# **Supplementary Information**

# Construction of Desert Rose Flower-Shaped NiFe LDH-Ni<sub>3</sub>S<sub>2</sub> Heterostructure via Seawater Corrosion Engineering for Efficient Water-Urea Splitting and Seawater Utilization

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#### Materials

Iron (III) nitrate nonahydrate (Fe(NO<sub>3</sub>)<sub>3</sub>·9H<sub>2</sub>O), iron(III) chloride hexahydrate (FeCl<sub>3</sub>·6H<sub>2</sub>O), iron(II) chloride tetrahydrate (FeCl<sub>2</sub>·4H<sub>2</sub>O), ammonium iron(II) sulfate hexahydrate ((NH<sub>4</sub>)<sub>2</sub>FeSO<sub>4</sub>·6H<sub>2</sub>O), urea, sodium chloride (NaCl, 99.5%), hydrochloric acid (HCl, 38%), potassium hydroxide (KOH, 85%), chloroplatinic acid hydrate (H<sub>2</sub>PtCl<sub>6</sub>×H<sub>2</sub>O, 99.9%), sulfuric acid (H<sub>2</sub>SO<sub>4</sub>), hydrogen peroxide (H<sub>2</sub>O<sub>2</sub>), poly(vinylidene fluoride) ((CH<sub>2</sub>CF<sub>2</sub>)<sub>n</sub>, M<sub>w</sub> ~534,000)), carbon black (99%), and 1-methyl-2-pyrrolidinone (C<sub>5</sub>H<sub>9</sub>NO, 99.5%) were purchased from Sigma-Aldrich (France) and used without further purification.

Acetone and ethanol were procured from Fisher Scientific SAS (Illkirch, France). Nickel foam (NF) ( $200 \times 300 \text{ mm}^2$ , thickness=1 mm) was obtained from Kunshan Lvchuang Electronic Technology Co., Ltd, China. Solar cell ( $50 \times 50 \text{ mm}^2$ , 2V) was purchased from Xu-Neng Trading Co., Ltd, China. The water used throughout the experiments was purified with an ultrapure water purification system Arium® comfort I from Sartorius (resistivity = 18.2 M\Omega.cm).

The H-type cell membrane (Dupont Proton Exchange Membrane N117) has the following characteristics: Thickness (183  $\mu$ m), density (360 g/m<sup>2</sup>), conductivity (0.083 S/cm), exchange capacity (0.89 meq/g). Pre-treatment method: the membrane was soaked in 5 wt.% of hydrogen peroxide (H<sub>2</sub>O<sub>2</sub>) for 1 h at 80°C, then placed in deionized water for 0.5 h, and finally treated with 5 wt.% of sulfuric acid for 0.5 h at 80°C.

### Characterization

The morphology and composition of the samples were assessed by scanning electron microscopy (SEM) using ULTRA 55 (Zeiss) equipped with a thermal field emission emitter and an energy dispersive X-ray spectroscopy (EDS) mapping detector. Transmission electron microscopy (TEM) imaging was performed by a TECNAI-G2 equipped with EDS spectrometer at a working voltage of 200 kV. X-ray diffraction (XRD) patterns were collected on a high flux Rigaku Smartlab rotated anode, working with a copper K $\alpha$  radiation (1.5418 ang) at an applied voltage of 45 kV and an anode current of 200 mA in the  $2\theta$  range of 5° to 80° at a scan rate of 5° min<sup>-1</sup>. X-ray photoelectron spectroscopy (XPS) measurements were acquired on a PHI 5000

Versa Probe-Scanning ESCA Microprobe (ULVAC-PHI, Japan/USA) instrument featuring a monochromatic Al-Ka (1486.6 eV) radiation. The water contact angle measurements were performed with Ossila L2004A1 analyzer using a 10  $\mu$ L water droplet at room temperature.

#### **Electrochemical measurements**

Electrochemical measurements were conducted using an electrochemical workstation (Autolab PGSTAT204, Metrohm) at ambient temperature in a standard three-electrode system using 1M KOH aqueous solution as the electrolyte, NiFe LDH-Ni<sub>2</sub>S<sub>3</sub> as the working electrode, carbon rod as the counter electrode and Ag/AgCl (3.5 M KCl) as the reference electrode. It should be noted that the current density is standardized to the geometrical surface area and all the potential measurements are calibrated to a reversible hydrogen electrode (RHE), based on the following equation S1:

$$E_{RHE} = E_{Ag/AgCl} + 0.0591 \times pH + 0.197$$
(S1)
$$E_{RHE} = E_{Ag/AgCl} + 1.014; 1M \text{ KOH solution (pH=13.8)}$$

For this system, we used 90% iR-correction to calculate LSV polarization curves. As shown in equation S2, the common 90% iR-correction could effectively eliminate the errors caused by solution resistance and open circuit potential;  $R_s$  (resistance of solution);  $V_{OCP}$  (open-circuit potential).

$$V = V_{\text{measured}} - I_{\text{measured}} * R_{\text{s}} * 90\% - V_{\text{OCP}}$$
(S2)

For the HER performance, LSV curves were recorded separately in 1M KOH, 1M KOH+0.5M NaCl and 1M KOH seawater at a scan rate of 5 mV s<sup>-1</sup> over the potential range from -1 to -1.7 V (*vs.* RHE). The HER operation stability of NiFe LDH-Ni<sub>2</sub>S<sub>3</sub> sample was conducted by chronopotentiometry at a current density of -10 mA cm<sup>-2</sup> for 35 h separately in 1M KOH, 1M KOH+0.5M NaCl and 1M KOH seawater solutions. The multistep chronoamperometric measurements were recorded at various overpotentials. In addition, 1000 CV cycles were acquired at a scan rate of 10 mV s<sup>-1</sup> in the potential range from -1 to -1.7 V (*vs.* RHE).

For the OER performance, cyclic voltammetry (CV) curves were conducted separately in 1M

KOH, 1M KOH+0.5M NaCl and 1M KOH seawater solutions in the potential range of  $0.80 \sim 1.80 \text{ V}$  (*vs.* RHE) at a scan rate of 5 mV s<sup>-1</sup>. The overpotential ( $\eta$ ) of OER process was calculated using  $\eta(V) = E_{(RHE)} - 1.23(V)$ . The OER operation stability of NiFe LDH-Ni<sub>2</sub>S<sub>3</sub> sample was conducted by chronopotentiometry at a current density of -10 mA cm<sup>-2</sup> for 35 h separately in 1M KOH, 1M KOH+0.5M NaCl and 1M KOH seawater solutions. The multistep chronoamperometric measurements were acquired at various overpotentials. In addition, 1000 CV cycles were recorded at a scan rate of 10 mV s<sup>-1</sup> in the potential range from 0.8 to 1.8 V (*vs.* RHE).

Electrochemical impedance spectroscopy (EIS) measurements were carried out separately in the potential range of  $1.014 \sim 1.514$  V (vs. RHE) for the OER or UOR and  $+0.114 \sim -0.386$  V (vs. RHE) for the HER in the frequency range of 100 kHz to 0.01 Hz at an amplitude of 10 mV. R<sub>s</sub> could be obtained from the resistance recorded at the frequency of 100 kHz.

The ECSA was determined by measuring the capacitive current associated with double-layer charging from the scan-rate dependence of the CV plots. The potential window of CV was 0.1-0.2 V *versus* RHE for HER process, while a potential window of 0.85-0.95 V (*vs.* RHE) was applied for the OER/UOR process at various scan rates (20, 40, 60, 80, 100 and 120 mV s<sup>-1</sup>). The double-layer capacitance (C<sub>dl</sub>) was calculated by plotting the  $\Delta J = (Ja - Jc)$  at the middle of potential against the scan rate, while the linear slope is twice of the double-layer capacitance C<sub>dl</sub>.

The two-electrodes system for full water splitting and overall urea electrolysis was assembled by using NiFe LDH-Ni<sub>2</sub>S<sub>3</sub> foam as both anode and cathode and powered by PGSTAT204 workstation or a 2.0 V solar cell in 1M KOH without or with 0.33M urea seawater solution. The chronopotentiometry curves were acquired at a current density of 10 mA cm<sup>-2</sup> for 65 h to evaluate the stability of the urea splitting cell.

#### Faradaic efficiency measurements for the HER, OER and UOR

The faradaic efficiency (*FE*) measurements were also conducted on the best performing catalyst, namely NiFe LDH-Ni<sub>2</sub>S<sub>3</sub>, to further evaluate its electrocatalytic performance towards

HER, OER and UOR and confirm electrochemical findings. Firstly, oxygen and hydrogen were collected using water displacement method during the overall water or urea splitting processes, and then the produced ideal gas volume with 100% theoretical Faradaic efficiency was calculated. To achieve this, the Faradic efficiency was obtained based on the following equation S3. We evaluated the electrochemical behavior by comparing the volume of gas produced by 100% Faradaic theoretical current with the amount of gas collected experimentally in two-electrode overall water splitting and urea splitting conditions.

Faradaic efficiency = 
$$[zF \times (n_{H2} \text{ or } n_{O2})] / Q$$
 (S3)

where z represents the number of electrons involved in the reaction (2 for H<sub>2</sub> and 4 for O<sub>2</sub>), n<sub>H2</sub> or n<sub>O2</sub> stands respectively for the number of H<sub>2</sub> or O<sub>2</sub> moles estimated approximately from the ideal gas law, F is the Faraday constant (96485 C mol<sup>-1</sup>), and Q is the charge consumed during the electrolysis process (Q = I × t, I is a given value of a constant applied current in the test and t is the reaction time (s) under the constant applied current). In this system, constant-current electrolysis was carried out at a current density of 50 mA cm<sup>-2</sup> under standard conditions for 1000 s.

## **Corrosion behavior measurements**



Figure S1. Corrosion polarization curves of nickel foam (NF) in (a) 0.5M NaCl, (b) 30 mM
Fe(NO<sub>3</sub>)<sub>3</sub>·9H<sub>2</sub>O, (c) 0.5M NaCl+30 mM Fe(NO<sub>3</sub>)<sub>3</sub>·9H<sub>2</sub>O aqueous solutions, (d) comparison of corrosion potentials (E<sub>corr</sub>), (e) comparison of corrosion current densities (I<sub>corr</sub>), (f) comparison of different solution pH values.



Figure S2. (a) SEM images at different magnifications, (b) elemental mapping images of Ni, C and O, and (c) EDX spectrum of the pristine Ni foam.



**Figure S3.** (a) SEM images at different magnifications, (b) the corresponding elemental mapping images of Ni, O, Na and Cl, (c) EDX spectrum and elemental composition (inset) of NaCl-Ni foam.



Figure S4. SEM and elemental mapping images of (a) NiFe LDH-1h, (b) NiFe LDH-6h, (c) NiFe LDH-12h and (d) NiFe LDH-18h. The corresponding EDX spectra and elemental composition of Ni, Fe, O, Na and Cl.



Figure S5. (a) SEM image, (b) the corresponding EDS elemental mapping images of Ni, Fe, S, and O, and (c) EDX spectrum and table of element distribution of NiFe LDH-Ni<sub>3</sub>S<sub>2</sub>.



Figure S6. XRD patterns of (a) Ni foam before (black) and after (red) immersion in 0.5M
NaCl aqueous solution for 12 h at room temperature. (b) XRD patterns of NiFe LDH prepared using different corrosion times.



Figure S7. LSV polarization curves of NiFe LDH prepared using various types of Fe salts for catalyzing (a) HER and (b) OER in 1M KOH at a scan rate of 5 mV s<sup>-1</sup>.



**Figure S8.** (a) Photographs of NiFe LDH catalysts prepared using various Fe(NO<sub>3</sub>)<sub>3</sub> concentrations and 0.5M NaCl solution, (b) LSV polarization curves of NiFe LDH prepared using various Fe(NO<sub>3</sub>)<sub>3</sub> concentrations for catalyzing HER in 1M KOH at a scan rate of 5 mV s<sup>-1</sup>, and (c) comparison of their overpotential values at the current densities of 10 and 100 mA cm<sup>-2</sup>.



**Figure S9.** (a) CV polarization curves of NiFe LDH prepared using various  $Fe(NO_3)_3$  concentrations in 0.5M NaCl solution for catalyzing OER in 1M KOH at a scan rate of 5 mV s<sup>-1</sup>. (b) Comparison of their overpotential values at the current densities of 50 and 100 mA cm<sup>-1</sup>





Figure S10. LSV polarization curves of NiFe LDH prepared using various corrosion times in 0.5M NaCl + 30 mM Fe(NO<sub>3</sub>)<sub>3</sub> for catalyzing (a) HER and (b) OER in 1M KOH at a scan rate of 5 mVs<sup>-1</sup>.



Figure S11. CV polarization process of NiFe LDH (0.5M NaCl) for catalyzing (a) HER and
(b) OER in 1M KOH at a scan rate of 5 mV s<sup>-1</sup>, demonstrating the catalyst surface reconstruction from NiFe LDH to NiFe-O(OH) *via* electrochemical activation.



Figure S12. LSV polarization curves of NiFe LDH-Ni<sub>3</sub>S<sub>2</sub> prepared through immersion in 0.5M Na<sub>2</sub>S by adjusting the reaction time for catalyzing the (**a**) HER, and (**b**) OER in 1M KOH at a scan rate of 5 mV s<sup>-1</sup>.



Figure S13. Nyquist and Bode plots at various potentials for HER process of (a, b) Ni foam,(c) NiFe LDH, and (c) NiFe LDH-Ni<sub>3</sub>S<sub>2</sub> foam.



Figure S14. CV curves in the potential range from 0.10 to 0.20 V acquired at various scan rates in 1M KOH solution of (a) Ni foam, (b) NiFe LDH and (c) NiFe LDH-Ni<sub>3</sub>S<sub>2</sub>. (d) Electrochemical double-layer capacitance as a function of scan rate.



Figure S15. Long-term stability measurement of the NiFe LDH-Ni<sub>3</sub>S<sub>2</sub> using multistep chronoamperometric curves for HER process.



Figure S16. (a) SEM images at different magnifications of the NiFe-Ni<sub>3</sub>S<sub>2</sub> after 1000 HER
 CV cycles, (b) the corresponding elemental mapping images, (c) EDX spectrum and element content analysis.



**Figure S17.** CV curves in the potential range from 0.85 to 0.95 V recorded at various scan rates in 1M KOH solution of (**a**) Ni foam, (**b**) NiFe LDH and (**c**) NiFe LDH-Ni<sub>3</sub>S<sub>2</sub>, and in 1M KOH+0.33 M urea solution of (**d**) Ni foam, (**e**) NiFe LDH and (**f**) NiFe LDH-Ni<sub>3</sub>S<sub>2</sub>.



Figure S18. (a) SEM images at different magnifications of the NiFe-Ni<sub>3</sub>S<sub>2</sub> after 1000 OER
CV cycles, (b) the corresponding elemental mapping images, (c) EDX spectrum and element content analysis.



Figure S19. XRD spectra of NiFe LDH-Ni $_3S_2$  after 1000 CV cycles of HER, OER and UOR measurements.



**Figure S20.** (a) Ni 2p, (b) Fe 2p, (c) O 1s and (d) S 2p XPS spectra of NiFe LDH-Ni<sub>3</sub>S<sub>2</sub> after 1000 CV cycles of HER, OER and UOR.



Figure S21. Nyquist (a) and Bode (b) plots for various potentials for the OER process of Ni foam.



Figure S22. Contact angle (CA) measurements of (a) Ni foam, (b) NiFe-LDH and (c) NiFe

LDH-Ni<sub>3</sub>S<sub>2</sub> using a 10  $\mu$ L water droplet.



**Figure S23.** (a) SEM images at different magnifications of the NiFe-Ni<sub>3</sub>S<sub>2</sub> after 1000 UOR CV cycles, (b) the corresponding elemental mapping images, (c) EDX spectrum and element



Figure S24. (a) Device diagram for measuring Faraday efficiency. (b) Diagram of the amount of H<sub>2</sub> and O<sub>2</sub> released over time in 1M KOH. (c) Enlarged diagram of the gas collected equipment *via* water displacement method during overall water splitting process.

Solution	рН-1	рН-2	рН-3	Average
1M KOH	13.8	13.8	13.8	13.8
1M KOH+0.5M NaCl	13.7	13.7	13.7	13.7
1M KOH seawater	13.8	13.8	13.8	13.8

**Table S1.** pH values of (a) 1M KOH, (b) 1M KOH+0.5M NaCl and (c) 1M KOH seawaterelectrolyte solutions.

**Table S2.** pH values of 30 mM (**a**) (NH<sub>4</sub>)<sub>2</sub>FeSO<sub>4</sub>·6H<sub>2</sub>O, (**b**) FeCl<sub>2</sub>·4H<sub>2</sub>O, (**c**) FeCl<sub>3</sub>·6H<sub>2</sub>O and (**d**) Fe(NO<sub>3</sub>)<sub>3</sub>·9H<sub>2</sub>O in 0.5M NaCl aqueous solutions.

Sample	Valence	pH-1	pH-2	рН-3	Average
$(NH_4)_2FeSO_4 \cdot 6H_2O$	Fe <sup>2+</sup>	3.68	3.69	3.69	3.69
FeCl <sub>2</sub> ·4H <sub>2</sub> O	Fe <sup>2+</sup>	2.38	2.39	2.39	2.39
FeCl <sub>3</sub> ·6H <sub>2</sub> O	Fe <sup>3+</sup>	2.08	2.10	2.11	2.10
$Fe(NO_3)_3 \cdot 9H_2O$	Fe <sup>3+</sup>	2.08	2.10	2.10	2.09

 Sample
 Ni (at. %)
 Fe (at. %)
 S (at. %)
 O (at. %)

 NiFe LDH-Ni<sub>3</sub>S<sub>2</sub> foam <sup>a</sup>
 11.35
 4.93
 6.24
 40.87

 NiFe LDH-Ni<sub>3</sub>S<sub>2</sub> foam <sup>b</sup>
 15.5
 12.8
 13.4
 38.3

**Table S3.** Surface composition of NiFe LDH-Ni2S3 catalyst determined by a XPS and b TEM-EDX analysis.

 Table S4. XPS analysis of Ni, Fe, S, O and C elements before and after HER, OER and UOR stability tests.

No	Sample	Ni (at. %)	Fe (at. %)	S (at. %)	O (at. %)	C (at. %)
1	NiFe foam	13.09	6.11		45.51	30.68
2	NiFe LDH-Ni <sub>3</sub> S <sub>2</sub>	11.35	4.93	6.24	40.87	35.42
3	NiFe LDH-Ni $_3$ S $_2$ after HER	10.96	2.4	1.81	52.62	31.31
4	NiFe LDH-Ni $_3$ S $_2$ after OER	14.81	5.16	0.93	44.57	34.52
6	NiFe LDH-Ni <sub>3</sub> S <sub>2</sub> after UOR	13.18	2.35	0.76	55.4	27.51

Catalutia	Preparation	Current	Overpotential	Tafel slope	Substrata	Dof
Catalytic	method	density	(mV)	(mV dec <sup>-1</sup> )	Substrate	Kel.
		(mA cm <sup>-2</sup> )				
NiFe LDH-Ni <sub>3</sub> S <sub>2</sub>	IC	10 100	101 240	95.3	Ni foam	This work
Ni <sub>5</sub> Co <sub>3</sub> Mo–OH	IC	10 100	52 249	59	Ni foam	1
Pt-Ni Fe LDH	IC	10	31	36	Ni foam	2
Fe-Ni <sub>3</sub> S <sub>2</sub>	HT	100	232	95	Ni foam	3
NiCo LDH	HT	10	271	141	Ni foam	4
NiCoP/NF	HT	10	122	88	Ni foam	5
NiFe LDH/NF	HT	10	210	62	Ni foam	6
Ni <sub>5</sub> Fe LDH	HT	10	130	89	Ni foam	7
(Ni, Fe)S <sub>2</sub> @MoS <sub>2</sub>	HT	10	130	101.2	СР	8
$Fe_{0.8}Ni_{0.15}S_{1.05}$	HT	10	263	103	RDE	9
Fe11.1%-Ni <sub>3</sub> S <sub>2</sub> /Ni	HT	10	126	89	Ni foam	10
Ni <sub>3</sub> Se <sub>4</sub> @NiFe LDH	HT	10 100	85 220	98.6	СР	11

**Table S5.** Comparison of the HER activity of NiFe LDH-Ni<sub>3</sub>S2 with other published metal-based HER electrocatalysts in 1M KOH.

RDE: rotating disk electrode

CP: carbon paper

I: ion corrosion method

HT: hydrothermal method

Catalytic	Preparation	Current	Overpotential	Tafel slope	Substrate	Ref.
	method	density	(mV)	(mV dec <sup>-1</sup> )		
		(mA cm-2)				
NiFe LDH-Ni <sub>3</sub> S <sub>2</sub>	IC	10	198	43.5	Ni foam	This
		100	240			work
NiFe LDH-Pt	IC	10	261	39.7	Ni foam	2
		100	320			
Ni <sub>5</sub> Co <sub>3</sub> Mo–OH	IC	100	304	56.4	Ni foam	1
	IJТ	10	210	50	NI: Comm	7
Ni <sub>5</sub> Fe LDH	ΠI	10	210	39	INI IOam	,
	ЦΤ	10	203	55 5	CD	11
Ni <sub>3</sub> Se <sub>4</sub> @NiFe LDH	пі	10	223	55.5	CP	
	UТ	100	230	53	PDE	9
$Fe_{0.8}Ni_{0.15}S_{1.05}$	111	100	540	55	KDE	
	НТ	100	326	157	Ni foam	12
pa-Nife LDH NS	111	100	520	107	TH Iouin	
Ea11 1% N; S	HT	100	252	61	Ni foam	10
1611.170–113.52						
Hapoa /FeNi-I DH-VaC	HT	100	300	46	RDG	13
(Ni,Fe,Coc-x-v) MocC	HT	10	212	55.1	Ni foam	14
		100	260			
MoNiFe-27% (oxy)	HT	10	242	23	Ni foam	15
		100	290			
NiFe-LDH@FeNi <sub>2</sub> S <sub>4</sub>	HT	100	240	29.4	Ni foam	16
0						
NIFE LDH/NIS	HT	100	277	60.1	Ni foam	17
Fe17.5%-Ni <sub>3</sub> S <sub>2</sub> /NF	HT	100	249	42	Ni foam	3

**Table S6.** Comparison of the OER activity of NiFe LDH-Ni<sub>3</sub>S2 with other published metal-based OER electrocatalysts in 1 M KOH.

RDE: rotating disk electrode CP: carbon paper

IC: ion corrosion method HT: hydrothermal method

Catalytic	Preparation	Current density	Potential	Substrate	Reference
	method	(mA cm <sup>-2</sup> )	(V)		
NiFe LDH-Ni <sub>2</sub> S <sub>3</sub> // NiFe LDH-Ni <sub>2</sub> S <sub>3</sub>	IC	50 100	1.58 1.63	Ni foam	This work
FQD/CoNi-LDH@ FQD/CoNi-LDH	HT	50 100	1.6 1.70	Ni foam	18
NiMoO <sub>4</sub> -300/NF@ NiMoO <sub>4</sub> -300/NF	HT	50	1.68	Ni foam	19
Fe-doped NiS–NiS <sub>2</sub> @ Fe-doped NiS–NiS <sub>2</sub>	НТ	50	1.67	Ni foam	20
NiCoP NS/CC@ NiCoP NS/CC	HT	50	1.50	Carbon cloth	21
$\begin{array}{c} CoS_2\text{-}MoS_2@\\ CoS_2\text{-}MoS_2 \end{array}$	HT	50	1.50	Ni foam	22
HC-NiMoS/Ti@ HC-NiMoS/Ti	HT	50	1.78	Ti mesh	23
NF/NiMoO-Ar@ NF/NiMoO-H <sub>2</sub>	HT	50 100	1.48 1.55	Ni foam	24

 Table S7. Comparison of the two-electrode system potential of the developed catalyst and

 reported UOR catalysts.

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