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Supporting Information

Polyaniline Induced Multi-functionalities in Interfacially Coupled

Electrocatalysts for Hydrogen/Oxygen Evolution Reactions

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Fig. S1. FESEM images of (a) random distribution between MoS_2 and carbon particles, and (b) nafion masking of MoS_2 /carbon particles/nafion dispersion prepared through ultrasonication method.



Fig. S2. XPS survey spectrum of PANi-CF.



Fig. S3. FESEM image of MoS₂@O-CF.



Fig. S4. XPS survey spectrum of MoS₂@PANi-CF.

Fig. S5. Mo 3d XPS deconvoluted spectra of (a) MoS₂@O-CF and (b) MoS₂.

Fig. S6. Raman spectrum of MoS₂@PANi-CF with a larger range.

Fig. S7. FESEM image of NiFeLDH@O-CF.

Fig. S8. XPS survey spectrum of NiFeLDH@PANi-CF.

Fig. S9. O1s deconvolution XPS spectra of (a) NiFeLDH@O-CF and (b) NiFeLDH.

Fig. S10. Deconvoluted (a) Mo 3d and (b) S 2p XPS spectra of MoS₂@PANi-CF and (c) Mo

3d and (d) S 2p XPS spectra of MoS₂@O-CF prolonged cycling in acidic medium.

Fig. S11. FESEM image of (a) MoS₂@PANi-CF and (b) MoS₂@O-CF after durability test.

Fig. S12. Deconvoluted (a) Ni 2*p* and (b) Fe 2*p* and (c) O 1*s* XPS spectra of NiFeLDH@PANi-CF and (d) Ni 2*p* and (e) Fe 2*p* and (f) O 1*s* XPS spectra of NiFeLDH@O-CF after cycling in alkaline medium.

Fig. S13. FESEM image of NiFeLDH@O-CF after durability test.

Fig. S14. The OER performance of NiFeLDH based catalysts in 0.5 $ext{M}$ H₂SO₄ electrolyte. (a) Polarization curves of NiFeLDH-based catalysts. (b) Tafel slopes of the NiFeLDH-based catalysts derived from the polarization curves of the catalysts. (c) Chronopotentiometric measurement of NiFeLDH catalysts at the current density of 20 mA cm⁻² in 0.5 $ext{M}$ H₂SO₄ electrolyte for 20 h.

Samples		Onset potential [V vs. RHE]	Tafel slope [mV dec ⁻¹]	Reference	
1T/2H MoS ₂		~0.12	110	[S1]	
1T MoS ₂		0.2	42	[S2]	
Ferromagnetic MoS ₂		0.1	59	[\$3]	
Defect rich MoS ₂		0.1	95	[S4]	
Defect rich MoS ₂		~0.15	50	[S5]	
Three-dimensional MoS ₂		0.2	98	[S6]	
MoS ₂ nanoflowers		0.2	52	[S7]	
MoS ₂ nanosheets		~0.25	38	[S8]	
Monolayer MoS ₂		~0.2	53	[\$9]	
Annealed MoS ₂		~0.25	71	[S10]	
MoS ₂ Nanomesh		~0.15	46	[\$11]	
MoS ₂ Nanodots		0.1	61	[812]	
Edge oriented MoS ₂		~0.25	50	[\$13]	
Micro/Nano MoS ₂		~0.17	74	[S14]	
MoS ₂ /graphene		~0.12	71	[\$15]	
MoS ₂ /carbon cloth		~0.1	42	[S16]	
MoS ₂ /carbon foam		~0.25	44	[\$17]	
MoS ₂ /vertical graphene/ carbon cloth		~0.15	53	[S18]	
MoS ₂ /Au/SiO ₂ /Si		~0.15	45	[S19]	
Vacancy-induced MoS ₂ @PANi-CF	acidic	0.03	35	This work	
	alkaline	0.04	40		
	neutral	0.05	31		

Table S1. The HER performance of the MoS_2 based catalysts.

Samples		Onset potential [V vs. RHE]	Tafel slope [mV dec ⁻¹]	Reference	
NiFeLDH/CNT		1.45	31	[S20]	
Three-dimensional NiFeLDH		1.46	40 [S21]		
NiFeLDH/Graphene		1.43	39	[S22]	
NiFeLDH/Carbon qua	antum dot	1.43	35 [\$23]		
NiFeLDH hollow s	pheres	1.45	53	[S24]	
NiFeLDH/Mesoporous oxide nanosphe	s graphene eres	1.50	63	[\$25]	
NiFeLDH/Reduced grap	ohene oxide	1.50	91	[S26]	
Plasma assisted oxyger NiFeLDH	n enriched	1.48	74	[S27]	
NiFeLDH edge a	ctive	1.47	35	[S28]	
NiFeLDH with oxygen	vacancies	1.45	48	[S29]	
Edge-enriched NiFeLDH		1.45	41	[S30]	
NiFeLDH/carbon	cloth	1.50	56	[\$31]	
NiFeLDH/sulfonated c	arbon dots	1.42	55	[832]	
NiFeLDH edge	rich	1.52	49	[\$33]	
Vacancy-induced	alkaline	1.44	47	This work	
NiFeLDH@PANi-CF	acidic	1.48	76		

Table S2. The OER performance of the NiFeLDH based catalysts.

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