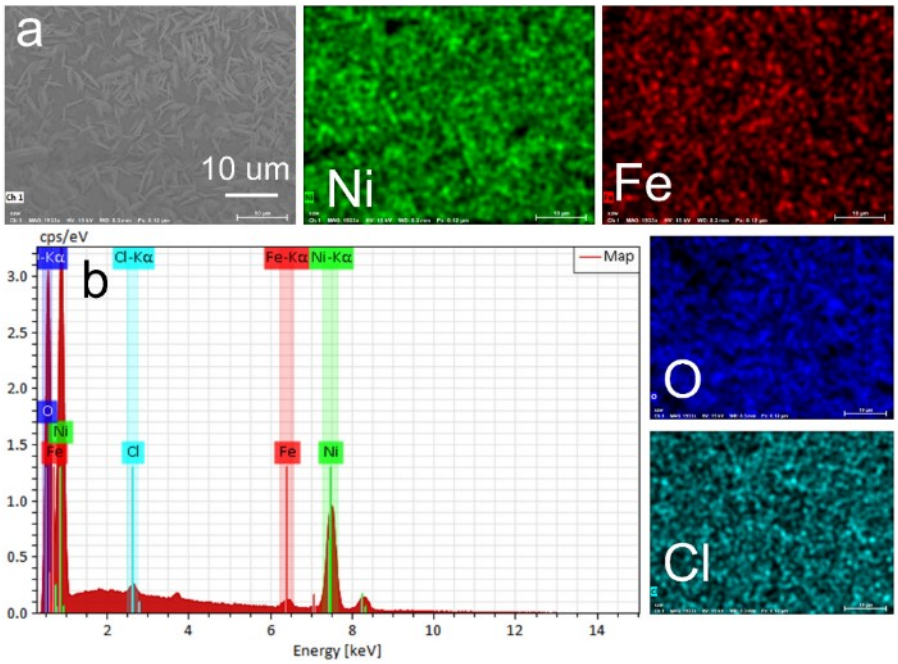

Fig. S12. EDS elemental mapping images of NiFe-LDH-Cl^- after OER test.

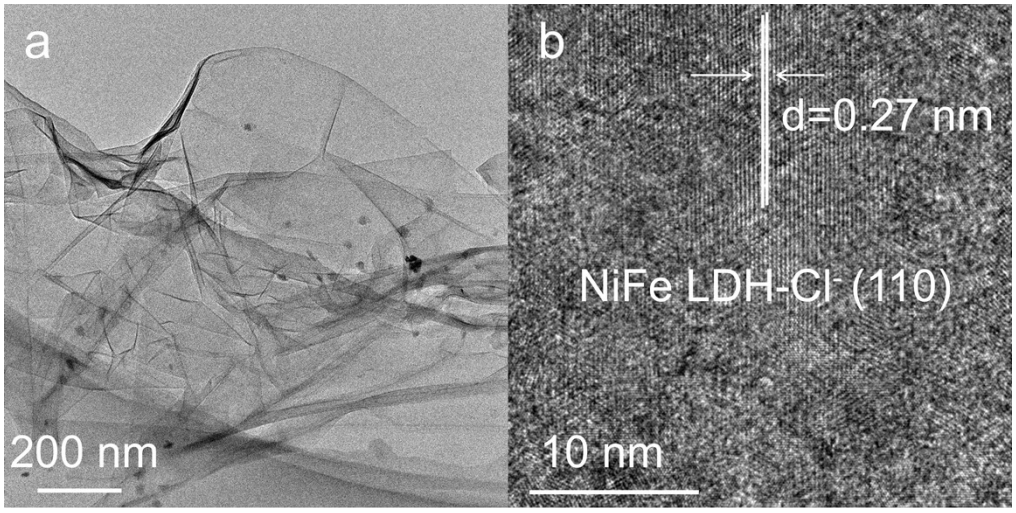

Fig. S13. (a) TEM and (b) HRTEM images of NiFe LDH-Cl^- after OER test.

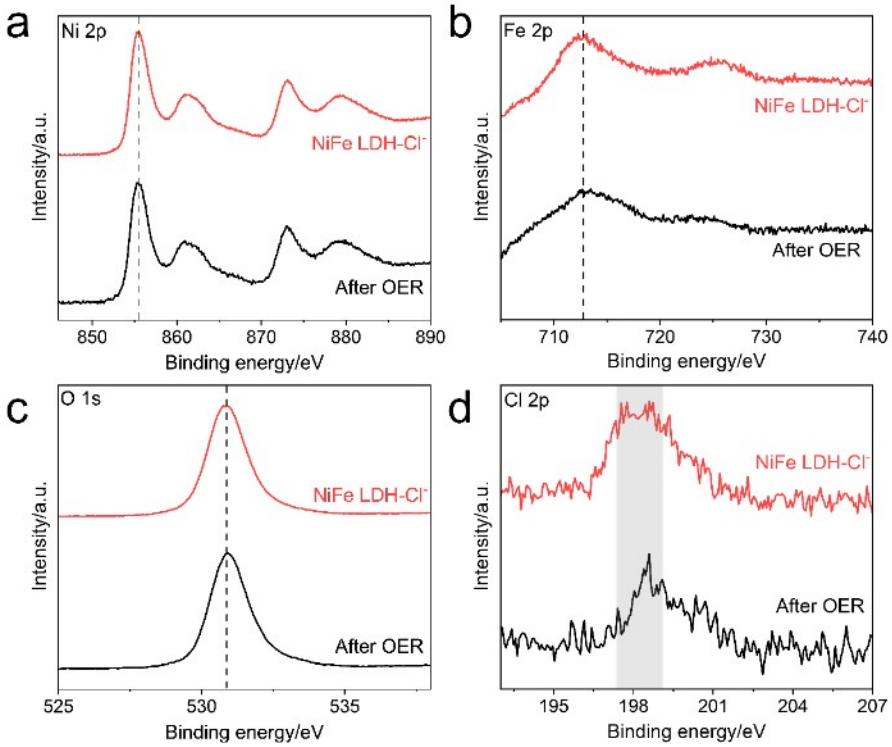

Fig. S14. (a) Ni 2p, (b) Fe 2p, (c) O 1s, and (d) Cl 2p XPS spectra of NiFe LDH-Cl^- before and after OER test.

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