Supporting Information

Epitaxial Growth of $Ni(OH)_2$ Nanoclusters on MoS_2 Nanosheets for Enhanced Alkaline Hydrogen Evolution Reaction

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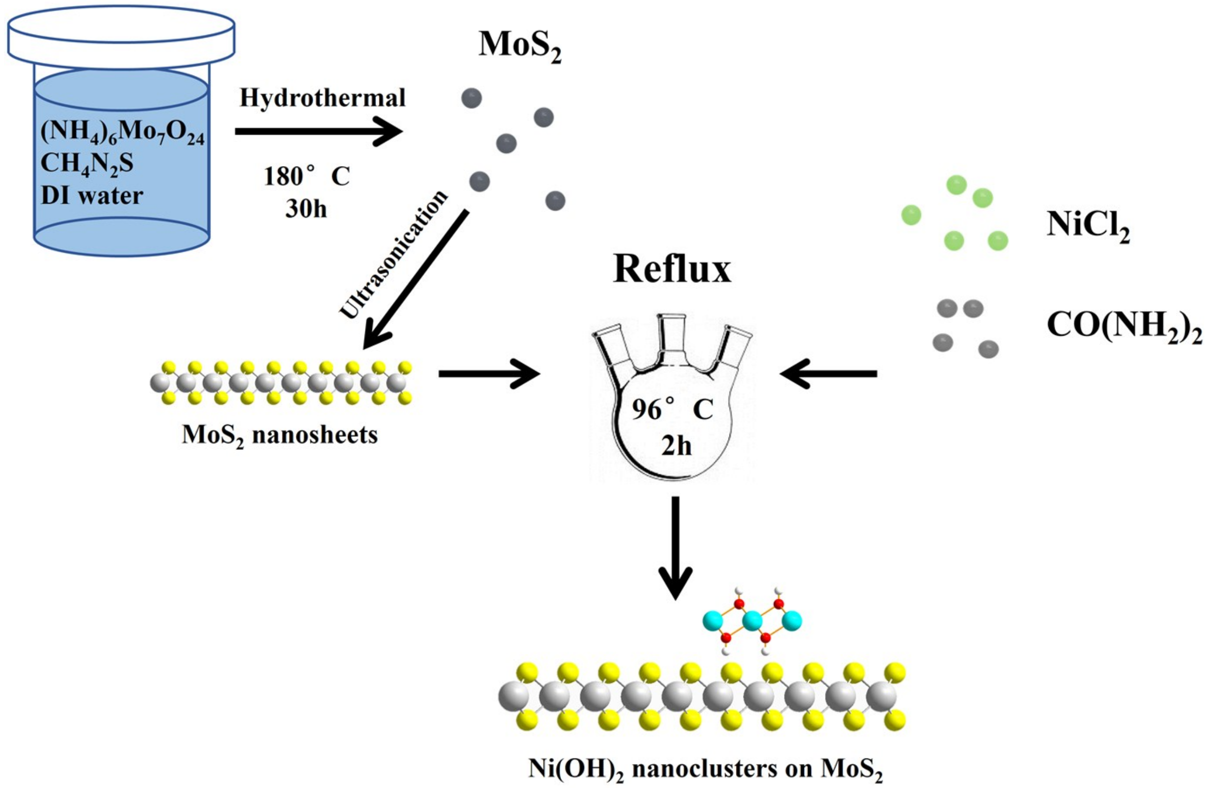

Fig. S1 Illustration scheme for materials synthesizing procedures.

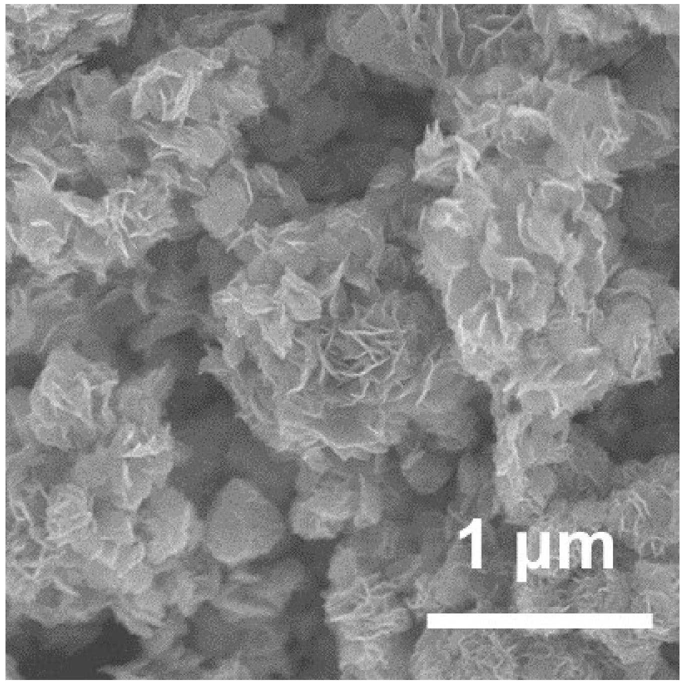

Fig. S2 SEM image of the pure MoS_2.

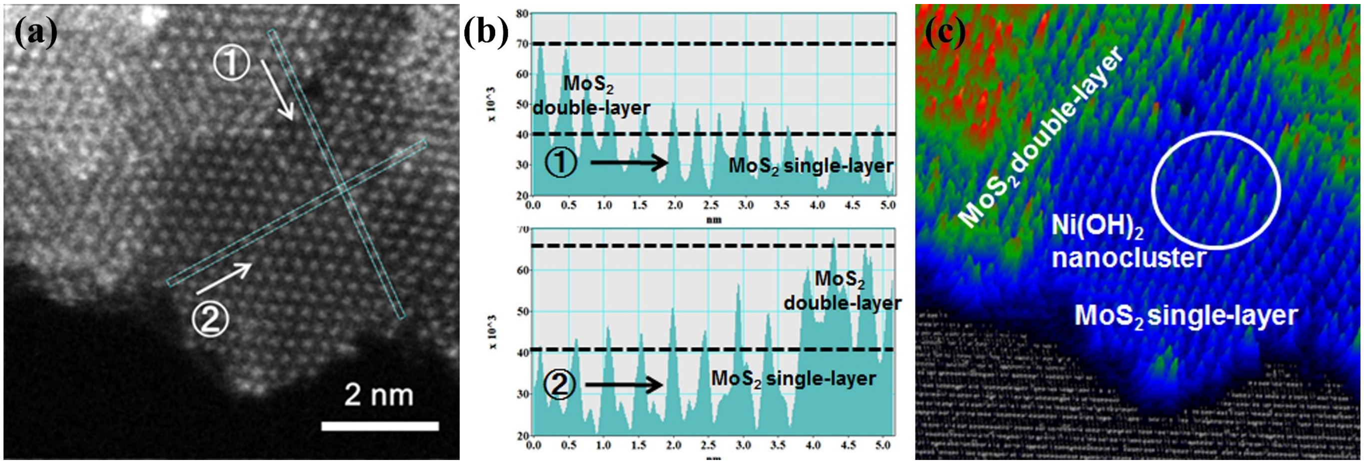

Fig. S3 (a) HAADF-STEM image of 0.2NM, the arrows indicate the direction of the intensity profiles. (b) the corresponding intensity profiles, showing a nanodomain with an average intensity between single-layer MoS_2 and double-layer MoS_2. (c) the corresponding surface plot image, illustrating $Ni(OH)_2$ nanocluster on the surface of single-layer MoS_2.

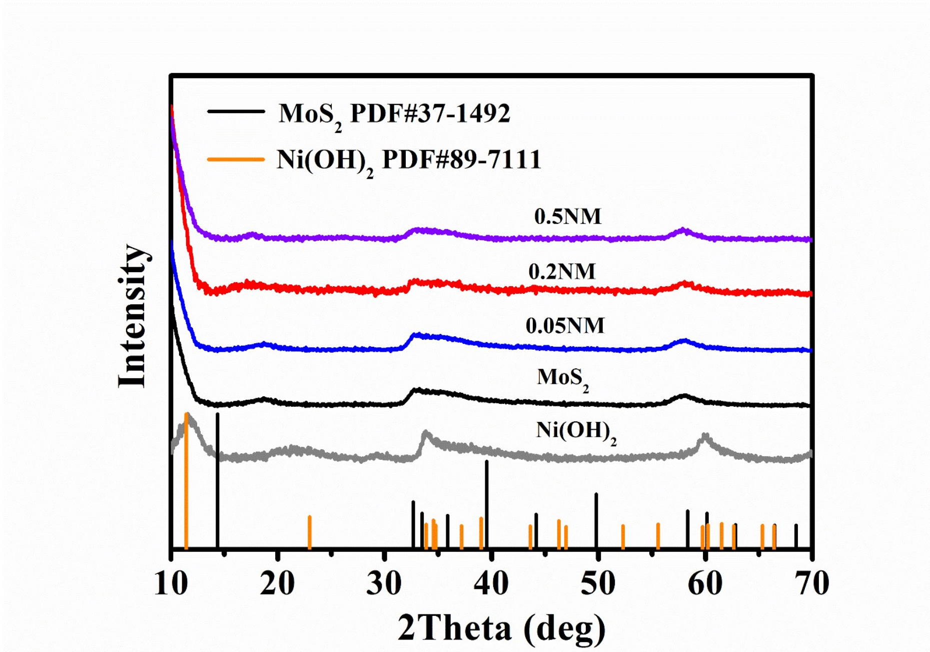

Fig. S4 XRD patterns of $Ni(OH)_2$, MoS_2, and $Ni(OH)_2/MoS_2$ heterostructures

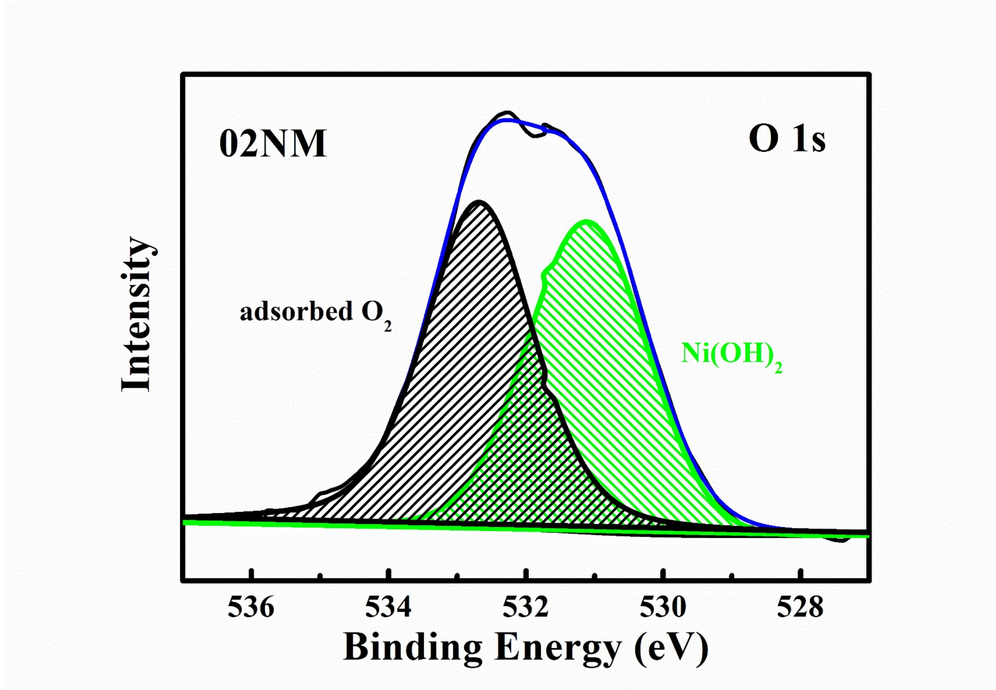

Fig. S5 The fitted XPS spectra of 0.2NM.

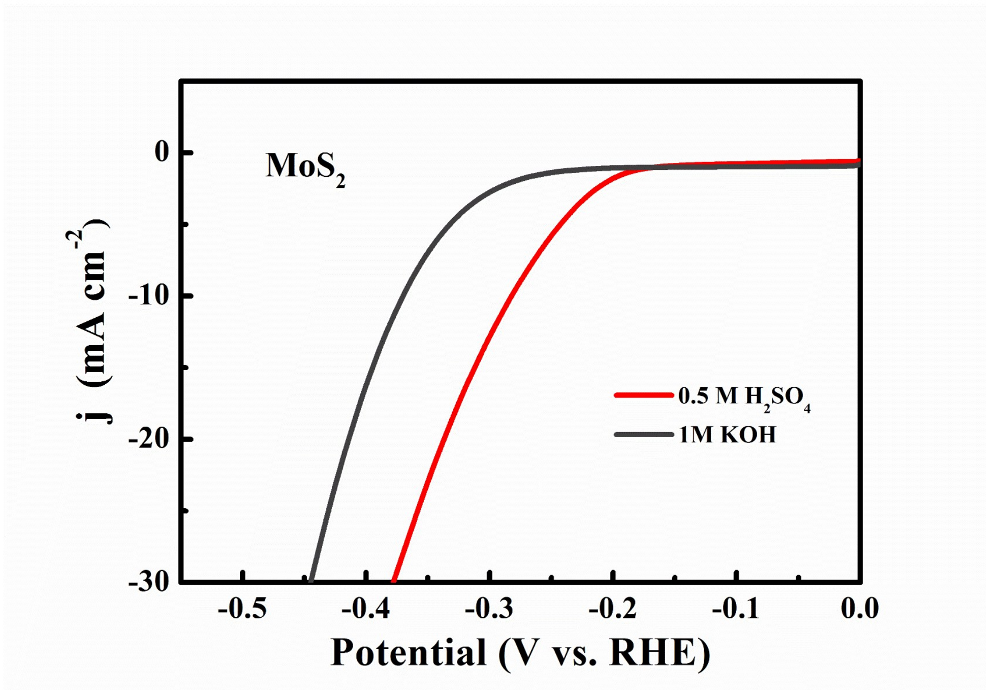

Fig. S6 LSV curves of the pure MoS_2 in 0.5M H_2SO_4 and 1M KOH.

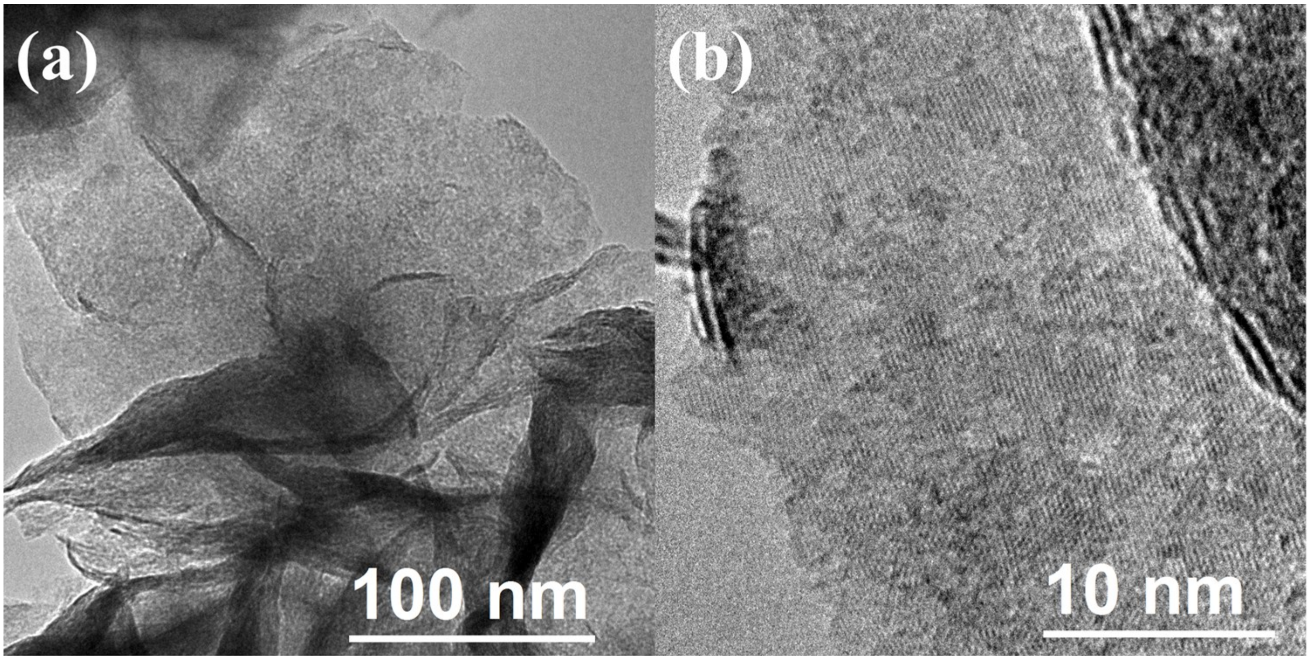

Fig. S7 (a) TEM and (b) HRTEM image of 0.2NM after stability test.

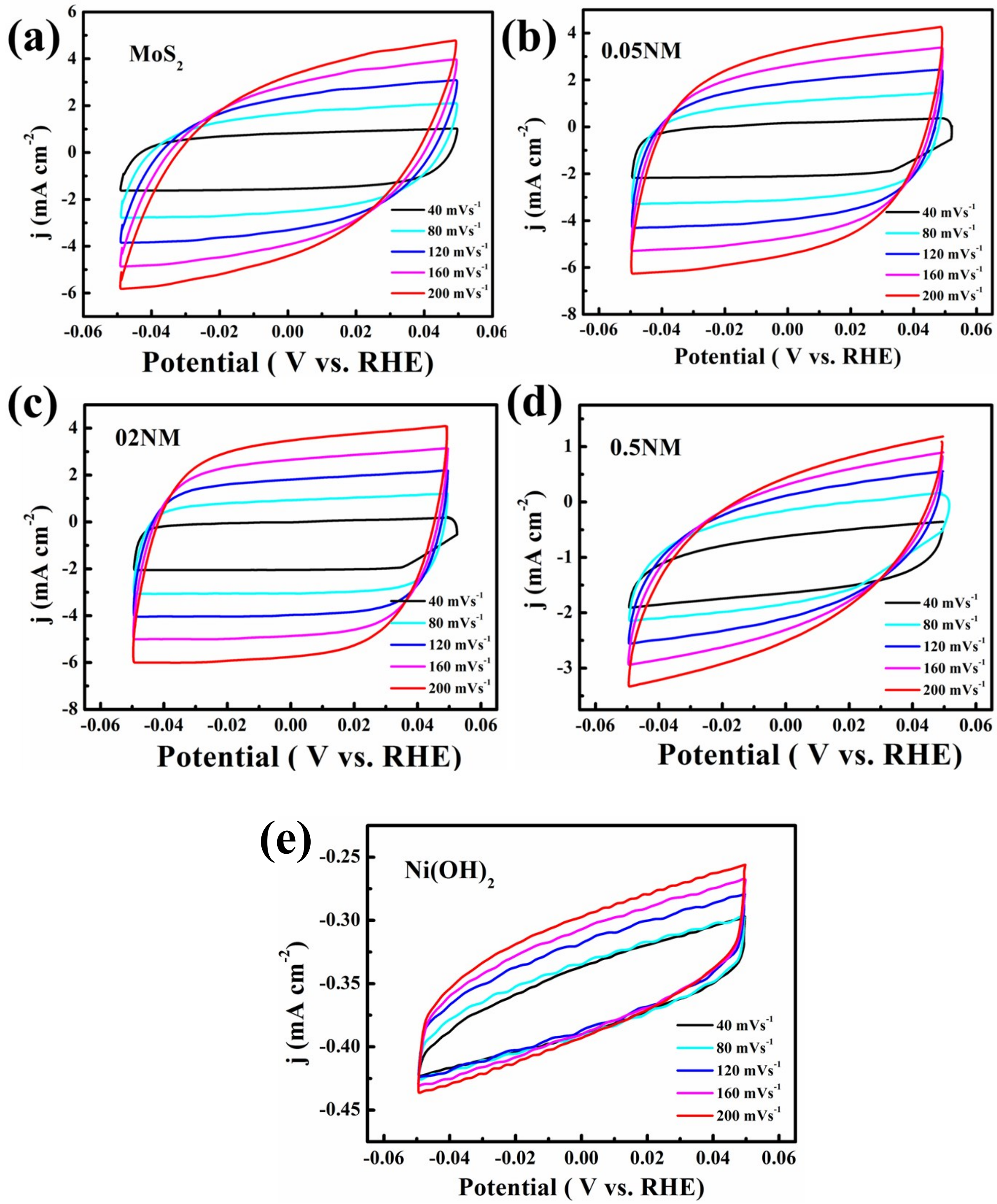

Fig. S8 CV curves of (a) MoS_2, (b) 0.05NM, (c) 0.2NM, (d) 0.5NM, and (e) $Ni(OH)_2$ at various scan rates

Note S1.

The atomic number (Z) of Ni and Mo are 28 and 42, respectively. For the $Ni(OH)_2/MoS_2$ heterostructures, the theoretical intensity ratio of the heterostructure to the MoS_2 surface is 1.44 (Eq. S1).

$$(Z_{Mo}^2 + Z_{Ni}^2)\big/ Z_{Mo}^2 = 1.44 \qquad \text{(S1)}$$

In Fig. 1(h) in the manuscript, the average intensity at the Mo sites of the single layer is ~ 275 × 10^3 counts while the average intensity of the $Ni(OH)_2/MoS_2$ heterostructure is ~ 350 × 10^3 counts. The average intensity of the background is ~ 90 × 10^3 counts. Therefore, the practical intensity ratio of the heterostructure to the MoS_2 is calculated to be 1.41, as shown in Eq. S2.

$$\frac{(350-90)\times 10^3}{(275-90)\times 10^3} = 1.41 \qquad \text{(S2)}$$

The practical intensity ratio of the proposed $Ni(OH)_2/MoS_2$ heterostructure is in good accordance with the theoretical value.

Table S1. The fitted equivalent elements for EIS of MoS_2 and 0.2NM

	R_s	R_{ad}	Q	n	R_{ct}	Q	n
MoS_2	4.163	31.77	0.00076	0.559	5010	0.00329	0.874
0.2NM	4.089	7.645	0.00602	0.379	141.3	0.00247	0.898

Table S2. HER activities of reported electrocatalysts in alkaline media

Catalysts	Electrolyte	Substrate	Overpotential (@10 mA cm^{-1})	Tafel slope	Mass loading	Reference
$Ni(OH)_2/MoS_2$	**1M KOH**	**GC**	**227**	**105**	**0.204**	**This work**
Commercial 20% Pt/C	1MKOH	GC	43	45		1
Commercial 20% Pt/C	1MKOH	NF	40	75	2.8	2
$CoSe_2/MoSe_2$	1MKOH	GC	218	76	0.204	3
NiS_2 nanocrystal	1MKOH	GC	540	139		4
$CoNi_2S_4$	1MKOH	GC	~270	85	0.213	5
Ni_3S_2	1MKOH	GC	335	97		6
$MoSe_2$	0.5M KOH	GC	310	93	1	7
$Mo_{0.75}W_{0.25}S_2$	0.5 M KOH	GC	264	84	1	7
$NiMo_3S_4$	0.1M KOH	GC	257	98	0.3	8
$CoNi_2S_4$	1MKOH	GC	~270	85	0.213	5
3D $MoSe_2@Ni_{0.85}Se$ Nanowire	1M KOH	NF	117	66	6.48	9
$MoS_2/NiS/MoO_3$	1M KOH	Ti Foil	91	55		10
$Ni\text{-}P/MoS_x$	1M KOH	FTO	140	64		11
MoS_2/NiCo-LDH	1M KOH	CFP	78	77		12
$Ni(OH)_2/Fe_2P$	1M KOH	Ti Mesh	76	105		13
$Pt/Ni(OH)_2$	1M KOH	CC	~150			14
MoS_2	1M KOH	CC	212	96		15
$MoS_2@Ni(OH)_2$	1M KOH	CC	80	60		15
Ni doped MoS_2	1M KOH	CC	98	60		16
$Co\text{-}MoS_2$	1M KOH	CC	203	158		16

GC: glassy carbon; CC: carbon cloth; NF: nickel foam; FTO: fluorine-doped tin oxide; CFP: carbon fiber paper

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