

**Controlled synthesis of bifunctional particle-like Mo/Mn-Ni<sub>x</sub>S<sub>y</sub>/NF  
electrocatalyst for highly efficient overall water splitting**

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**Table. S1** Comparison of HER properties for catalysts of superior electrochemistry materials

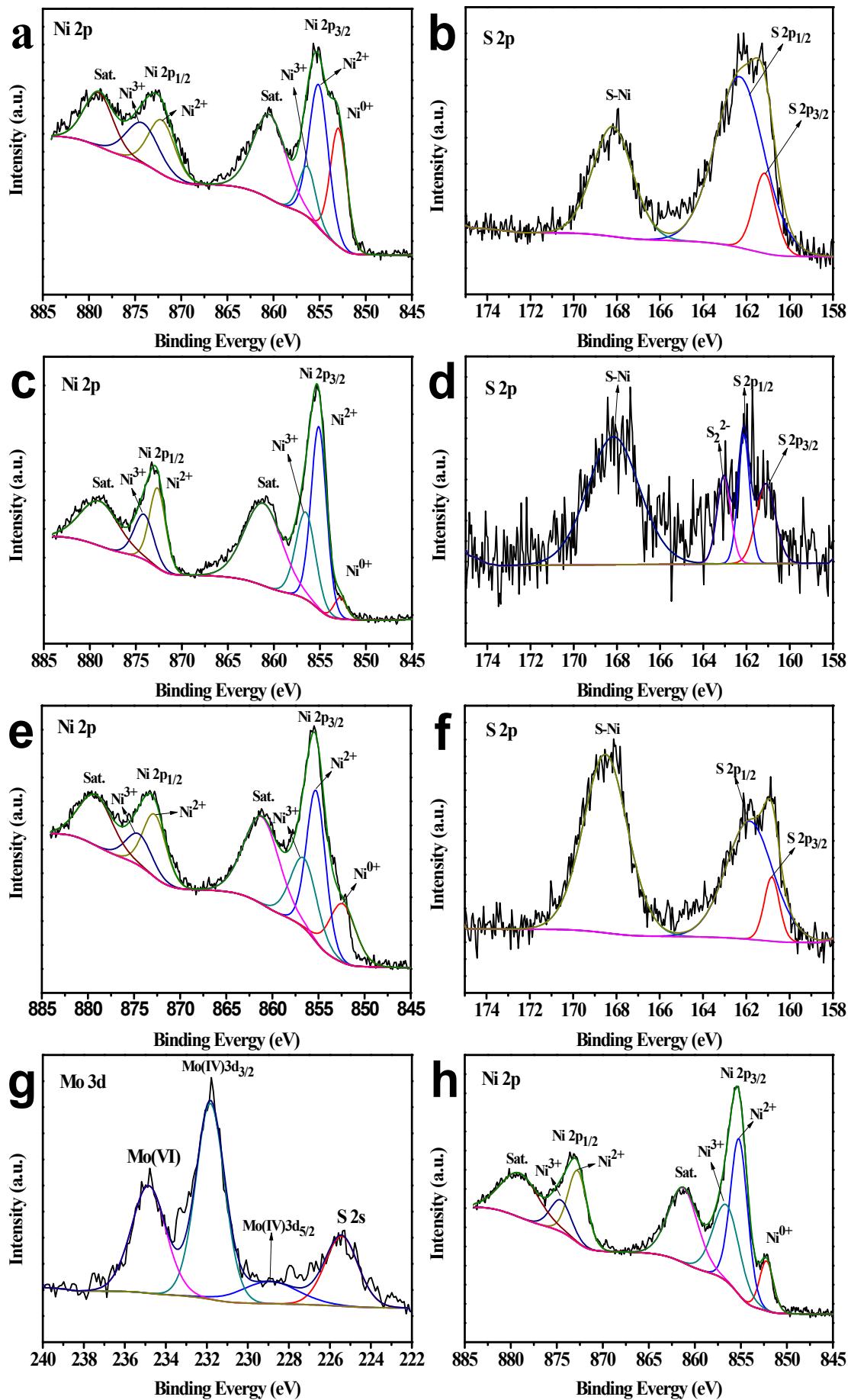
Catalyst	Overpotential (mV vs. RHE)	Ref
Mo/Mn-Ni <sub>x</sub> S <sub>y</sub> /NF	144 mV@10 mA cm <sup>-2</sup>	This work
Mo, S- codoping NiSe	88 mV@10 mA cm <sup>-2</sup>	[1]
Sn-Ni <sub>3</sub> S <sub>2</sub> /NF	170 mV@100 mA cm <sup>-2</sup>	[2]
2H-MoS <sub>2</sub> /G/NF	117 mV@10 mA cm <sup>-2</sup>	[3]
Ni <sub>x</sub> Co <sub>3-x</sub> S <sub>4</sub> /Ni <sub>3</sub> S <sub>2</sub>	136 mV@10 mA cm <sup>-2</sup>	[4]
Co <sub>1</sub> Mn <sub>1</sub> CH/NF	180 mV@10 mA cm <sup>-2</sup>	[5]
MoS <sub>2</sub> -Ni <sub>3</sub> S <sub>2</sub>	211mV@10 mA cm <sup>-2</sup>	[6]
MoSe <sub>2</sub> -CoSe <sub>2</sub>	148 mV@10 mA cm <sup>-2</sup>	[7]
CoS <sub>x</sub> /Ni <sub>3</sub> S <sub>2</sub>	204 mV@10 mA cm <sup>-2</sup>	[8]
P-Doped Co-Ni-S	187 mV@100 mA cm <sup>-2</sup>	[9]
Er-doped CoP NMs	66 mV@10 mA cm <sup>-2</sup>	[10]
Mo, S-codoping NiSe/NF	88 mV@10 mA cm <sup>-2</sup>	[22]
N,S-codoped Zn <sub>0.975</sub> Co <sub>0.025</sub> S/CoS <sub>2</sub>	152 mV@10 mA cm <sup>-2</sup>	[23]

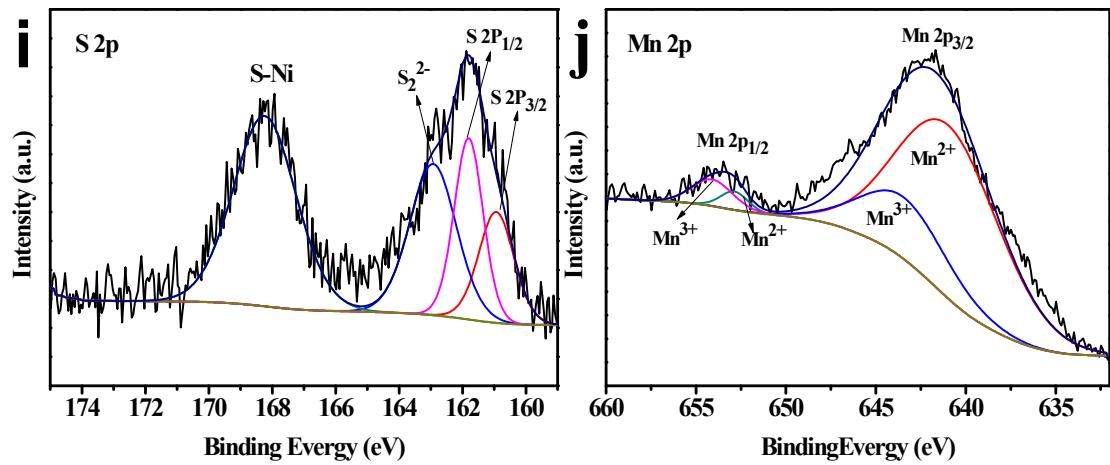
**Table. S2** Comparison of OER properties for catalysts of superior electrochemistry materials

Catalyst	Overpotential (mV vs. RHE)	Ref
Mo/Mn-Ni <sub>x</sub> S <sub>y</sub> /NF	162 mV @ 50 mA cm <sup>-2</sup>	This work
CoS <sub>x</sub> /Ni <sub>3</sub> S <sub>2</sub>	280 mV@ 20 mA cm <sup>-2</sup>	[8]
Er-doped CoP NMs	256mV@10 mA cm <sup>-2</sup>	[10]
MoS <sub>2</sub> /NiS	271 mV @10 mA cm <sup>-2</sup>	[11]
CoOOH	262 mV @10 mA cm <sup>-2</sup>	[12]
Fe-doped Ni <sub>3</sub> S <sub>2</sub>	253 mV @100 mA cm <sup>-2</sup>	[13]
Ni <sub>x</sub> Co <sub>3-x</sub> S <sub>4</sub> /Ni <sub>3</sub> S <sub>2</sub>	160 mV@10 mA cm <sup>-2</sup>	[4]
CS-NiFeCu	180 mV@10 mA cm <sup>-2</sup>	[14]
NiS <sub>1.03</sub> -NSCs	270 mV@10 mA cm <sup>-2</sup>	[15]
Cu <sub>2</sub> S/CF	336 mV@20 mA cm <sup>-2</sup>	[16]
Ce-doped NiFe-LDH/CNT	227 mV@10 mA cm <sup>-2</sup>	[17]
P-Doped Co-Ni-S	292 mV@100 mA cm <sup>-2</sup>	[9]
Sn-Ni <sub>3</sub> S <sub>2</sub> /NF	270 mV@100 mA cm <sup>-2</sup>	[2]
N,S-codoped Zn <sub>0.975</sub> Co <sub>0.025</sub> S/CoS <sub>2</sub>	270 mV@10 mA cm <sup>-2</sup>	[23]
P,N Co-doped PNGF	320 mV@10 mA cm <sup>-2</sup>	[24]

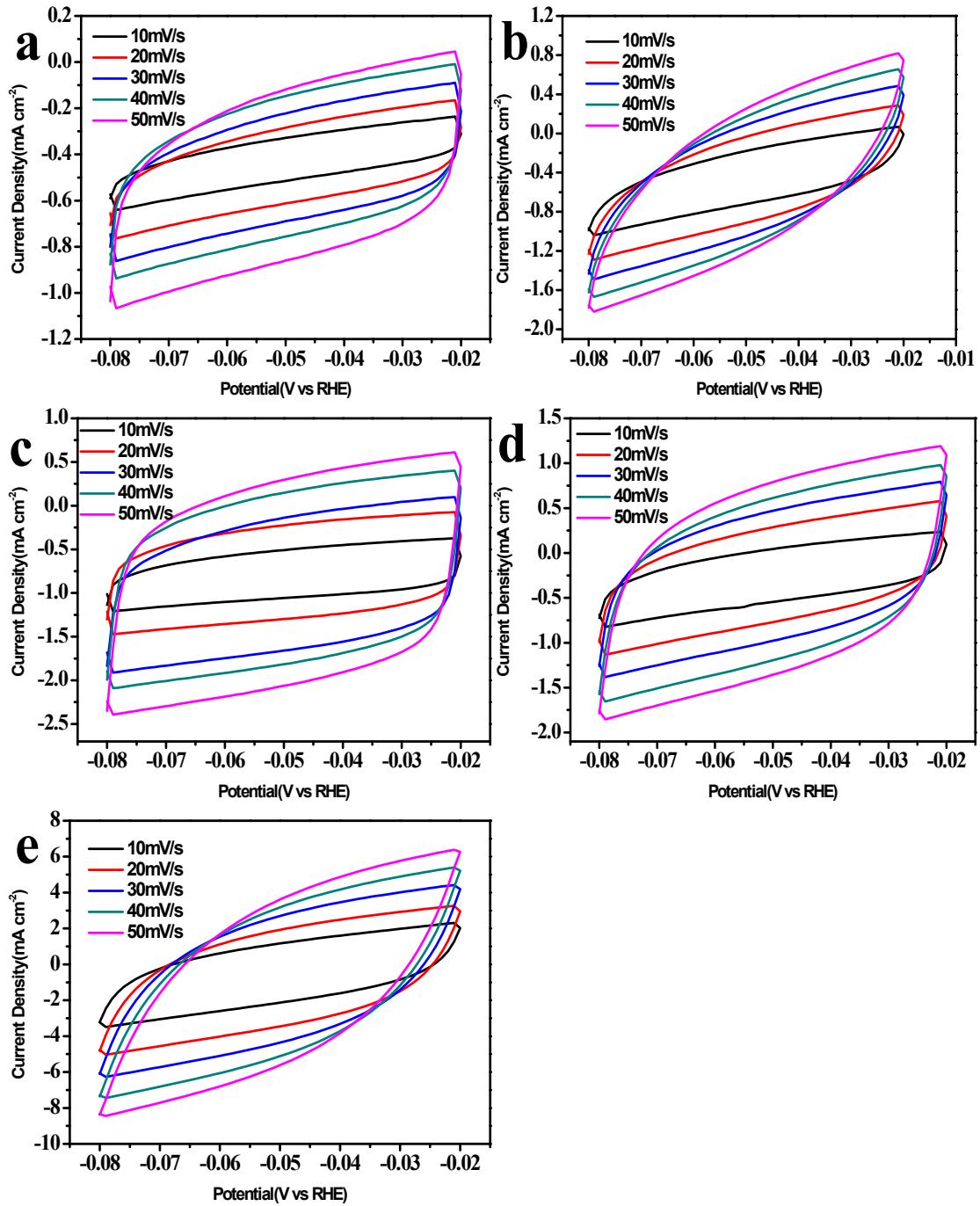
**Table. S3** Comparison of overall water splitting properties for catalysts of superior electrochemistry materials

Catalyst	Overpotential (mV vs. RHE)	Ref
Mo/Mn-Ni <sub>x</sub> S <sub>y</sub> /NF	1.49 V @ 10 mA cm <sup>-2</sup>	This work
Er-doped CoP NMs	1.58 V@10 mA cm <sup>-2</sup>	[10]
MoS <sub>2</sub> /NiS	1.61 V @10 mA cm <sup>-2</sup>	[9]
Ni <sub>x</sub> Co <sub>3-x</sub> S <sub>4</sub> /Ni <sub>3</sub> S <sub>2</sub>	1.53 V@10 mA cm <sup>-2</sup>	[4]
Co <sub>1</sub> Mn <sub>1</sub> CH/NF	1.68 V@10 mA cm <sup>-2</sup>	[5]
Zn-doped Co <sub>3</sub> O <sub>4</sub>	1.39 V@10 mA cm <sup>-2</sup>	[18]
MoP/Ni <sub>2</sub> P	1.55 V@10 mA cm <sup>-2</sup>	[19]
Co <sub>3</sub> O <sub>4</sub> @MoS <sub>2</sub> /CC	1.59 V@10 mA cm <sup>-2</sup>	[20]
NiFe-P/NF	1.56 V@10 mA cm <sup>-2</sup>	[21]
CoS <sub>x</sub> /Ni <sub>3</sub> S <sub>2</sub>	1.57 V@10 mA cm <sup>-2</sup>	[8]
P-Doped Co-Ni-S	1.60 V@10 mA cm <sup>-2</sup>	[9]
Sn-Ni <sub>3</sub> S <sub>2</sub> /NF	1.46 V@10 mA cm <sup>-2</sup>	[2]
N,S-codoped Zn <sub>0.975</sub> Co <sub>0.025</sub> S/CoS <sub>2</sub>	1.59 V@10 mA cm <sup>-2</sup>	[23]

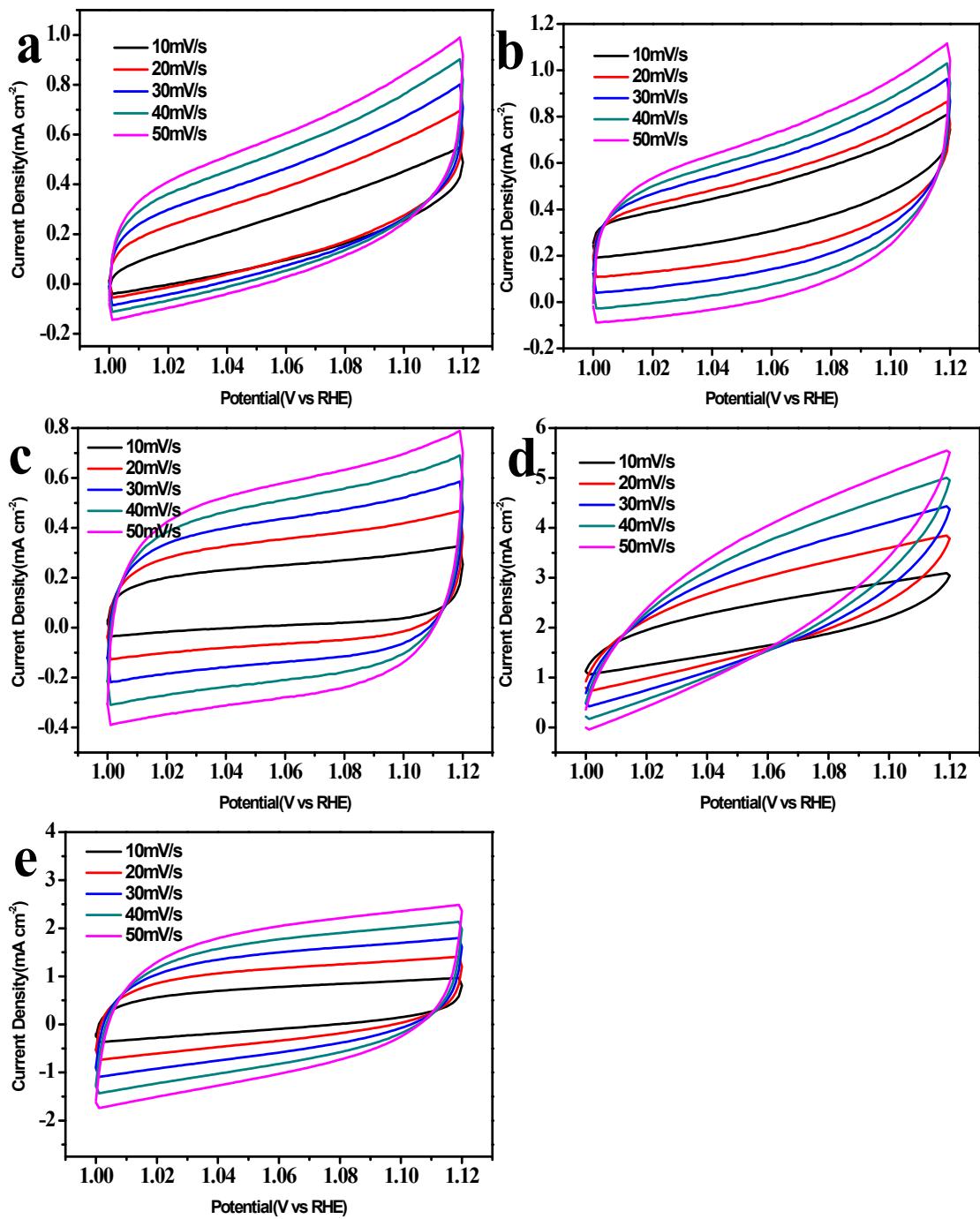




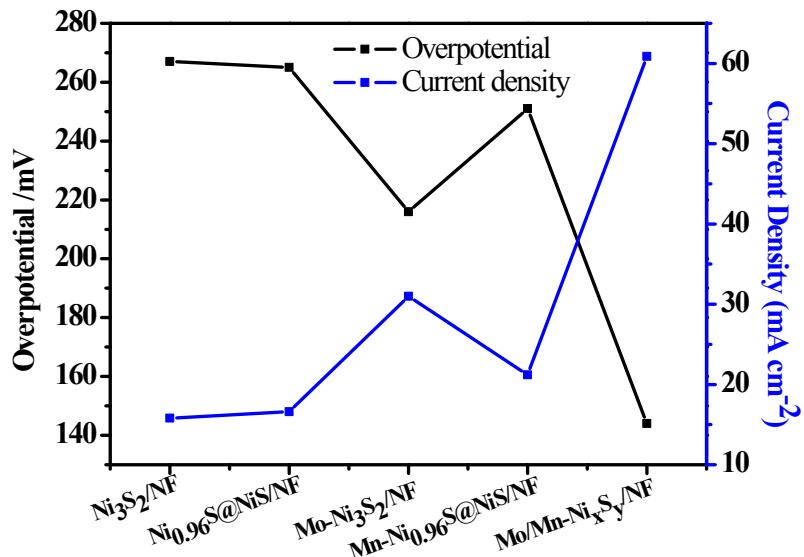
**Fig. S1.** High-magnification XPS spectra (a) Ni 2p and (b) S 2p of  $\text{Ni}_3\text{S}_2/\text{NF}$ , (c) Ni 2p and (d) S 2p of  $\text{NiS@Ni}_{0.96}\text{S}/\text{NF}$ , (e) Ni 2p, (f) S 2p and (g) Mo 3d of  $\text{Mo-Ni}_3\text{S}_2/\text{NF}$ , (h) Ni 2p, (i) S 2p, and (j) Mn 2p of  $\text{Mn-NiS@Ni}_{0.96}\text{S}/\text{NF}$ .



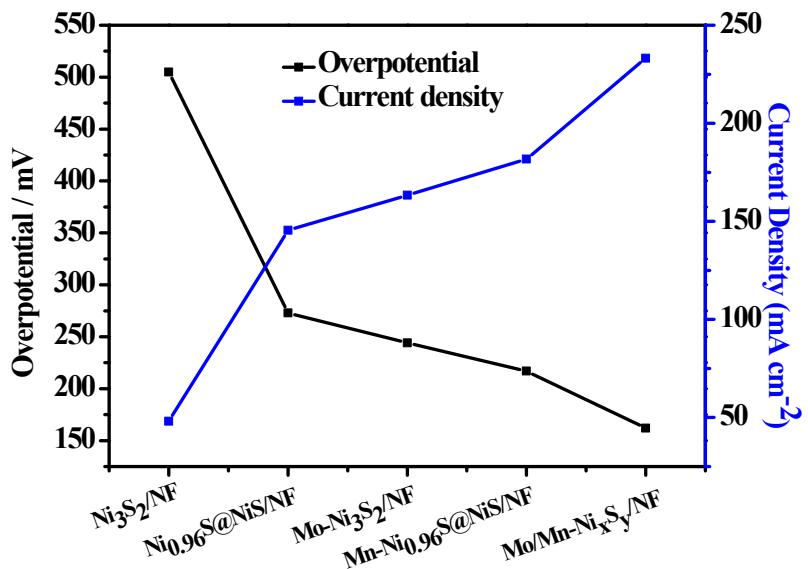
**Fig. S2.** CVs for HER of (a)  $\text{Ni}_3\text{S}_2/\text{NF}$ ; (b)  $\text{NiS}@\text{Ni}_{0.96}\text{S}/\text{NF}$ , (c)  $\text{Mo-Ni}_3\text{S}_2/\text{NF}$ , (d)  $\text{Mn-NiS}@\text{Ni}_{0.96}\text{S}/\text{NF}$ , (e)  $\text{Mo/Mn-Ni}_x\text{S}_y/\text{NF}$  with different scan rates ( $10-50 \text{ mV s}^{-1}$ ) in the region from -1.08 to -1.02 vs RHE.



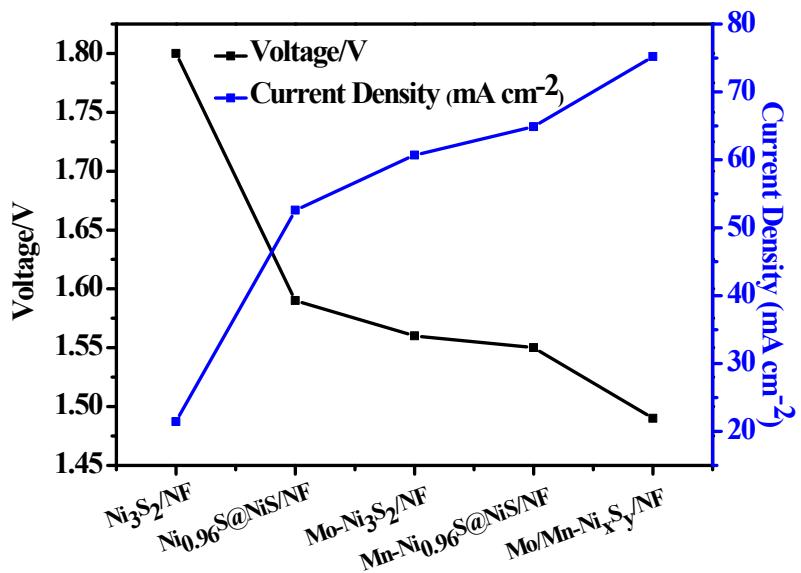
**Fig. S3.** CVs for OER of (a)  $\text{Ni}_3\text{S}_2/\text{NF}$ ; (b)  $\text{NiS}@\text{Ni}_{0.96}\text{S}/\text{NF}$ ; (c)  $\text{Mo}-\text{Ni}_3\text{S}_2/\text{NF}$ ; (d)  $\text{Mn}-\text{NiS}@\text{Ni}_{0.96}\text{S}/\text{NF}$ , (e)  $\text{Mo/Mn-Ni}_x\text{S}_y/\text{NF}$  with different scan rates ( $10-50 \text{ mV s}^{-1}$ ) in the region from 1.00 to 1.12 vs RHE.



**Fig. S4** Comparison of the overpotentil (HER) required for achieving current density of 10 mA cm<sup>-2</sup>, and the current density at overpotential of 300 mV for Ni<sub>3</sub>S<sub>2</sub>/NF, NiS@Ni0.96S/NF, Mo-Ni<sub>3</sub>S<sub>2</sub>/NF, Mn-NiS@Ni0.96S/NF, and Mo/Mn-Ni<sub>x</sub>S<sub>y</sub>/NF.



**Fig. S5** Comparison of the overpotentil (OER) required for achieving current density of 50 mA cm<sup>-2</sup>, and the current density at overpotential of 500 mV for Ni<sub>3</sub>S<sub>2</sub>/NF, NiS@Ni0.96S/NF, Mo-Ni<sub>3</sub>S<sub>2</sub>/NF, Mn-NiS@Ni0.96S/NF, and Mo/Mn-NixSy/NF.



**Fig. S6** Comparison of the voltage required for achieving current density of 10 mA cm<sup>-2</sup>, and the current density at voltage of 1.9 V for Ni<sub>3</sub>S<sub>2</sub>/NF, NiS@Ni<sub>0.96</sub>S/NF, Mo-Ni<sub>3</sub>S<sub>2</sub>/NF, Mn-NiS@Ni<sub>0.96</sub>S/NF, and Mo/Mn-Ni<sub>x</sub>S<sub>y</sub>/NF.

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