SUPPORTING INFORMATION

Methanol-assisted synthesis of Ni^{3+}-doped ultrathin NiZn-LDH nanomeshes for boosted alkaline water splitting

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Electrochemical active surface area (ECSA) calculation:

The ECSA of the electrode is estimated from the double-layer capacitance (C_{dl}) of the catalyst according to the following equation:[1-4]

$$\text{ECSA} = C_{dl} / Cs \quad (1)$$

Where, C_s is the specific capacitance of a flat standard electrode. In this work, we give C_s a value of 40 μF cm^{-2}.

The C_{dl} can be calculated by linearly fitting at 0.1 V vs scan rate. The C_{dl} can be calculated according to the following equation:

$$C_{dl} = j / v \quad (2)$$

Where, j is the difference of the anodic and cathodic current densities with no-Faradaic and v is the different scan rate

Take the OER performance of Ni(II, III)Zn-LDH/NF-nm as an example (Slope = 34.7 mF cm^{-2} i.e. C_{dl} = 4.164 mF)

ECSA = C_{dl} / Cs = 4.164 / 0.04 = 52.05 cm^{-2}

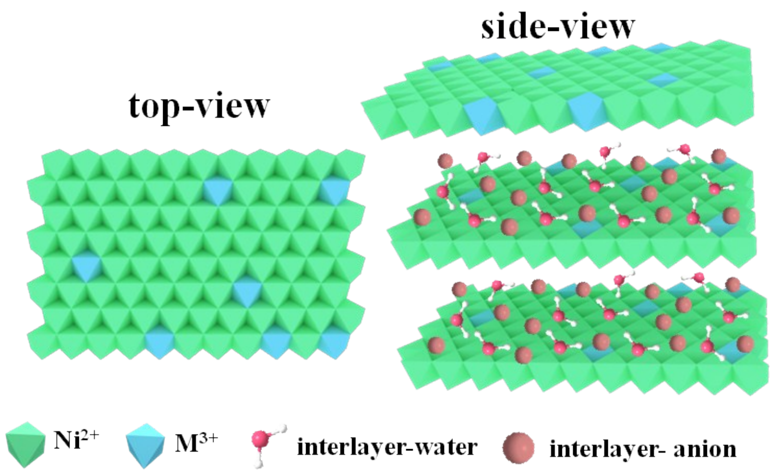

Figure S.1 Schematic diagram of the crystalline structure of Ni(II)-based LDH.

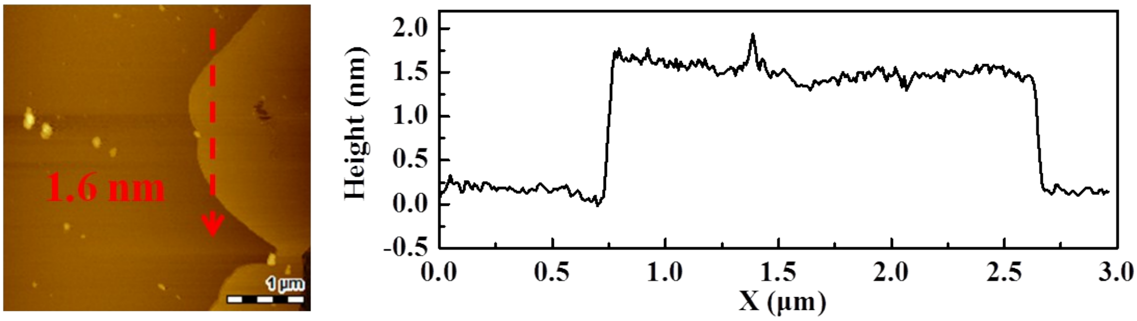

Figure S.2 AFM image and the height profile of Ni(II, III)Zn-LDH/NF-nm.

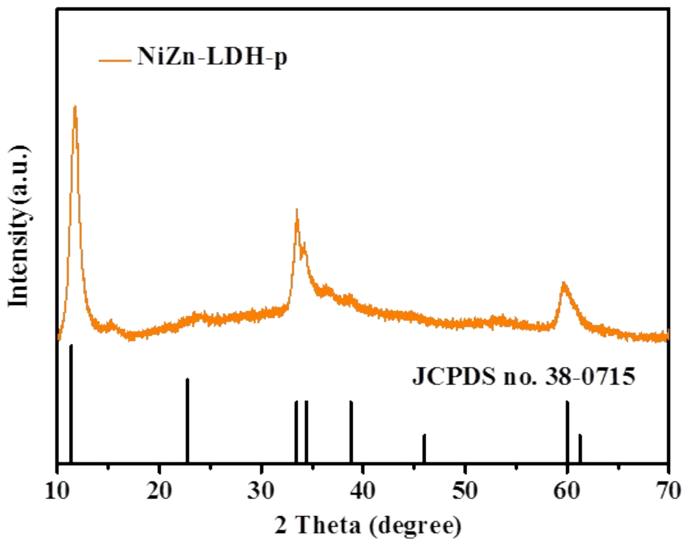

Figure S.3 XRD patterns of NiZn-LDH-p.

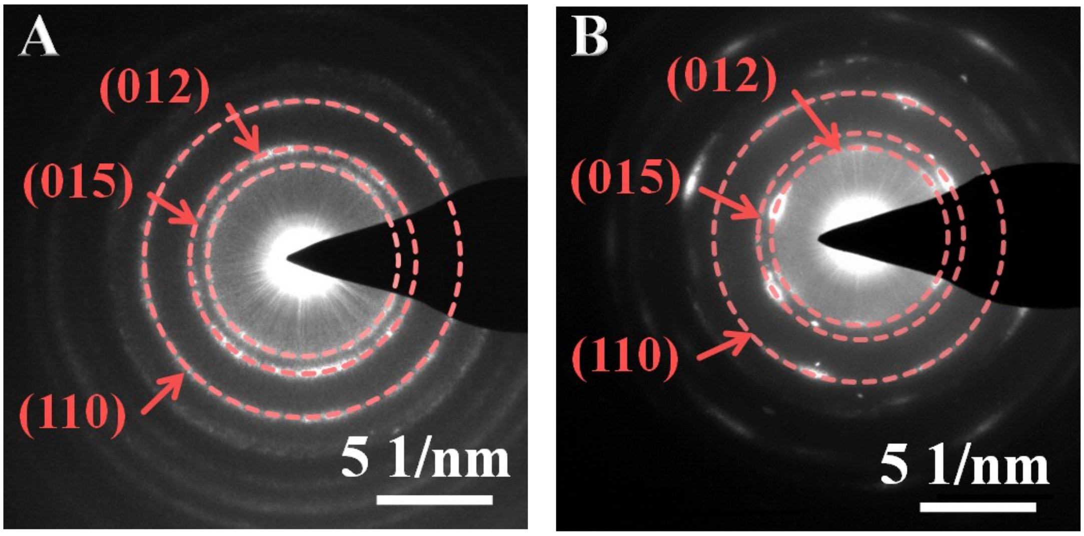

Figure S.4 SAED patterns of the NiZn-LDH-p (A) and the Ni(II, III)Zn-LDH/NF-nm (B).

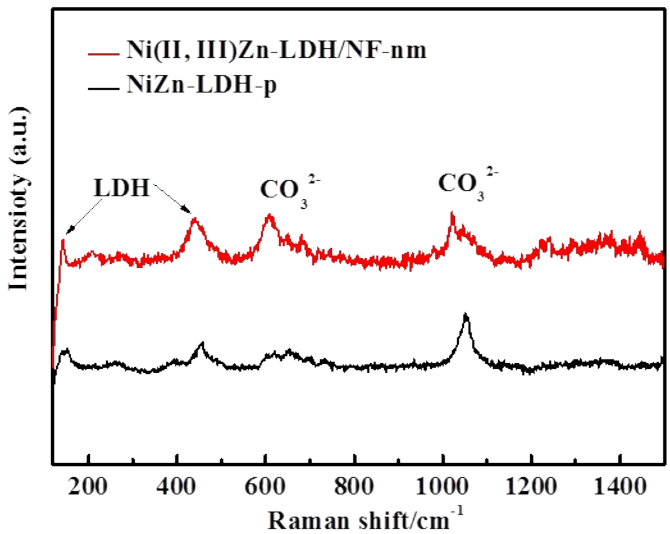

Figure S.5 Raman spectra of Ni(II, III)Zn-LDH/NF-nm and NiZn-LDH-p.

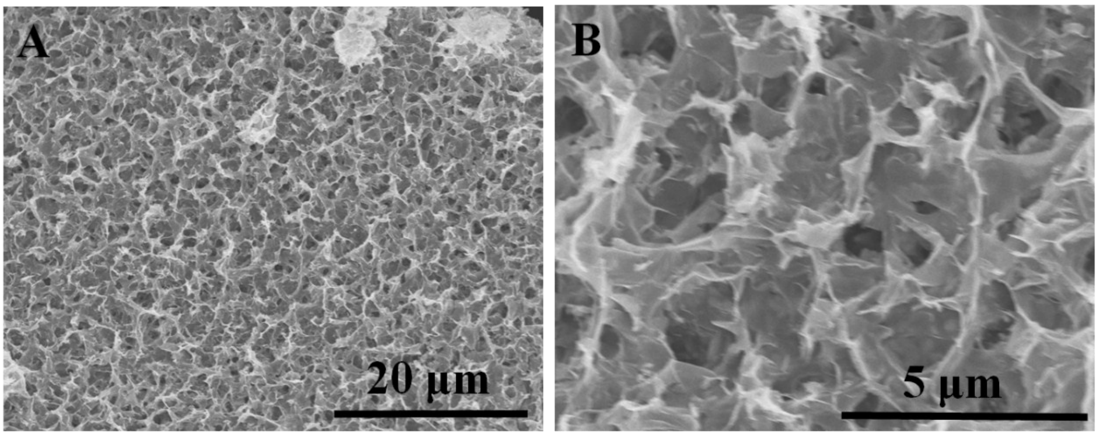

Figure S.6 SEM images of the NiZn-LDH/NF-ns.

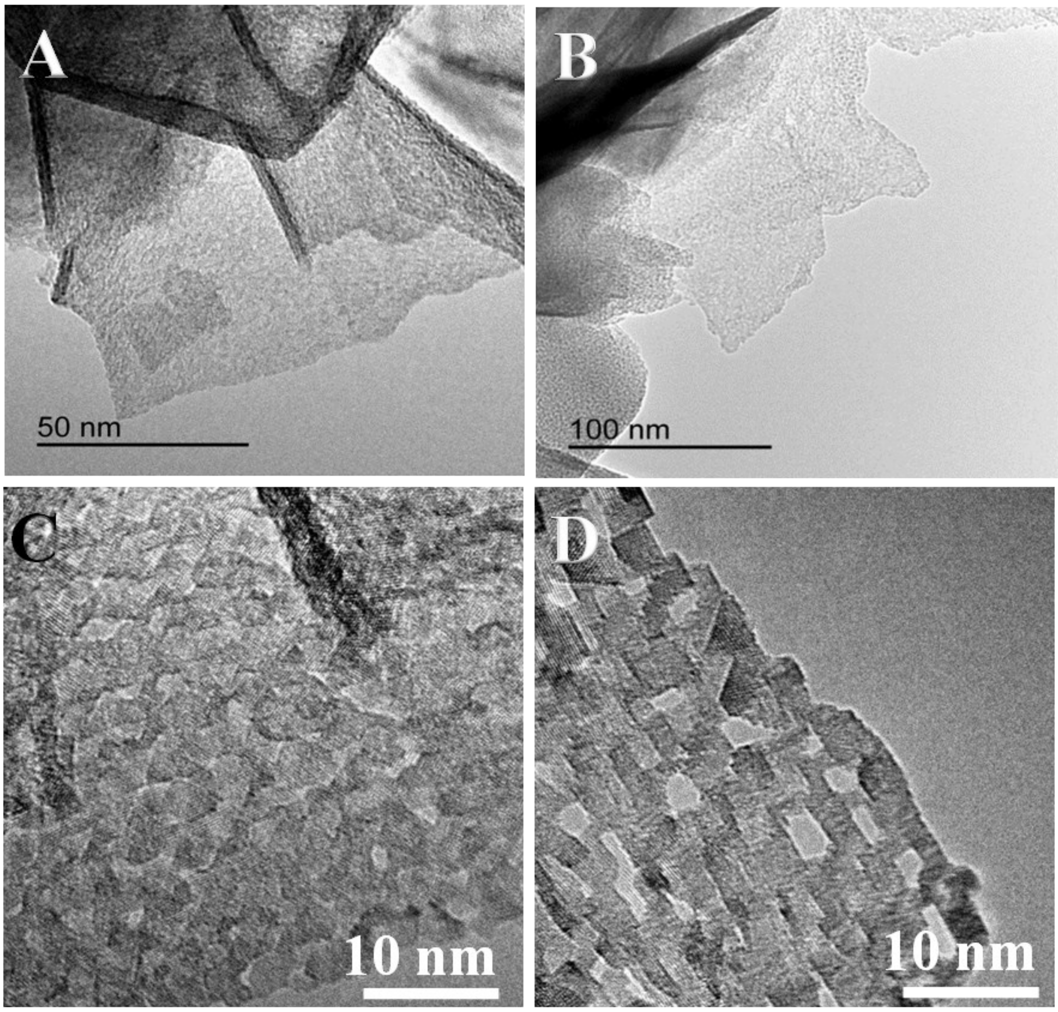

Figure S.7 HRTEM pattern of the NiZn-LDH/NF-ns (A, C) and Ni(II, III)Zn-LDH/NF-nm (B, D).

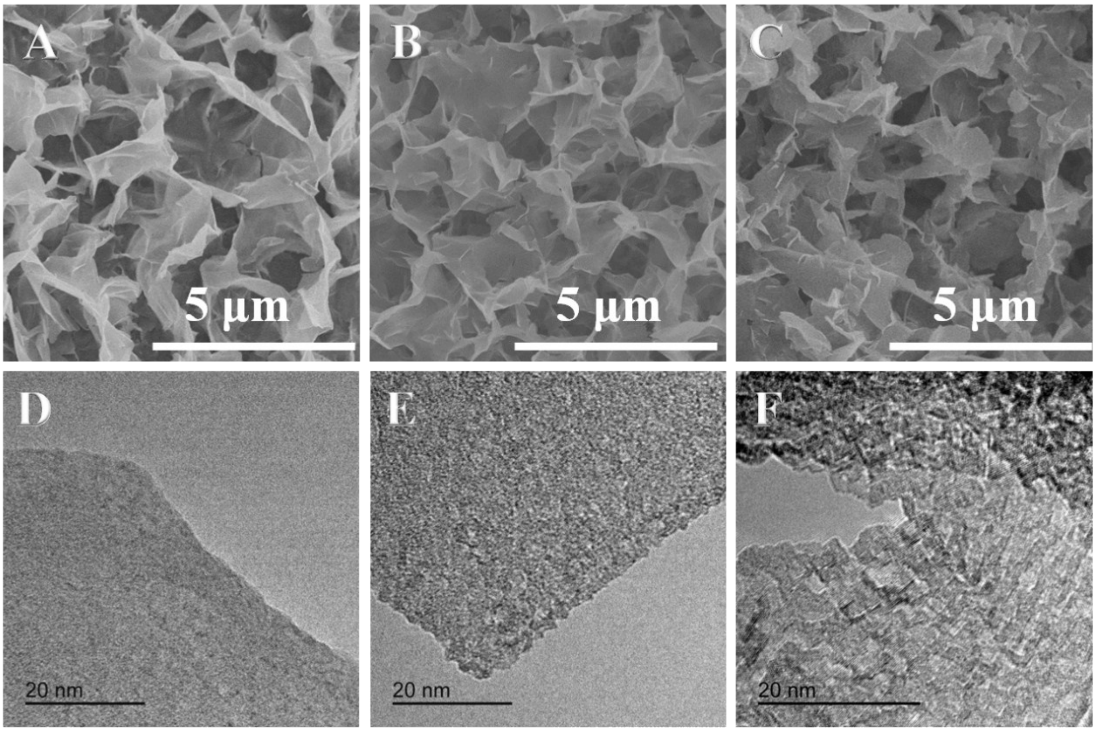

Figure S.8 SEM images (A-C) and TEM images (D-F) of the Ni(II, III)Zn-LDH/NF-nm samples prepared at the reaction times of 5 h, 13 h and 21 h, respectively.

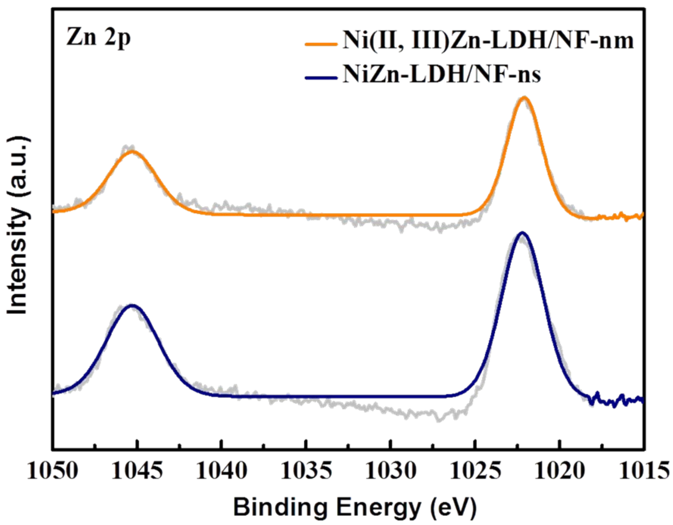

Figure S.9 Zn 2p XPS spectra of Ni(II, III)Zn-LDH/NF-nm and NiZn-LDH/NF-ns.

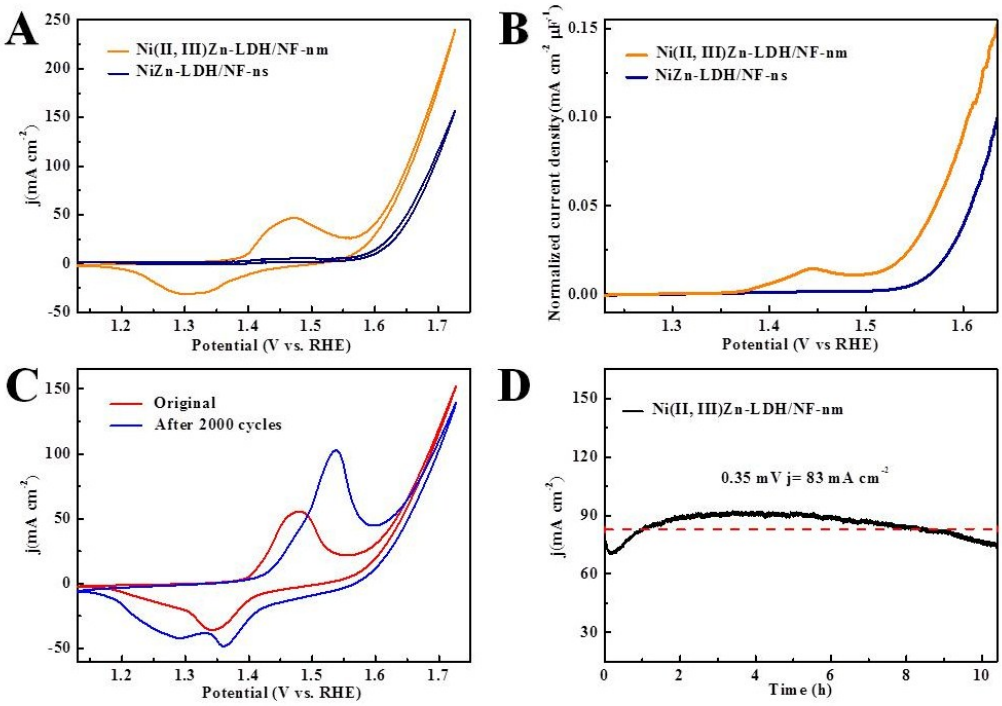

Figure S.10 (A) Cyclic voltammetry (CV) curves of the Ni(II, III)Zn-LDH/NF-nm and the NiZn-LDH/NF-ns; The normalized LSV curves of the OER (B); (C) Comparative CV curves before and after 2000 cycles; (D) Long-term electrochemical OER durability test for the Ni(II, III)Zn-LDH/NF-nm.

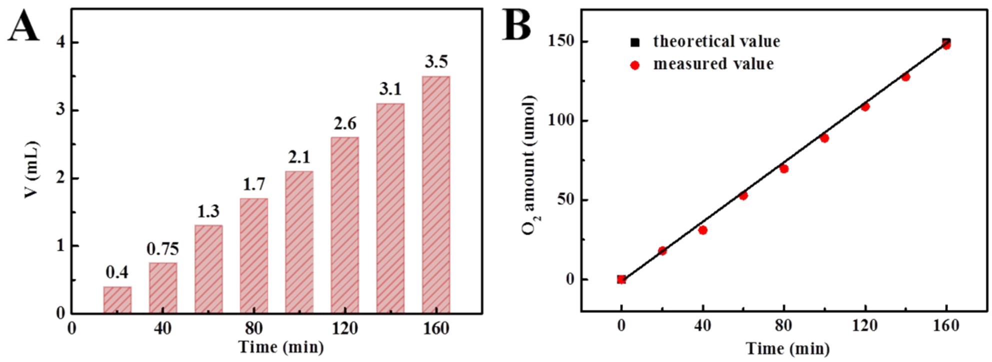

Figure S.11 (A) The volume of the oxygen-produced gas from the OER of Ni (II, III) Zn-LDH / NF-nm was recorded every 20 minutes; (B) The oxygen generation efficiency of Ni (II, III) Zn-LDH / NF-nm OER when current density is maintained at 50 mA cm^{-2}.

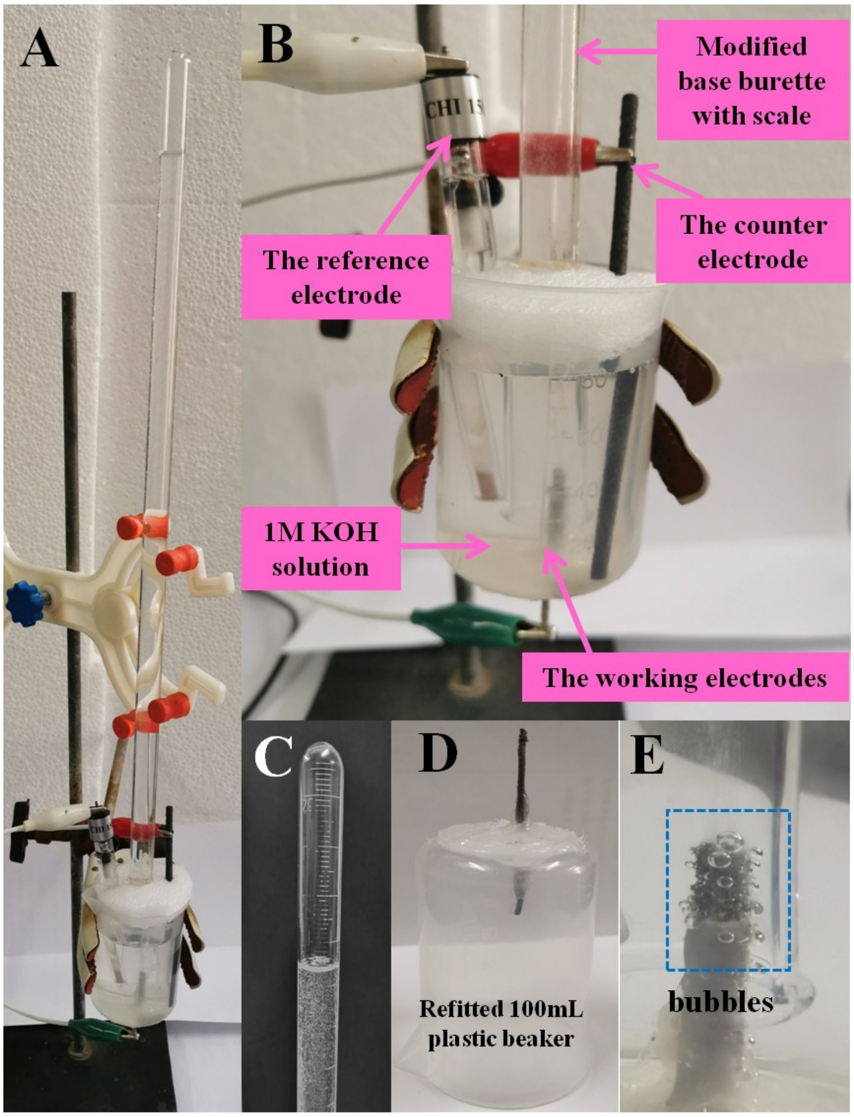

Figure S.12 (A) Optical image of the quantitative oxygen or hydrogen evolution device under potentiostatic electrolysis; (B) the corresponding instruments for the quantitative OER or HER analysis; (C) the enlarged modified base buret for collecting the evolved gas; (D) Refitted 100 mL plastic beaker for storing 1 M KOH solution; and (E) the bubbles evolved on the electrode.

In order to make the quantitative analysis of the OER or HER during the experiments, we built a device for measuring the evolved gas amount, as shown in Figure S.12. Taking the OER as an example, firstly, we collected O_2 gas by water drainage method after the OER have been initiated for ~40 min when the soluble O_2 gas is saturated in water. Then, we calculated the moles of O_2 generated from OER with an ideal gas law and the theoretical amount of O_2 evolved by using the Faraday law, respectively. Note that the passage of 96485.4 C charge causes 1 equivalent of reaction. Furthermore, the Faradic efficiency of the as-prepared catalysts for the OER or HER can be determined by the ratio of the actually evolved O_2 or H_2 amount, respectively, during the electrocatalysis to the theoretically evolved O_2 or H_2 amount.

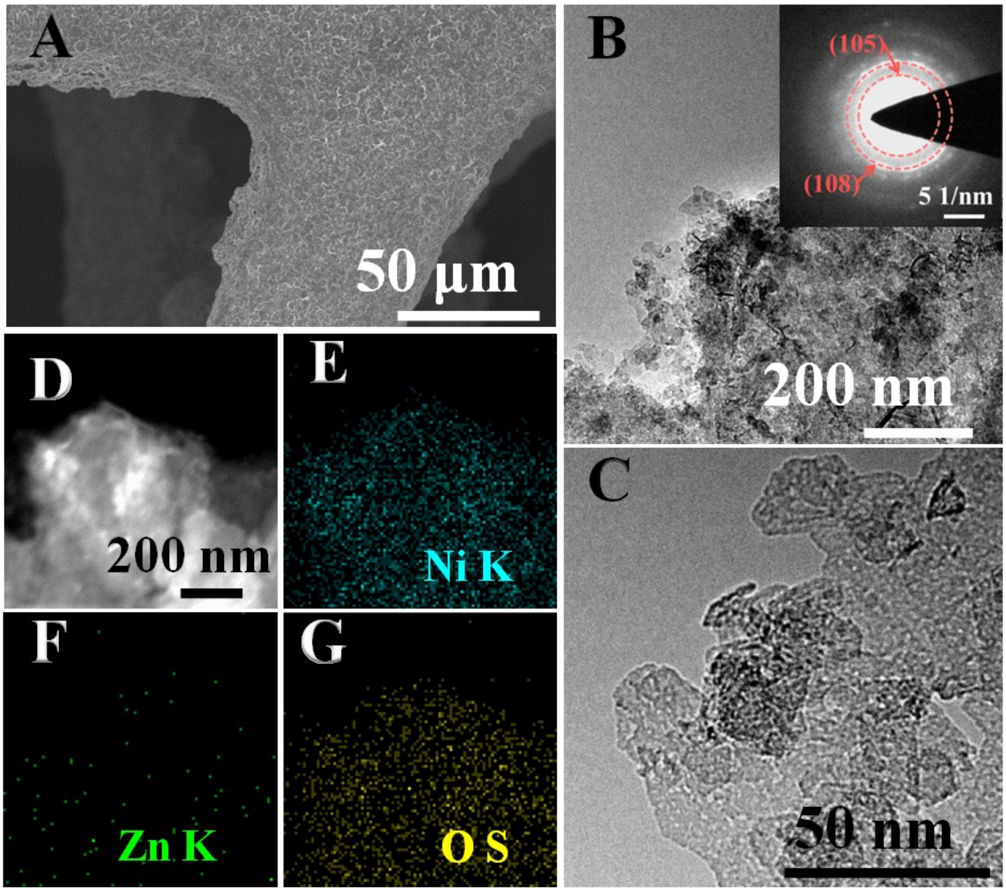

Figure S.13 (A) SEM and (B,C) TEM imagesof the Ni(II, III)Zn-LDH/NF-nm after OER test; (D-G): elemental mapping analysis images; Insert in (B): SAED pattern of Ni(II, III)Zn-LDH/NF-nm after OER test.

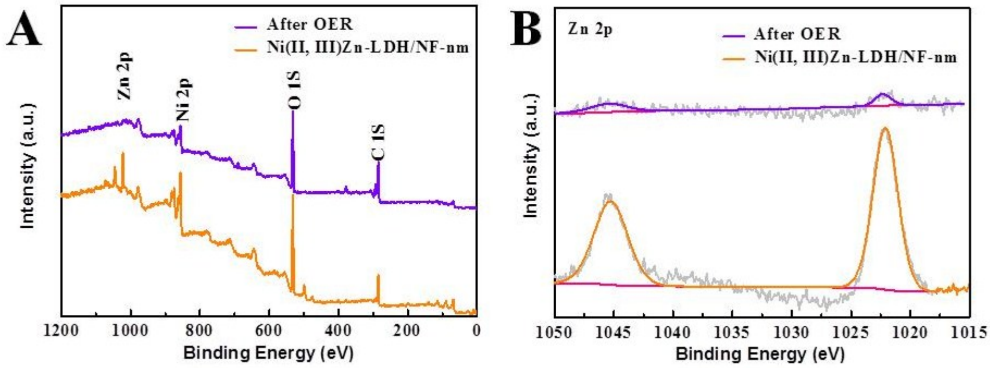

Figure S.14 (C) XPS survey spectra and (D) Zn 2p spectra of Ni(II, III)Zn-LDH/NF-nm before and after OER test.

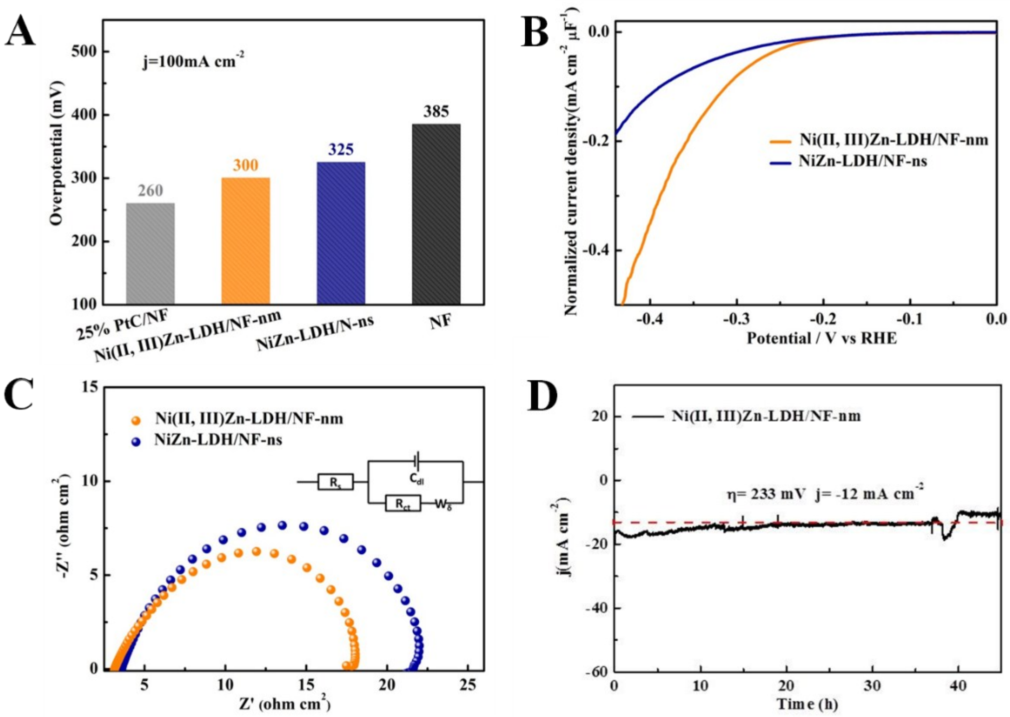

Figure S.15 (A) Overpotential of HER comparison of various catalysts at 100 mA cm^{-2}; The normalized LSV curves of the HER (B); (C) HER Nyquist plots of catalysts (inset shows the equivalent circuit used); Long-term electrochemical HER durability test for the Ni(II, III)Zn-LDH/NF-nm.

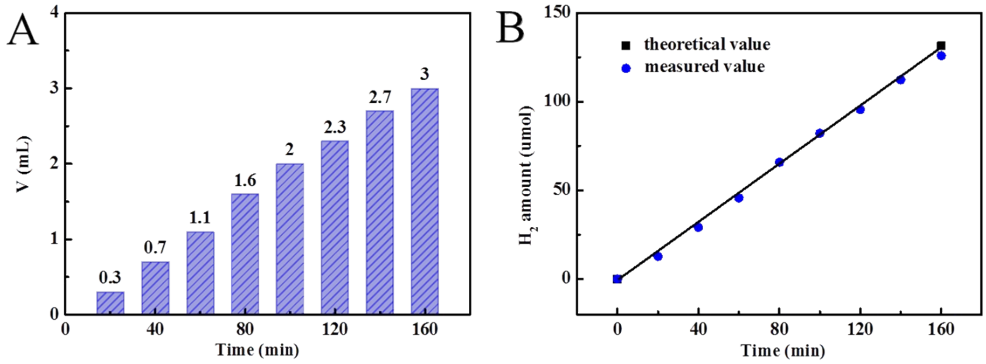

Figure S.16 (A) The volume of the hydrogen-produced gas from the HER of Ni (II, III) Zn-LDH / NF-nm was recorded every 20 minutes; (B) Hydrogen production efficiency for HER under when the i-t measurements were performed at 22 mA cm^{-2}.

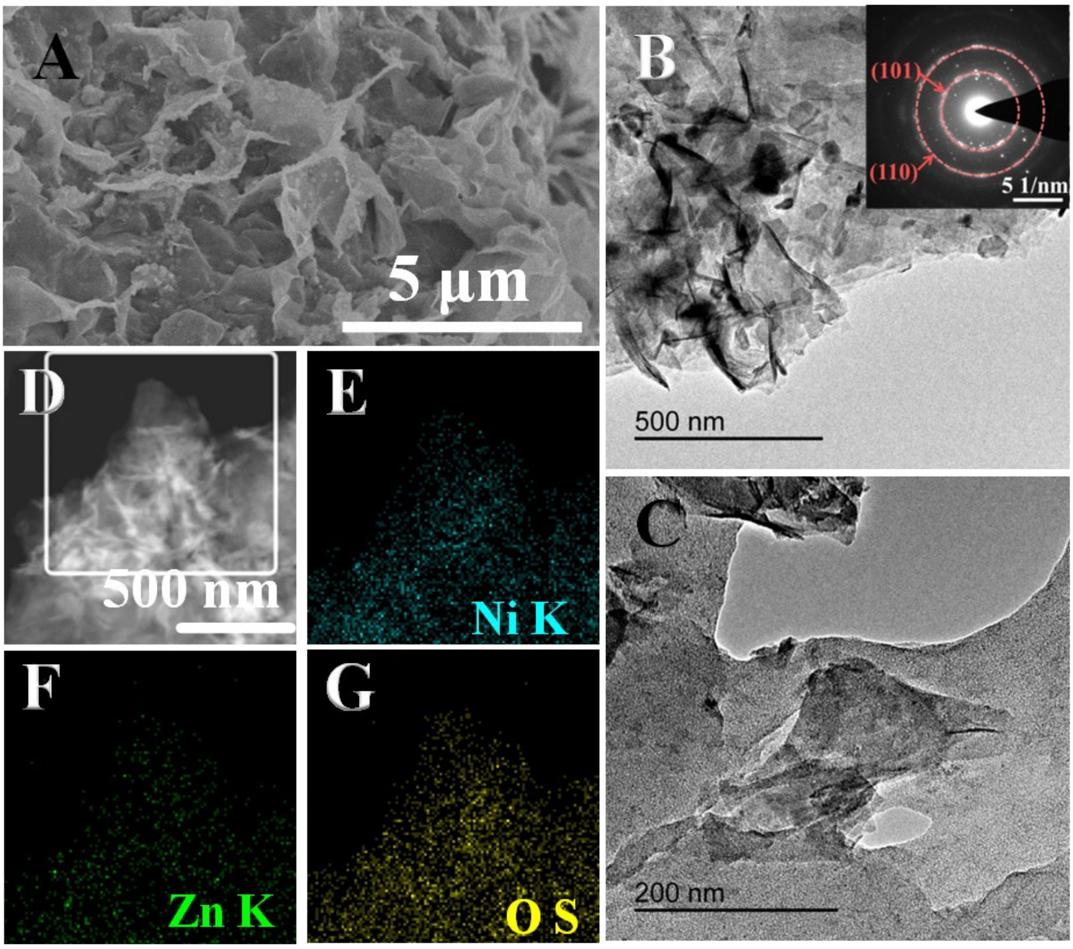

Figure S.17 SEM (A) and TEM images (B,C)of Ni(II, III)Zn-LDH/NF-nm after HER. (D-G) Elemental mapping images of Ni(II, III)Zn-LDH/NF-nm after HER. Insert in (B): SAED pattern of Ni(II, III)Zn-LDH/NF-nm after HER.

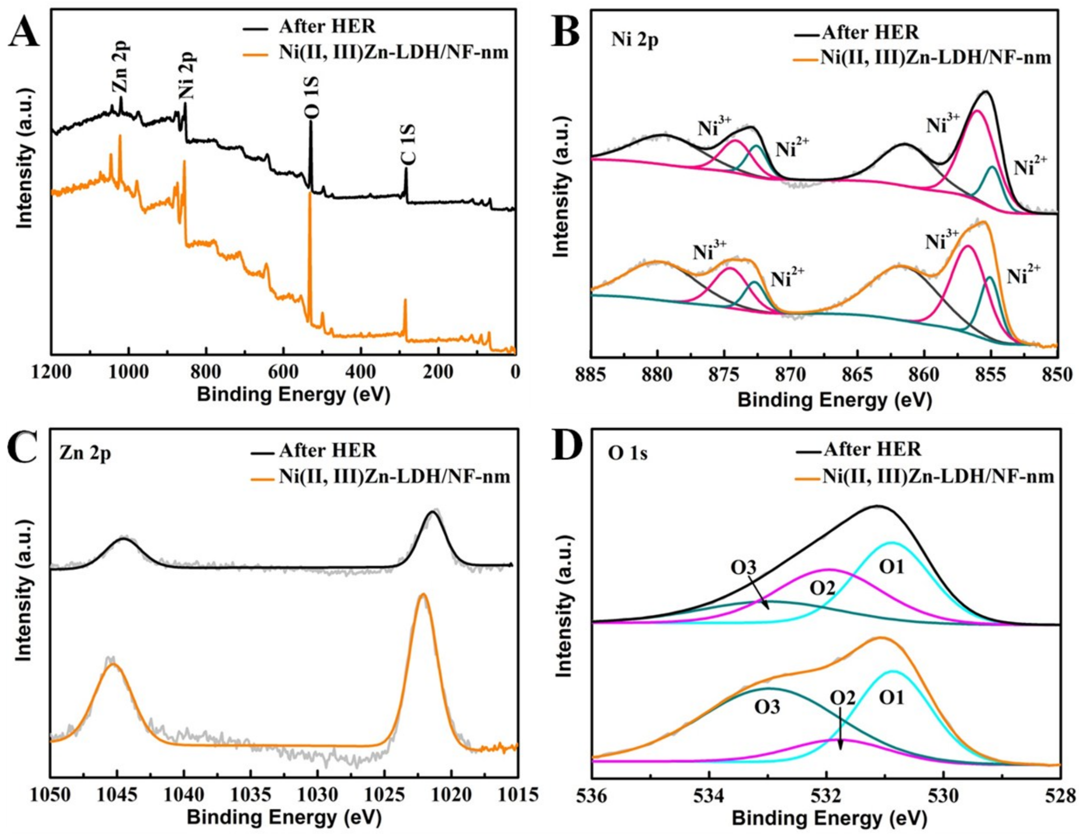

Figure S.18 XPS spectra of Ni(II, III)Zn-LDH/NF-nm and Ni(II, III)Zn-LDH/NF-nm after HER. (A) survey spectra. (B) Ni 2p. (C) Zn 2p. (D) O 1s.

Table S.1 Comparison of the charge transfer resistance (Rt) and internal resistance (Rs) of Ni(II, III)Zn-LDH/NF-nm and NiZn-LDH/NF-ns.

Nyquist plots of OER	Ni(II, III)Zn-LDH/NF-nm	NiZn-LDH/NF-ns
Rs	2.74	2.94
Rct	1.13	7.73

Table S.2 Comparison of the electrochemically active surface area (ECSA) of OER for Ni(II, III)Zn-LDH/NF-nm and NiZn-LDH/NF-ns.

Results of analysis	Ni(II, III)Zn-LDH/NF-nm	NiZn-LDH/NF-ns
ECSA	52.05 cm^{-2}	31.95 cm^{-2}

Table S.3 Comparison of the charge transfer resistance (Rt) and internal resistance (Rs) of Ni(II, III)Zn-LDH/NF-nm and NiZn-LDH/NF-ns.

Nyquist plots of HER	Ni(II, III)Zn-LDH/NF-nm	NiZn-LDH/NF-ns
Rs	3.49	3.96
Rct	13.13	17.14

Table S.4 Comparison of the electrochemically active surface area (ECSA) of HER for Ni(II, III)Zn-LDH/NF-nm and NiZn-LDH/NF-ns.

Results of analysis	Ni(II, III)Zn-LDH/NF-nm	NiZn-LDH/NF-ns
ECSA	15.75 cm^{-2}	7.80 cm^{-2}

Table S.5 Comparison of the OER catalytic performance of Ni(II, III)Zn-LDH/NF-nm with other recently reported hydroxide-based electrocatalysts in alkaline solution.

Catalyst	Overpotential (mV, at 10 mA cm^{-2})	Overpotential (mV, at 100 mA cm^{-2})	mass loading (mg cm^{-2})	Reference
Ni(II, III)Zn-LDH/NF-nm	**≈150**	**=320**	**0.107**	***This work***
oxygen-doped NiFe-LDH	**-**	**≈370**	**0.28**	***ACS Sustainable Chem. Eng. 2019, 7, 4247–4254***
$Ni_{0.83}Fe_{0.17}(OH)_2$	**=245**	**-**	**0.204**	***ACS Catal. 8, 2018, 5382-5390***
NiV-LDH/NF	**-**	**=391**	**0.71**	***Nanoscale, 2019, 11, 8855-8863***
1.0 h-$Ni(OH)_2$	**-**	**≈400**	**0.687**	***Green Chem., 2019, 21, 578-588***
ES-CoAl-LDH	**=270**	**=320**	**5**	***Dalton Trans., 2019, 48, 5214-5221***
Co_4Fe_2-LDHs/$Co(OH)_2$-NWs	**=220**	**=231**	**-**	***Chem. Commun., 2019, 55, 4218-4221***
ZnNiNGr	**=290**	**-**	**-**	***ACS Appl. Energy Mater, 2018, 1, 5500-5510***
$Ni_3V_1Fe_1$ LDH	**=269**	**≈410**	0.28	***Inorg. Chem. Front., 2019, 6, 1890***
β-$Ni(OH)_2$ nanomeshes	**-**	**≈360**	**0.285**	***Adv. Mater. 2017 29, 1604765.***
NiCo-Fe1.0	**=283**	**≈350**	**0.283**	***ChemSusChem, 2019, 10.1002/cssc.201901364***
holey $Ni(OH)_2$	**=335**	**≈370**	**0.204**	***small 2017, 13, 1700334***
$Ni_{0.75}Fe_{0.125}V_{0.125}$-LDHs/NF	**=231**	**≈270**	**1.42**	***Small, 2018, 14, 4724***
water-plasma exfoliated CoFe LDH/NF	**=232**	**≈360**	**0.408**	***Adv. Mater. 2017, 29, 1701546***
σ-FeOOH NSs/NF	**=265**	**≈470**	**0.16**	***Adv. Mater. 2018, 1803144***
NiFe(II,III)-LDH	**=220**	**=307**	**2**	***Small 2019, 1902551***
NiFeRu-LDH	**=225**	**≈250**	**-**	***Adv. Mater. 2018, 30，1706279***
NiLa-LDH NSAs	**=209**	**≈321**	**-**	***Journal of Energy Chemistry, 2019, 33, 125-129***
$CoFe_2O_4$ NSs	**=275**	**-**	**0.36**	***J. Mater. Chem. A, 2019, 7, 7328-7332***
oxygen-enriched NiFe-	**=310**	**≈370**	**0.204**	***Adv. Mater., 2017, 29,***

LDH nanosheets				***1701546***
NiFe-$NiCoO_2$	**=286**	**≈383**	**0.6**	***Journal of Energy Chemistry, 2019, 33, 74-80***
PA-$CoS_x(OH)_y$	**=261**	**-**	**0.400**	***J. Mater. Chem. A,2018, 6,24311–24316 |***
Co_1Mn_1CH	**-**	**=322**	**5.6**	***J. Am. Chem. Soc., 2017, 139, 8320-8328***
V_{Ni}-α-$Ni(OH)_2$	**=308**	**≈383**	**0.142**	***ACS Energy Lett. 2018, 3, 1373−1380***

Table S.6 Comparison of the HER catalytic performance of Ni(II, III)Zn-LDH/NF-nm with other recently reported hydroxide-based electrocatalysts in alkaline solution.

catalyst	Overpotential (mV, at 10 mA cm^{-2})	Overpotential (mV, at 100 mA cm^{-2})	Mass loading (mg cm^{-2})	Reference
Ni(II, III)Zn-LDH/NF-nm	**=183**	**=300**	**0.107**	***This work***
NiFe(II,III)-LDH	**=120**	**≈310**	**2**	***Small 2019, 1902551***
NiV-LDH/NF	**-**	**=333**	**0.71**	***Nanoscale, 2019, 11, 8855-8863***
$Fe_{1.5}Ni_{0.5}OOH$/EGSI	**=228**	**≈310**	**-**	***ChemistrySelect, 2019, 4, 2153-2159***
NiFeLDH@NiCoP/NF	**≈140**	**≈320**	**2**	***Adv. Funct. Mater., 2018, 28, 1706847***
σ-FeOOH NSs/NF	**=108**	**≈180**	**0.16**	***Adv. Mater. 2018, 1803144***
NiFe-LDHs/NF	**=145**	**≈300**	**1.42**	***Small, 2018, 14, 4724***
$Ni_{0.75}Fe_{0.125}V_{0.125}$-LDHs/NF	**=125**	**≈220**	**1.42**	
Co_1Mn_1CH	**-**	**=328**	***5.6***	***J. Am. Chem. Soc., 2017, 139, 8320-8328***

Table S.7 Comparison of overall water splitting performance of Ni(II, III)Zn-LDH/NF-nm to recently reported hydroxide-based electrocatalysts in alkaline solution.

catalyst	Potential (mV, at 10 mA cm^{-2})	Potential (mV, at 50 mA cm^{-2})	Stability test (time, h)	Reference
Ni(II, III)Zn-LDH/NF-nm	**=1.68**	**≈1.78**	**16 h**	***This work***
$Ni_{0.75}Fe_{0.125}V_{0.125}$-LDHs/NF	**=1.591**	**≈1.74**	**15 h**	***Small, 2018, 14, 4724***
Fe_x Ni_yOOH/EGSI	**=1.69**	**≈1.91**	**-**	***ChemistrySelect, 2019, 4, 2153-2159***
NiFe LDH/NF	**≈1.66**	**≈1.86**	**-**	***Adv. Funct. Mater., 2018, 28, 1706847***
σ-FeOOH NSs/NF	**=1.62**	**≈1.83**	**60 h**	***Adv. Mater. 2018, 1803144***
Co_1Mn_1CH	**=1.68**	**≈1.87**	**13 h**	***J. Am. Chem. Soc., 2017, 139, 8320-8328***

References

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