Supporting Information

Multiphases interface coupling of Ni-based sulfides composites for large-current-density oxygen evolution electrocatalysis in alkaline freshwater/simulated seawater/seawater

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Experimental section

1. Materials and Characterizations

Pattern X-ray diffraction (XRD) was recorded using a Bruke D8 equipped with Cu-Ka radiation. X-ray photon-electron spectroscopy (XPS) analysis was performed using a VG ESCALAB MK II spectrophotometer with Mg-Kα radiation (1253.6 eV). Scanning electron microscopy (SEM) test was carried out on a Hitachi S-4800 instrument at an accelerating voltage of 5kV. The high-resolution transmission electron microscopy (HRTEM) and selected area electron diffraction (SAED) patterns were performed on a FEI Talos-F200S electron microscope. The compositions of the products were examined using energy dispersive X-ray spectroscopy (EDX) attached to the EM system. Specific surface areas and pore size were measured using Brunauer-Emmett-Teller (BET). Atomic Force Microscope (AFM, BioScope Resolve, Bruker, USA) was also utilized to characterize the morphology of NNSN/NF sample using ScanAsyst-Air probe (Bruker). The optical absorption studies of the synthesized sample were analysed with the absorption spectra using UV–Vis-NIR spectrophotometer (Shimadzu). The contact angles were measured using Contact Angle/surface tension measuring instrument (Dataphysics OCA20).

2. Preparation of NNSN/NF-xh (x=8, 11 and 14)

Sonicate NF (1×3 cm^2) in 3M HCl solution for 30 min to remove the NiO layer on the surface of the NF. It is then sonicated three times with deionized water and ethanol to remove residual HCl and organic impurities on the surface of NF, and then dried in an oven at 60°C.

6.63 mmol CH$_4$N$_2$S and 0.26 mmol Ni(NO$_3$)$_2$·6H$_2$O were accurately weighed and dissolved in 25 mL of CH$_3$CH$_2$OH and then magnetically stirred for 20 min to form a clarified and homogeneous solution, and then NF was put into the solution, and the solution was transferred into a 50 mL PTFE-lined stainless steel reactor with a piece of NF in it. The reaction was kept at 160 °C for 8 h, 11 h and 14 h, respectively. The as-obtained materials were washed with DI water and ethanol repeatedly and then dried at 60 °C for 12 h to obtain the NNSN/NF-xh (x=8, 11 and 14) samples.

3. Electrochemical measurements

Electrochemical tests were performed on the CHI760e workstation, using a typical three-electrode system, Ag/AgCl as the reference electrode, graphite rod as the counter electrode, and the NNSN/NF as the working electrode. All potentials are expressed with reference to a reversible hydrogen electrode (RHE) by the Nernst equation: $E_{RHE} = E_{Ag/AgCl} + 0.059 pH + 0.197$. The electrolytes are 1M KOH, 1M KOH+0.5M NaCl and 1M KOH + seawater, and its pH is 14. All linear sweep

voltammetry (LSV) curves are 95% iR corrected and the sweep rate is 5 mV s^{-1}. The Tafel data were fitted according to the equation η = b log(j) + α (j: current density, and b: Tafel slope). The durability tests were carried out by repeating the potential at a scan rate of 50 mV s^{-1} for 3000 cycles. The i-t test was carried out at a constant potential to surveil the variation of the current density during the long-term OER process. CV measurements of the electrocatalysts were performed in a potential range of 0.94–1.04V (vs. RHE) at scan speeds of 10-50 mV s^{-1} to calculate the double-layer capacitance (C$_{dl}$) of the catalysts. ECSA was evaluated by measuring C$_{dl}$ of CV at non-Faradaic potential. EIS is measured in the frequency range of 0.1 Hz-100 kHz and the voltage is 0.5 V.

Section 2 Supplementary Structural Section
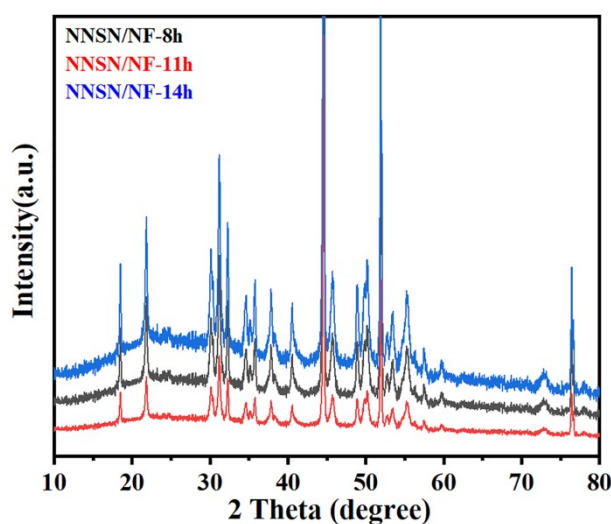
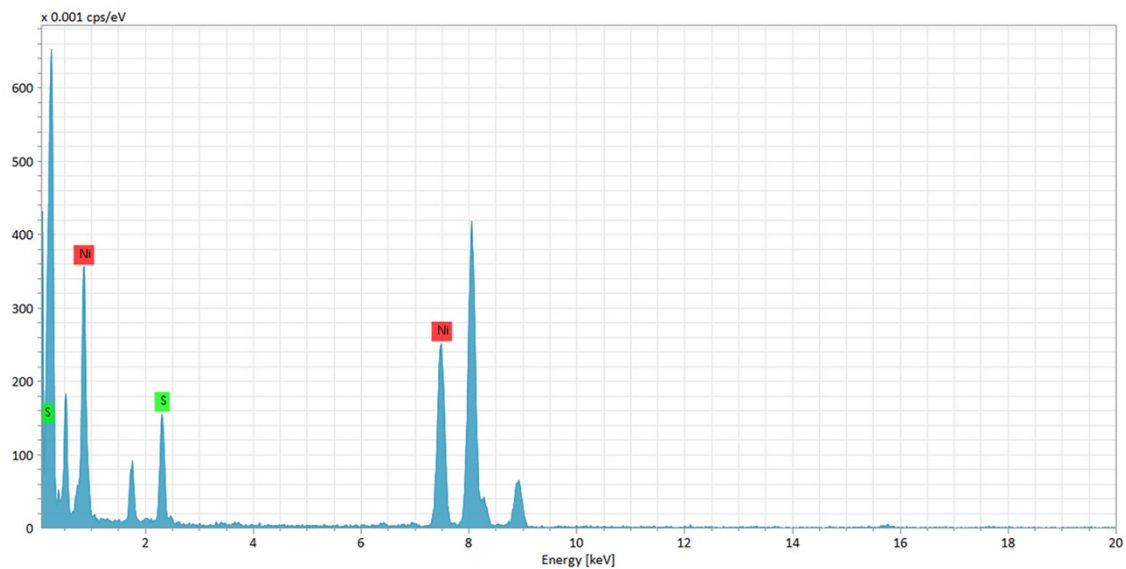**Fig. S1** XRD patterns of NNSN/NF-xh (x=8, 11 and 14)

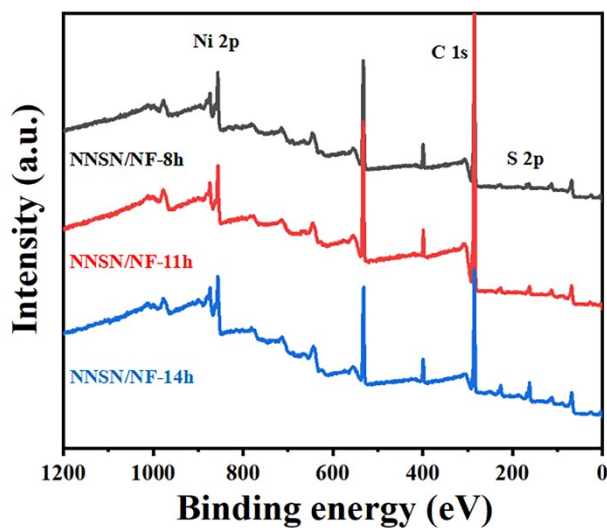**Fig. S2** EDX patterns of NNSN/NF-11h

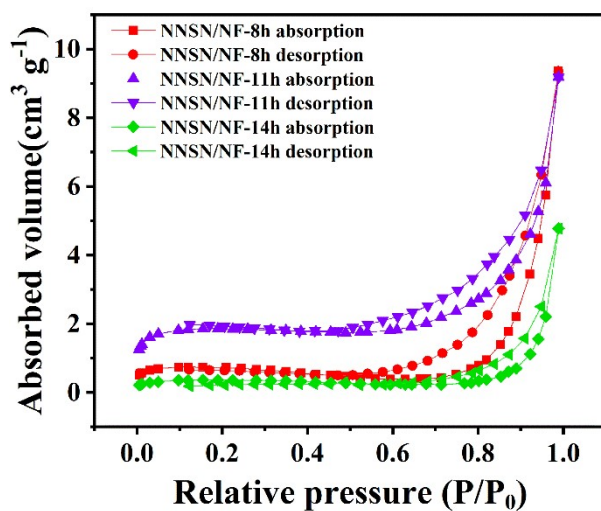**Fig. S3** XPS spectrum of NNSN/NF-8,11 and 14h

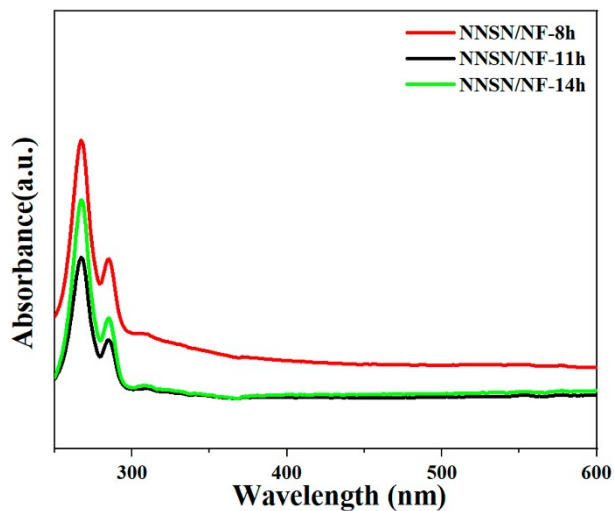**Fig. S4** N$_2$ adsorption–desorption isotherms of NNSN/NF-xh (x=8,11 and 14).

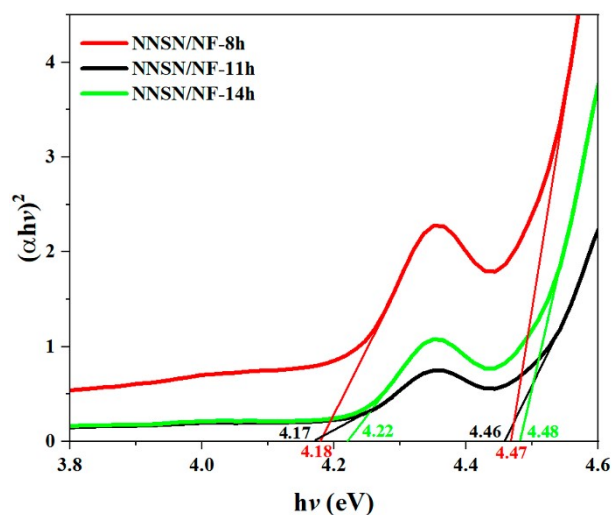**Fig. S5** The UV-visible absorption spectra of NNSN/NF-xh (8,11 and 14).

Fig.S6 The corresponding curves of $(\alpha h\nu)^2$ versus $h\nu$ of NNSN/NF-xh (x=8,11 and 14) samples.

Fig. S7 C_{dl} plots of NNSN/NF-8h

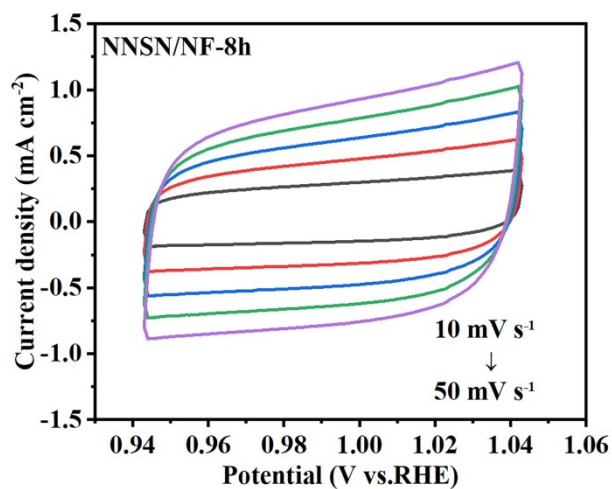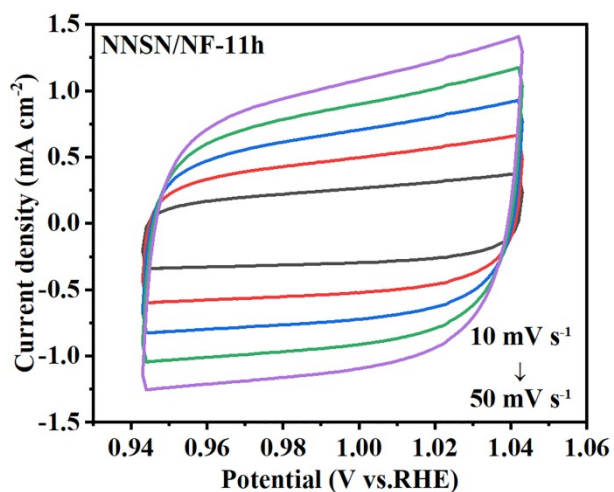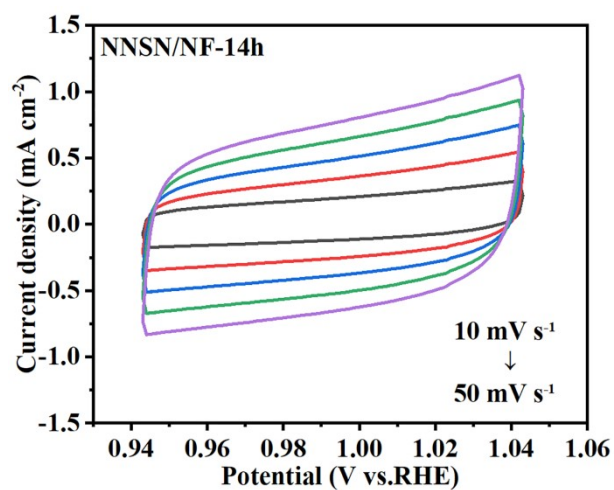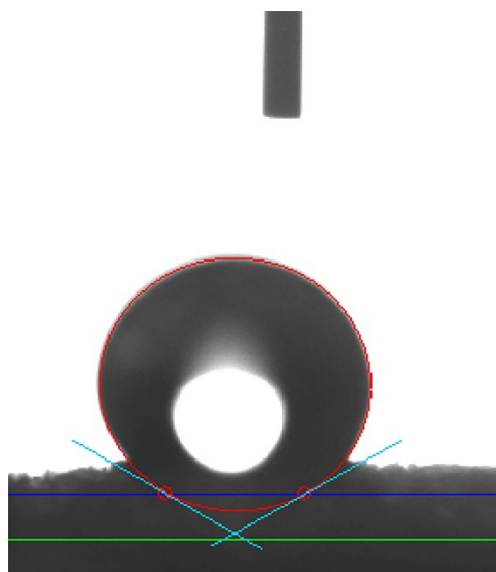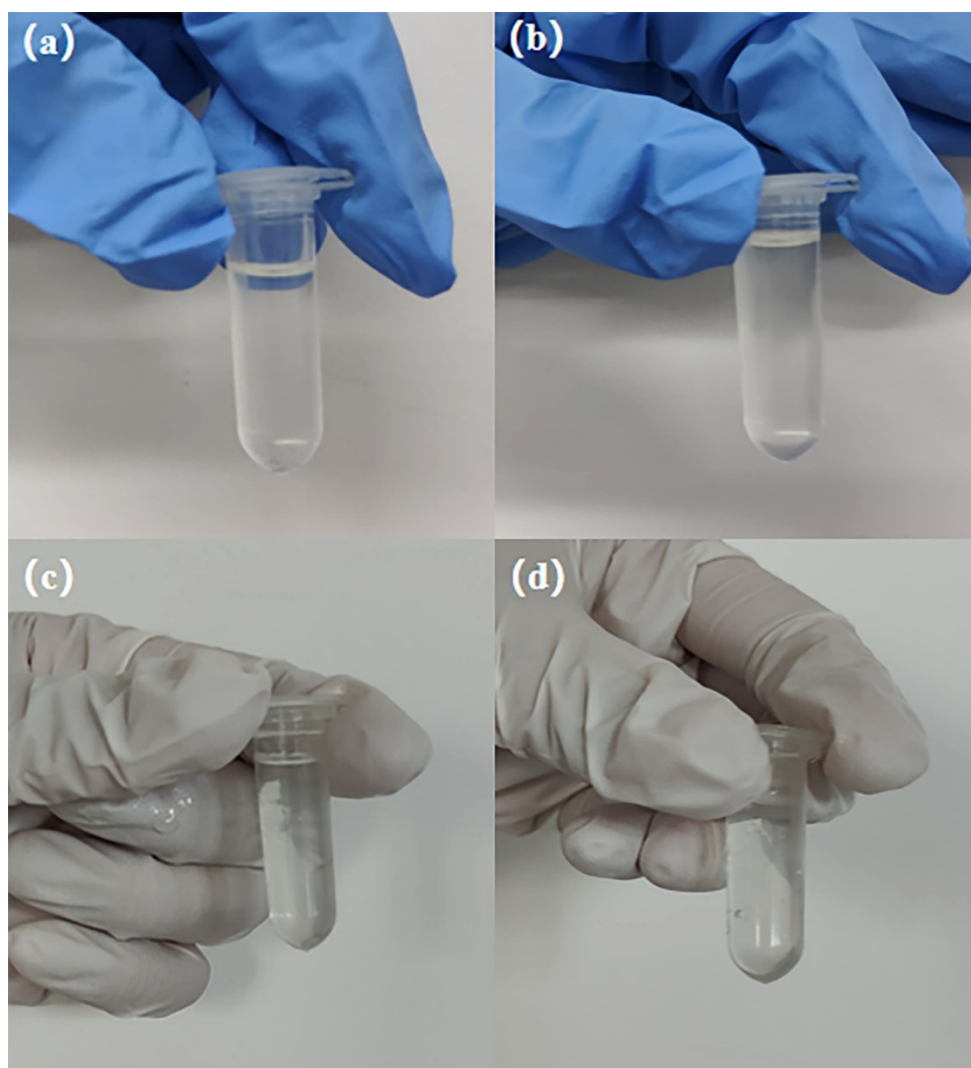**Fig. S8** C_{dl} plots of NNSN/NF-11h

Fig. S9 C_{dl} plots of NNSN/NF-14h

Fig. S10 Digital images of the wetting process of 1M KOH droplet at the NF.

Fig. S11 (a and c) the 1.0 M KOH + 0.5 M NaCl and 1.0 M KOH +seawater electrolyte after 3000 CV cycles. (b and d) the 1.0 M KOH + 0.5 M NaCl and 1.0 M KOH +seawater electrolyte after 3000 CV cycles with the addition of KI.

Table S1. Comparison of OER performance of similar catalysts in 1.0 M KOH solution.

Catalyst	Electrolyte	j [mA cm^{-2}]	η [mV]	Tafel [mV dec^{-1}]	Ref.
FeOOH/Co$_9$S$_8$/Ni$_3$S$_2$	1.0 M KOH	1000	265	32	S1
NiFeS/NF	1.0 M KOH	500	272	56.37	S2
NiCo@C–NiCoMoO/NF	1.0 M KOH	1000	390	75.15	S3
Fe-NiS/NF	1.0 M KOH	500	222	41.7	S4
S-(Ni,Fe)OOH/NF	1.0 M KOH	1000	355	48.9	S5
Ni@C–MoO$_2$/NF	1.0 M KOH	1000	400	52.34	S6
FeCoNiMnOOH/NF	1.0 M KOH	500	378	62.1	S7
V-Ni$_3$Se$_2$/NF	1.0 M KOH	500	370	62	S8
Ni-Fe-OH@Ni$_3$S$_2$/NF	1.0 M KOH	500	530	93	S9
Fe-NiMoSe@C	1.0 M KOH	1000	446	34.38	S10
NNSN/NF	1.0 M KOH	1000	377	45.93	This work

Table S2. Comparison of OER performance of similar catalysts in 1.0 M KOH+0.5 M NaCl solution.

Catalyst	Electrolyte	j [mA cm^{-2}]	η [mV]	Tafel [mV dec^{-1}]	Ref.
NiFeS/NF	1.0 M KOH + 0.5 M NaCl	500	288	---	S2
NiCoS/NF	1.0 M KOH + 0.5 M NaCl	1000	430	---	S11
NiCoHPi@Ni$_3$N/NF	1.0 M KOH + 0.5 M NaCl	500	425	---	S12
R-NiCo(OH)F	1.0 M KOH + 0.5 M NaCl	1000	550	---	S13
MOEE	30% KOH + 0.5 M NaCl	1000	470	---	S14
NiFe hydroxide/NiS$_x$-Ni	1.0 M KOH + 0.5 M NaCl	400	510	---	S15
Fe-Ni(OH)$_2$/Ni$_3$S$_2$@NF	1.0 M KOH + 0.5 M NaCl	10	269	46	S16
Fe$_x$-Ni$_3$S$_2$/NF	1.0 M KOH + 0.5 M NaCl	100	306	80	S17
Ni(OH)$_2$@NiFe-PBA	1.0 M KOH + 0.5 M NaCl	100	342	57.1	S18
NNSN/NF	1.0 M KOH + 0.5 M NaCl	1000	377	45.93	This work

Table S3. Comparison of OER performance of similar catalysts in 1.0 M KOH+ seawater.

Catalyst	Electrolyte	j [mA cm^{-2}]	η [mV]	Tafel [mV dec^{-1}]	Ref.
S-(Ni,Fe)OOH/NF	1.0 M KOH + seawater	1000	462	---	S5
NiCoS/NF	1.0 M KOH + seawater	1000	470	---	S11
NiFe LDH-CeW@NFF	1.0 M KOH + seawater	500	330	81.2	S19
Ru-Ni(Fe)P$_2$/NF	1.0 M KOH + seawater	1000	520	---	S20
BZ-NiFe-LDH/CC	1.0 M KOH + seawater	500	610	---	S21
NiMoN/NiFeN	1.0 M KOH + seawater	100	307	---	S22
S-NiFeSe$_2$	1.0 M KOH + seawater	100	367	32	S23
Ni(OH)$_2$-TCNQ/GP	1.0 M KOH + seawater	100	382	75	S24
NNSN/NF	1.0 M KOH + seawater	1000	476	54.66	This work

Table S4. Comparison of OER performance of similar catalysts in 1.0 M KOH solution.

Catalyst	Electrolyte	j [mA cm^{-2}]	η [mV]	Tafel [mV dec^{-1}]	Ref.
RuO$_2$	1.0 M KOH	500	495	98.01	S2
RuO$_2$	1.0 M KOH	100	377	67.4	S25
RuO$_2$	1.0 M KOH + seawater	10	620	---	S26
IrO$_2$	1.0 M KOH + seawater	500	763	132	S27
NNSN/NF-11h	1.0 M KOH	1000	377	45.93	This work
NNSN/NF-11h	1.0 M KOH + seawater	1000	476	54.66	This work

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