

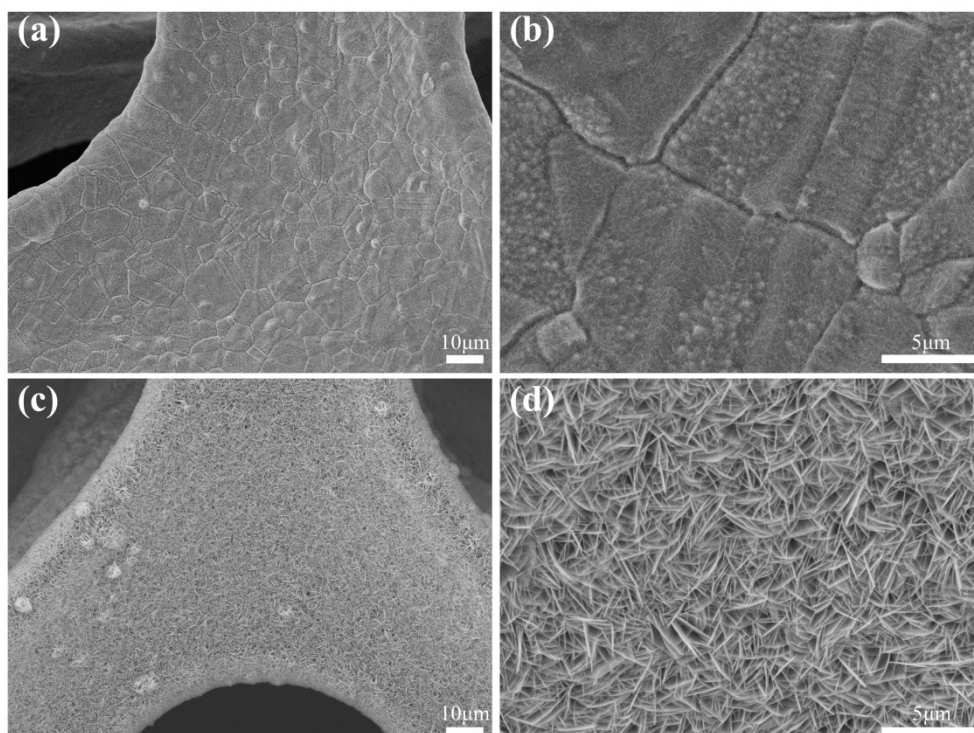
## Supporting Information

### **High-efficiency NiFeSe<sub>4</sub>/NiSe<sub>2</sub> bifunctional electrocatalyst with outstanding oxygen evolution reaction and overall water splitting performance**

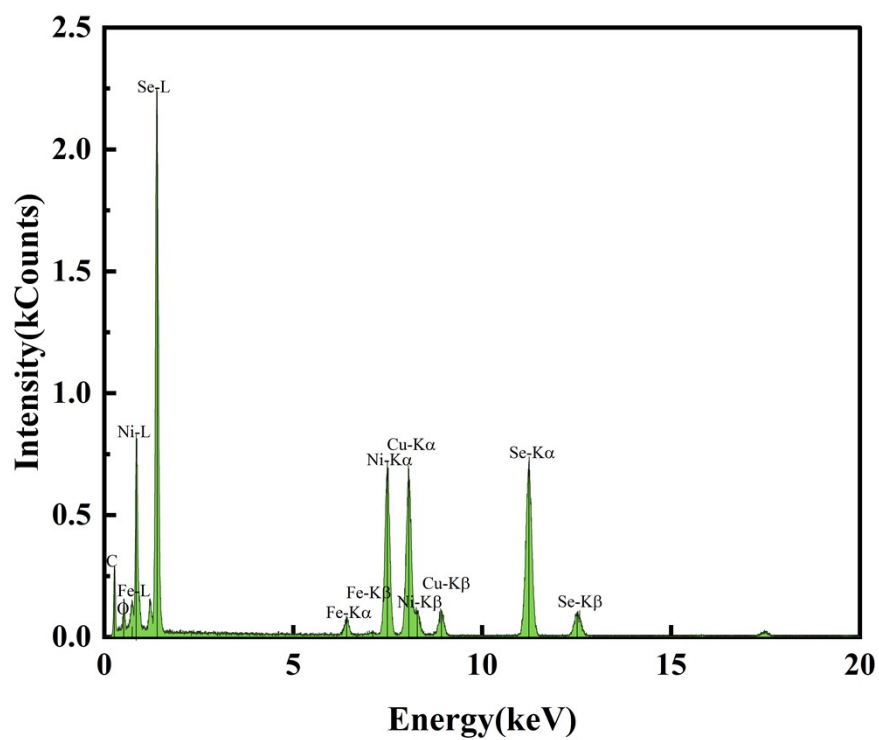
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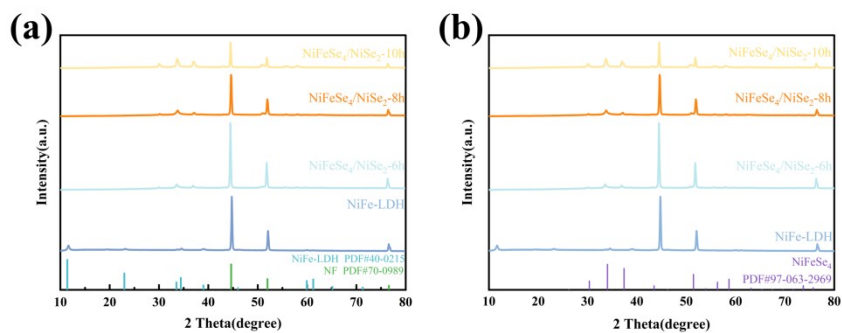
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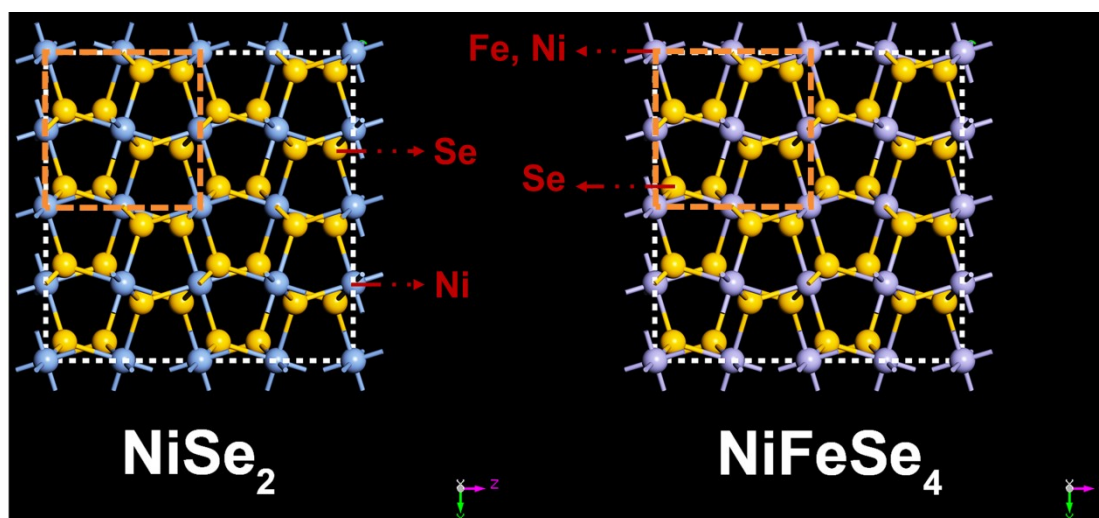
**Figure S1** SEM image of catalyst (a-b) nickel foam; (c-d) NiFe-LDH.



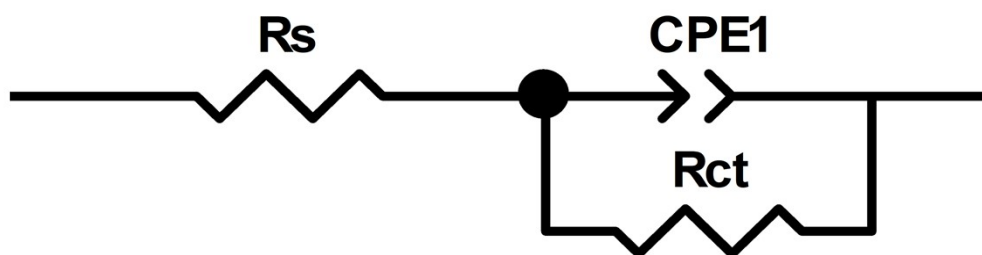
**Figure S2** Energy dispersion spectrum of NiFeSe<sub>4</sub>/NiSe<sub>2</sub>-8h.



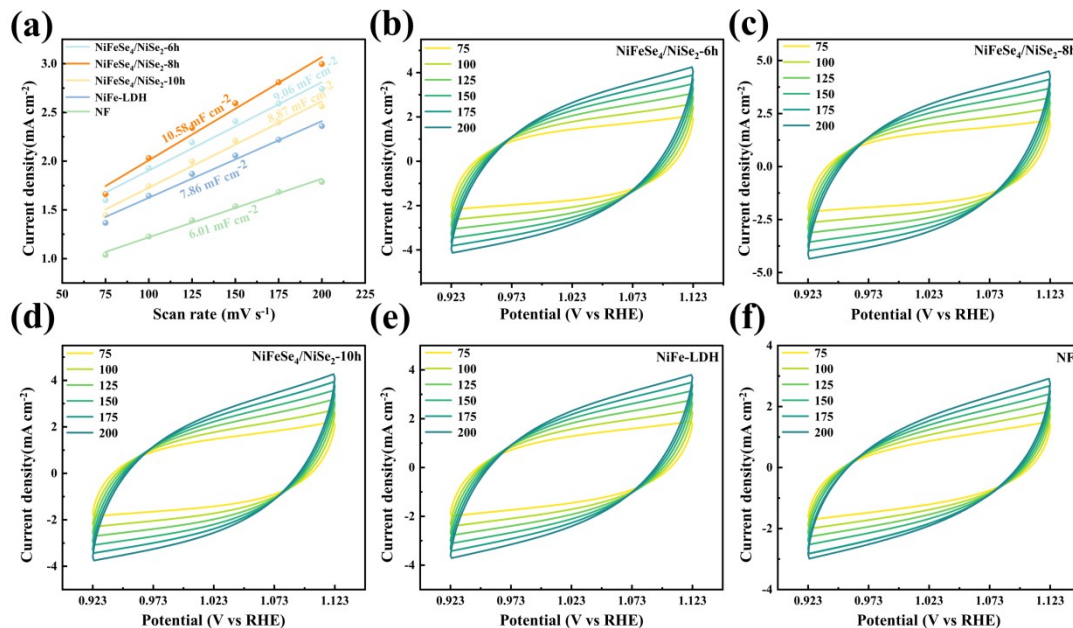
**Figure S3** XRD patterns of NiFe-LDH, NiFeSe<sub>4</sub>/NiSe<sub>2</sub>-6h, NiFeSe<sub>4</sub>/NiSe<sub>2</sub>-8h and NiFeSe<sub>4</sub>/NiSe<sub>2</sub>-10h.



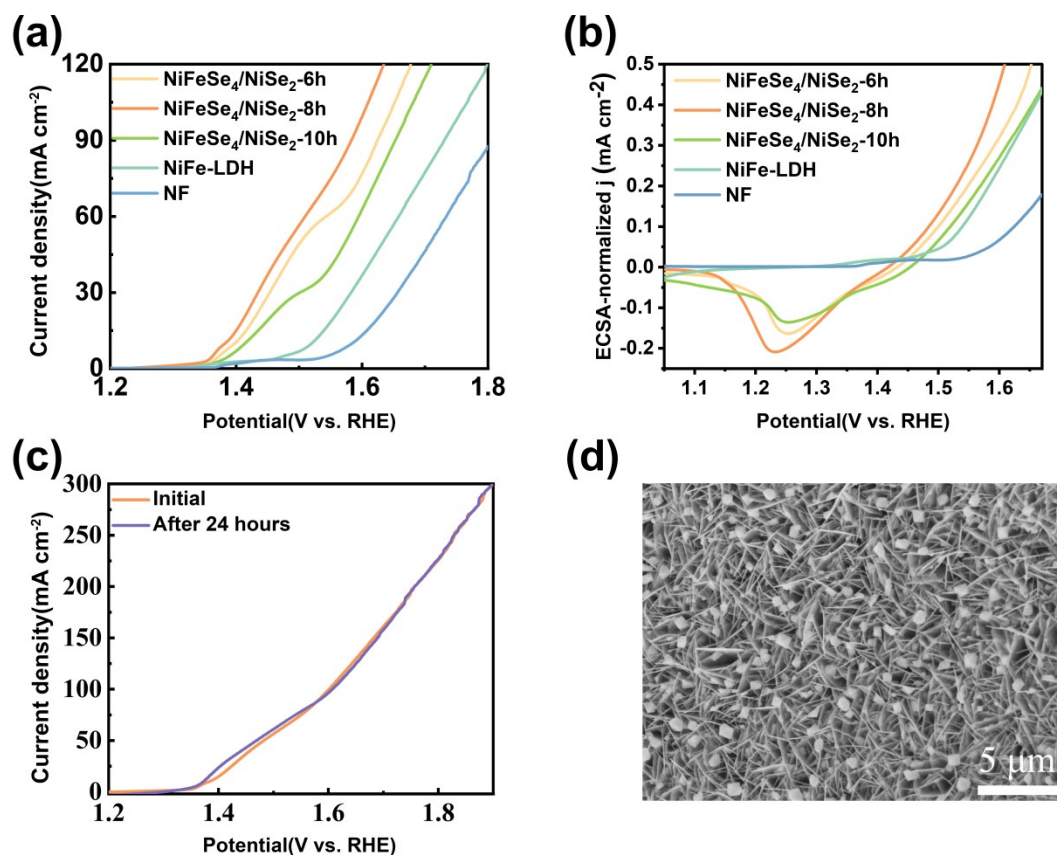
**Figure S4** Crystal structure of NiSe<sub>2</sub> and NiFeSe<sub>4</sub>.



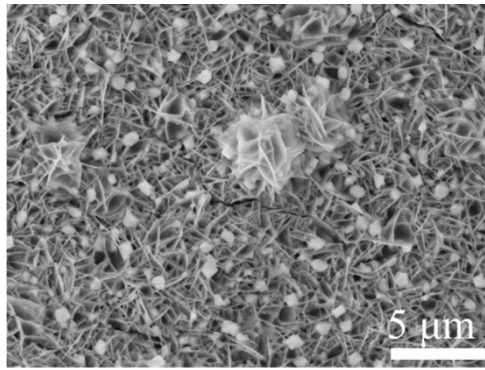
**Figure S5** Equivalent Circuit Model (ECM): Randall circuit model.



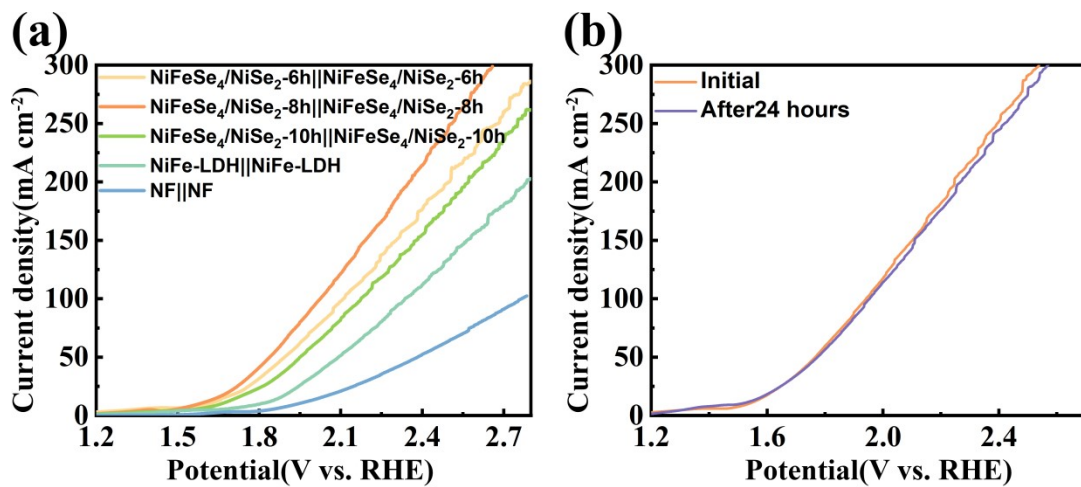
**Figure S6** Shows the double-layer capacitors of NiFeSe<sub>4</sub>/NiSe<sub>2</sub>-6h, NiFeSe<sub>4</sub>/NiSe<sub>2</sub>-8h, NiFeSe<sub>4</sub>/NiSe<sub>2</sub>-10h, NiFe-LDH and NF, respectively.



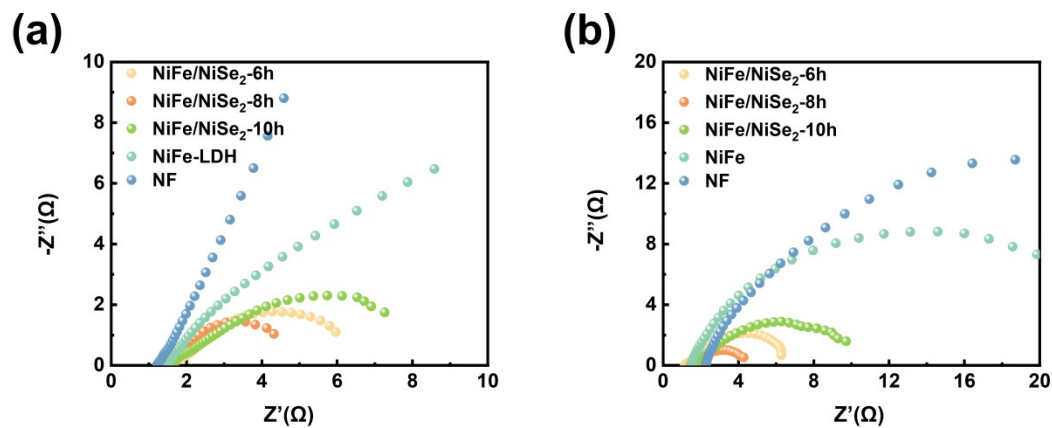
**Figure S7** (a) LSV curves of forward sweep; (b) normalization of current density using ECSA; (c) performance comparison of NiFeSe<sub>4</sub>/NiSe<sub>2</sub>-8h before and after stability test; (d) morphology of NiFeSe<sub>4</sub>/NiSe<sub>2</sub>-8h material after OER stability test.



**Figure S8** Morphology of NiFeSe<sub>4</sub>/NiSe<sub>2</sub>-8h material after HER stability test



**Figure S9** (a) LSV diagrams of NF, NiFe, NiFeSe<sub>4</sub>/NiSe<sub>2</sub>-8h, NiFeSe<sub>4</sub>/NiSe<sub>2</sub>-8h and NiFeSe<sub>4</sub>/NiSe<sub>2</sub>-8h; (b) performance comparison of NiFeSe<sub>4</sub>/NiSe<sub>2</sub>-8h before and after the total water splitting stability test.



**Figure S10:** (a) OER impedance diagram without fitting; (b) HER impedance diagram without fitting



**Table S1** Energy dispersive X-ray spectrum analysis of NiFeSe<sub>4</sub>/NiSe<sub>2</sub>-8h.

element	line style	atomic percent	weight percentage	Mass Error (%) Fit
Fe	K	3.29	2.59	0.71
Ni	K	36.78	30.52	0.24
Se	K	59.93	66.89	0.37
total		100	100	

**Table S2** Reaction impedances of OER and HER for NiFeSe<sub>4</sub>/NiSe<sub>2</sub>-6h, NiFeSe<sub>4</sub>/NiSe<sub>2</sub>-8h, NiFeSe<sub>4</sub>/NiSe<sub>2</sub>-10h, NiFe-LDH and NF.

Catalytic materials	OER			HER				
	R <sub>s</sub> : resistance	Ohmic	R <sub>ct</sub> : transfer resistance	Charge	R <sub>s</sub> : resistance	Ohmic	R <sub>ct</sub> : transfer resistance	Charge
NiFeSe <sub>4</sub> /NiSe <sub>2</sub> -6h		1.859		5.1		1.212		6.137
NiFeSe <sub>4</sub> /NiSe <sub>2</sub> -8h		1.714		3.279		1.542		3.281
NiFeSe <sub>4</sub> /NiSe <sub>2</sub> -10h		1.789		8.652		1.868		8.841
NiFe-LDH		1.45		25.25		1.449		24.18
NF		1.286		979.9		2.148		35.49

**Table S3** overall water splitting performance comparison.

Catalyst	Current density (mA cm <sup>-2</sup> )	Potential(V)	Reference
NiFeSe <sub>4</sub> /NiSe <sub>2</sub> -8h	10	1.569	This work
Ni <sub>1</sub> Fe <sub>10</sub> -LDH@Ni <sub>3</sub> S <sub>2</sub> /NF	10	1.650	1
(Ni,Co) <sub>0.85</sub> Se NSAS	10	1.650	2
Fe <sub>3</sub> O <sub>4</sub> /NiFe LDH/Fe <sub>3</sub> O <sub>4</sub>	10	1.648	3
NiSe <sub>2</sub>	10	1.640	4
NiCo <sub>2</sub> S <sub>4</sub> @NiFe-LDH/NF	10	1.600	5
NiWO <sub>4</sub> @NiSe <sub>2</sub> /NF	10	1.600	6
CoFe/NiFe-LDH	10	1.590	7
Cr-NiSe <sub>2</sub> -N/NF	10	1.590	8
Co <sub>9</sub> S <sub>8</sub> @NiFe-LDH	10	1.585	9

## References

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