

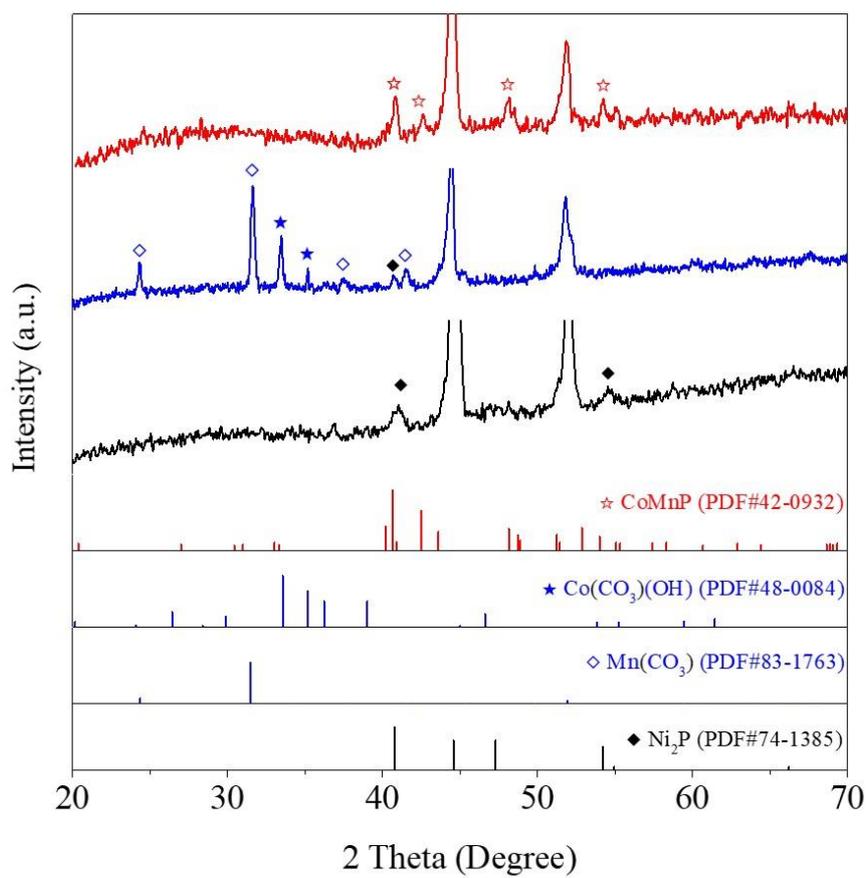
## Supporting Information

# A Unique Sandwich Structure of CoMnP/Ni<sub>2</sub>P/NiFe electrocatalyst for Highly Efficient Overall Water Splitting

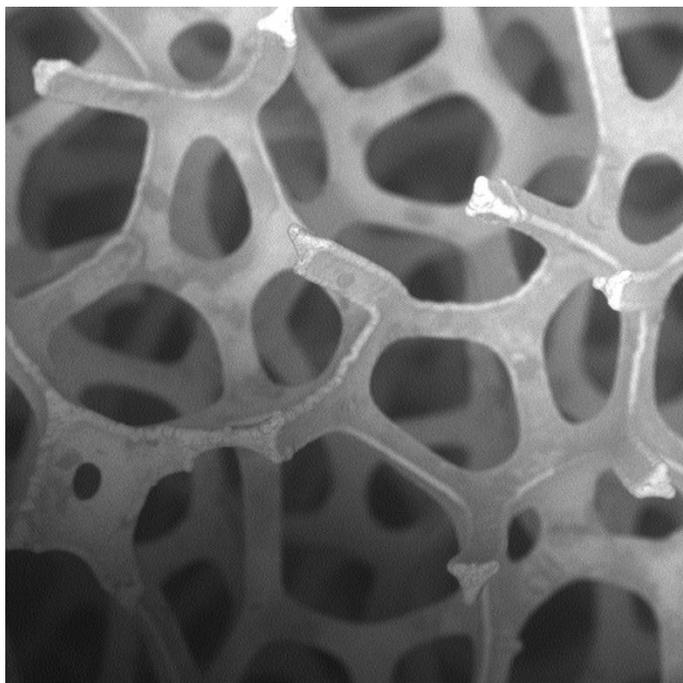
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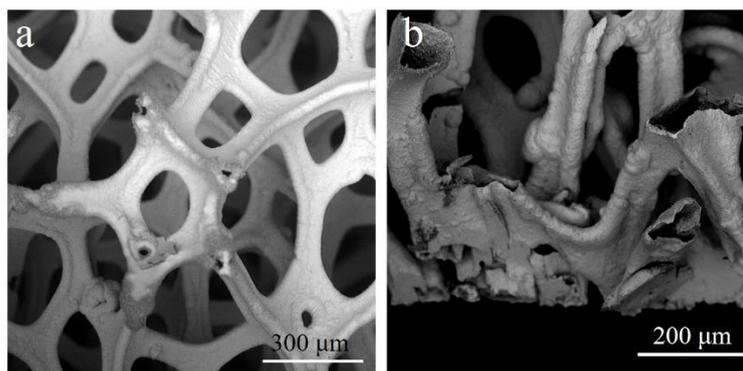
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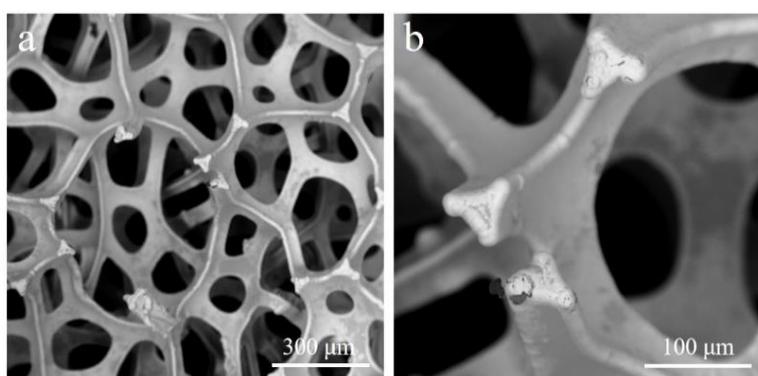
**Fig S1.** XRD patterns of Ni<sub>2</sub>P (red), CoMnCH (blue) and Ni<sub>2</sub>P/CoMnP (black) on NiFe foam.



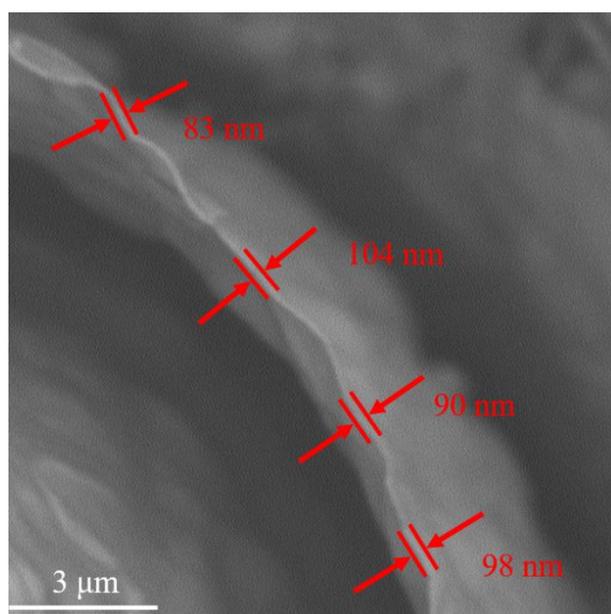
**Fig S2.** The SEM image of a NiFe foam.



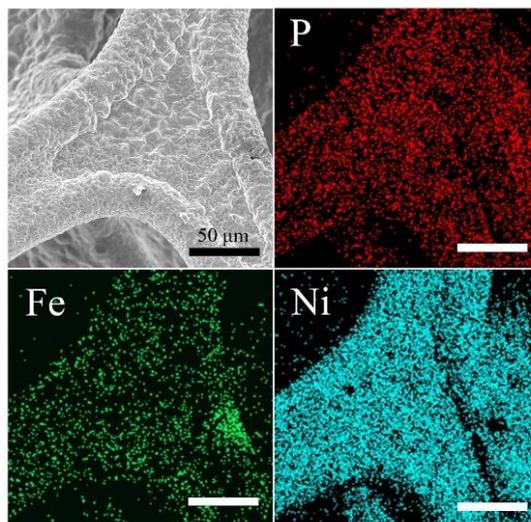
**Fig S3.** The SEM image of NiFe foam after acid treatment: top view (a) and side view (b).



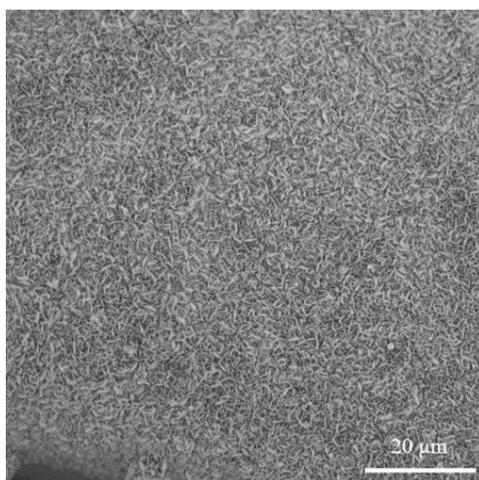
**Fig S4.** SEM images of the nickel foam processed with the same condition of acid treatment as compared with the one of NiFe foam. The Ni foam is found to maintain a complete structure, rather than a hollow structure, suggesting that the formation of the hollow structure on NiFe foam substrates can be mainly attributed to the different dissolution rate of Ni and Fe during the acid treatment.



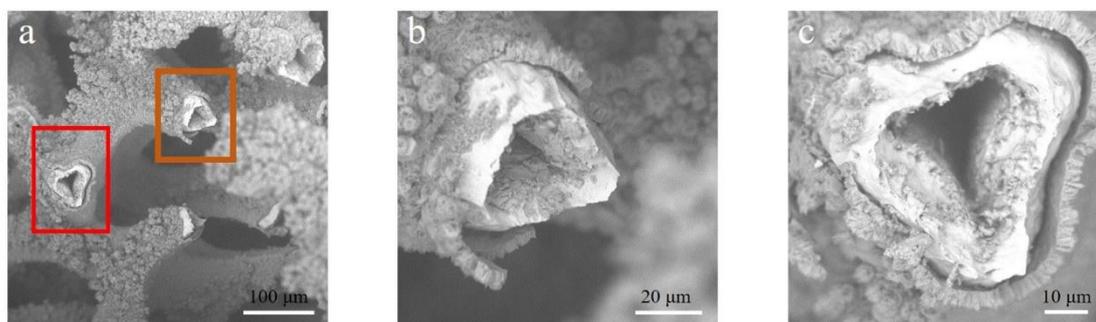
**Fig S5.** The thickness of the Ni<sub>2</sub>P interlayer estimated from the SEM image.



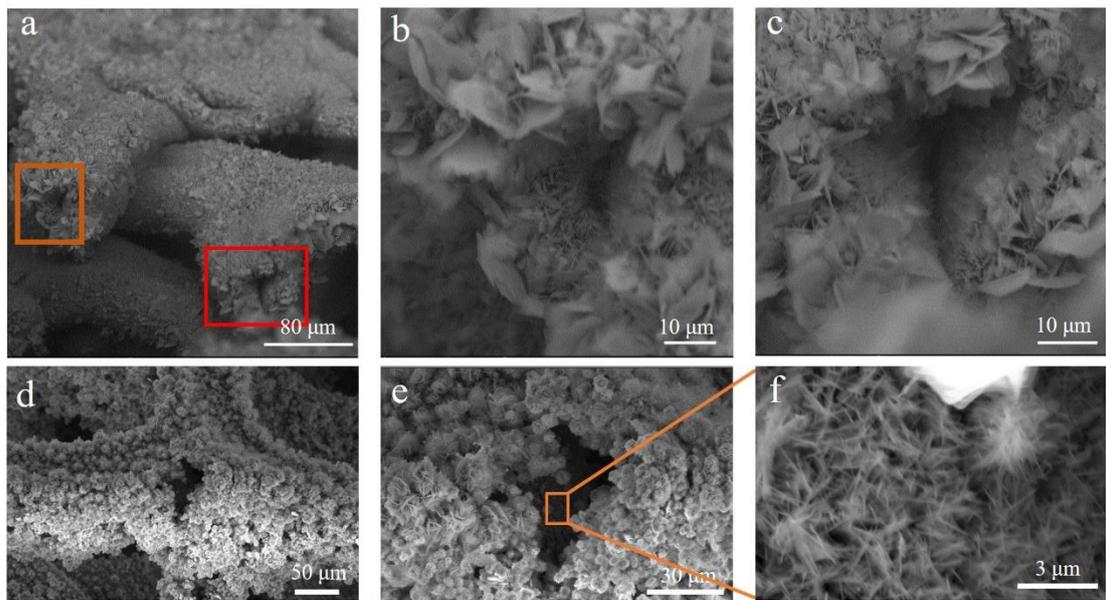
**Fig S6.** The SEM images of Ni<sub>2</sub>P/NiFe and corresponding EDX images.



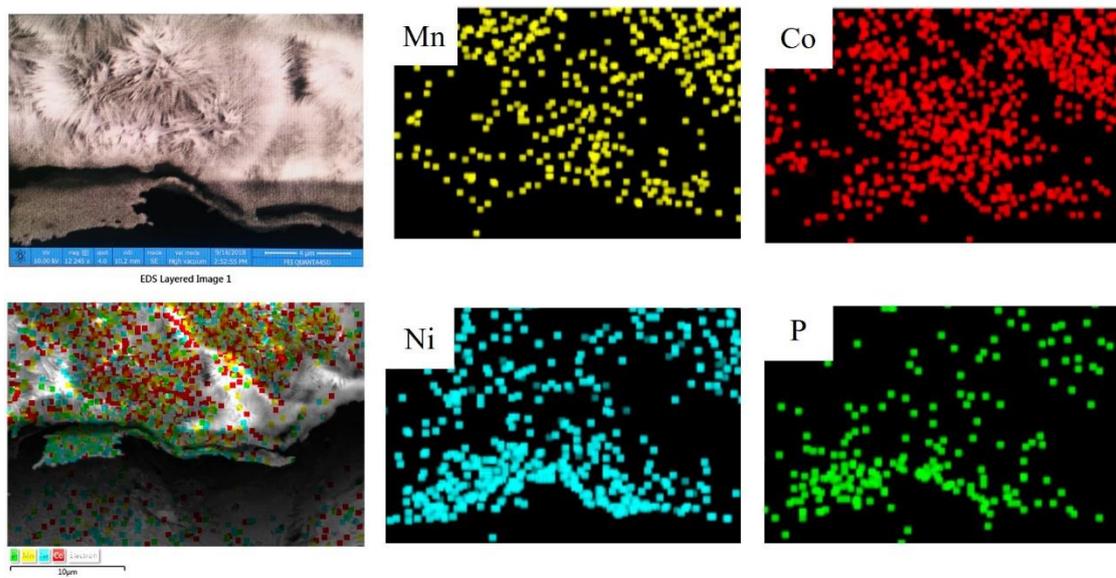
**Fig S7.** The SEM image of Ni<sub>2</sub>P foam after acid treatment.



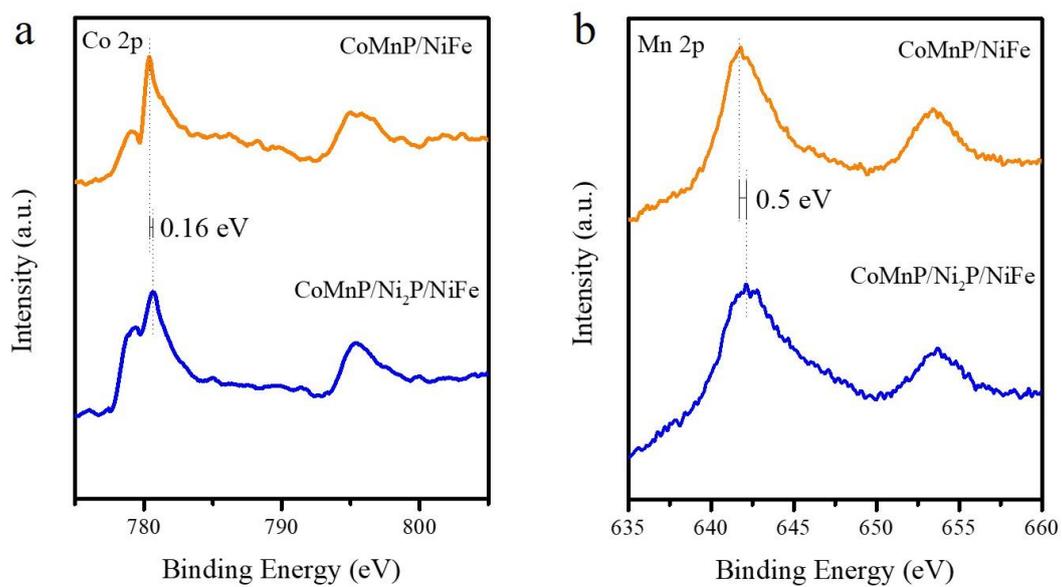
**Fig S8.** The SEM images of a) CoMnP/Ni<sub>2</sub>P/NiFe, b) zoom-in orange region and zoom-in red region.



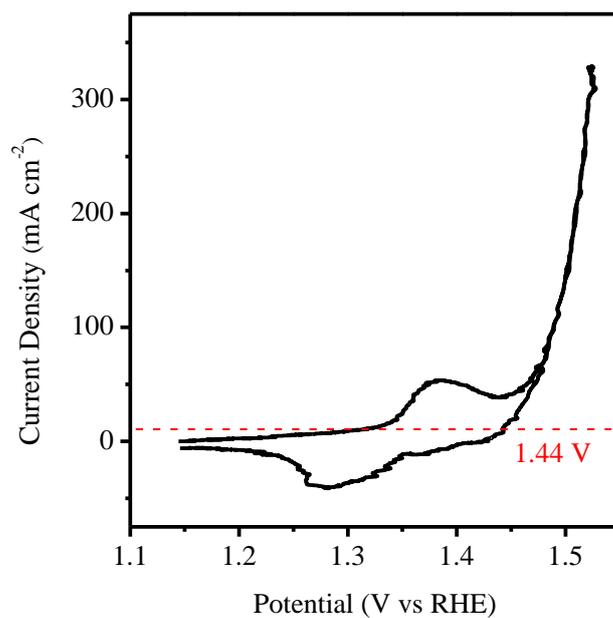
**Fig S9.** The SEM images of a) and d) CoMnCH/Ni<sub>2</sub>P on the different sites of NiFe foam. b) zoom-in orange region and c) zoom-in red region. e) and f) are the enlarged images of d).



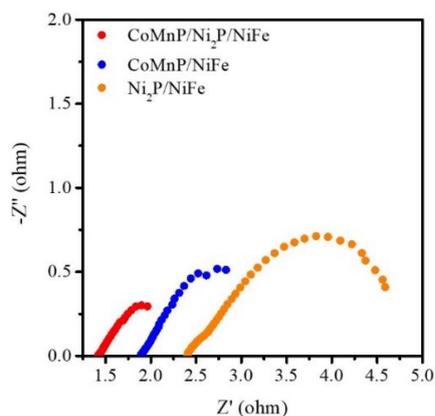
**Fig S10.** The SEM images of CoMnCH/Ni<sub>2</sub>P/NiFe and corresponding elemental mappings.



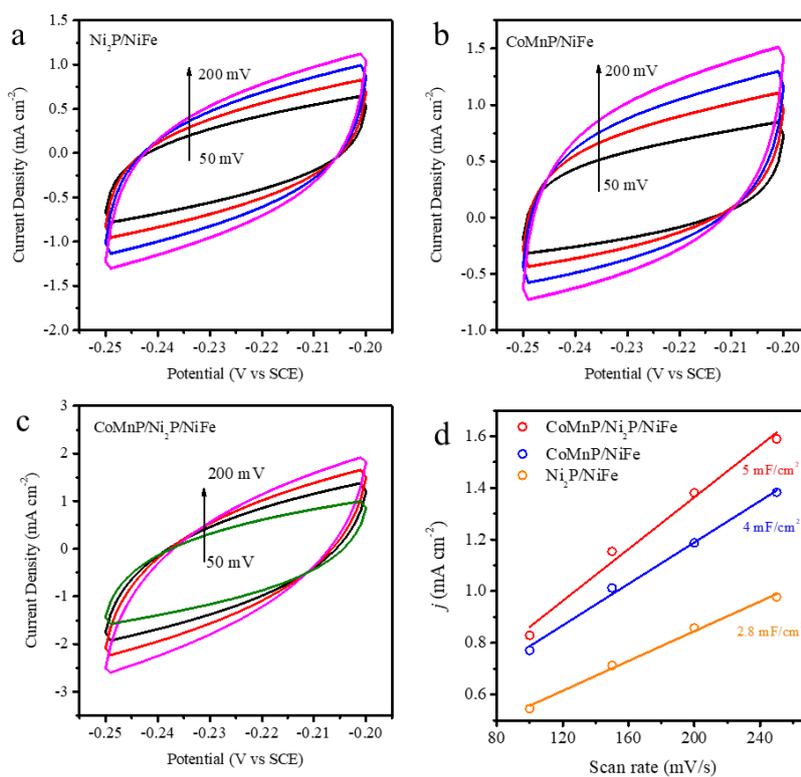
**Fig S11.** XPS characterization of CoMnCH/Ni<sub>2</sub>P/NiFe and CoMnP/Ni<sub>2</sub>P/NiFe (a) Co 2p; (b) Mn 2p.



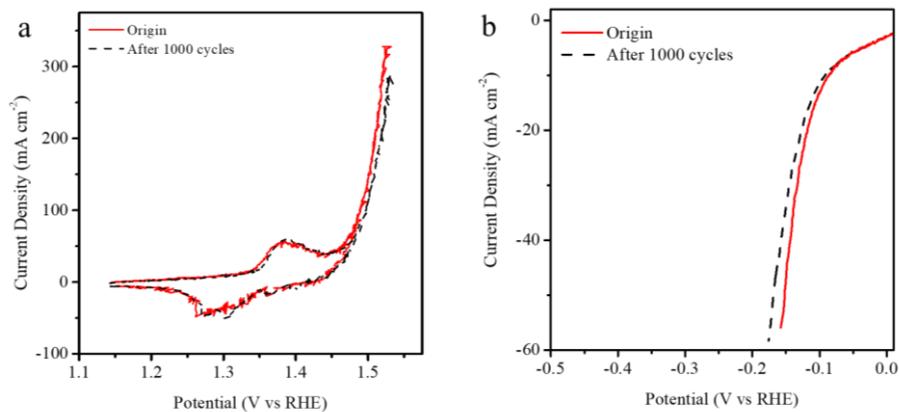
**Fig S12.** The CV curve of CoMnP/Ni<sub>2</sub>P/NiFe. The scan rate is 5 mV/s.



**Fig S13.** EIS curves of Ni<sub>2</sub>P, CoMnP and Ni<sub>2</sub>P/CoMnP on NiFe foam at a potential of 1.5 V versus SCE.

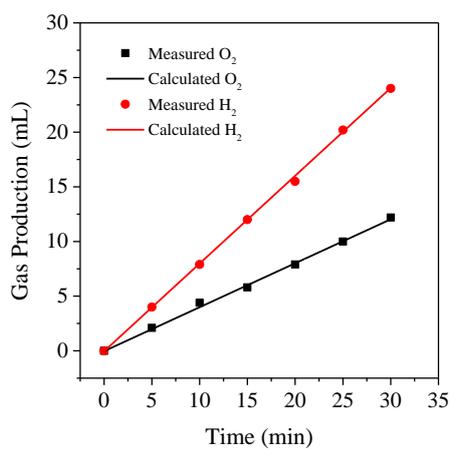


**Fig S14.** The  $C_{dl}$  by plotting the current density against scan rate to fit a linear regression measured at the voltage from -0.25 V to -0.20 V (vs SCE).

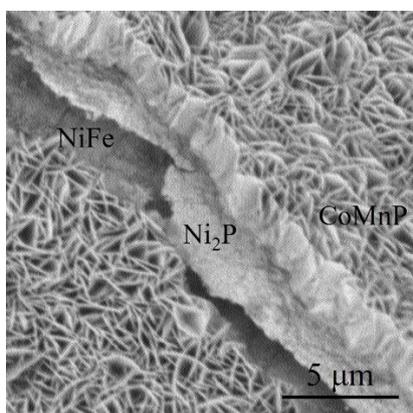


**Fig S15.** The a) OER and b) HER stability evolution of CoMnP/Ni<sub>2</sub>P/NiFe at 1 M

KOH solution.



**Fig S16.** Measured and calculated volumes of the evolved H<sub>2</sub> and O<sub>2</sub> over the CoMnP/Ni<sub>2</sub>P/NiFe electrodes at a constant voltage of 1.7 V in 1 M KOH solution.



**Fig S17.** SEM image of the CoMnP/Ni<sub>2</sub>P/NiFe structure after the reaction of 35 hours.

Table 1. Comparison of different bifunctional electrocatalysts for overall water splitting reported in the literature.

Catalyst	Substrate	Overpotential (10 mA/cm <sup>2</sup> )	Reference
<b>CoMnP/Ni<sub>2</sub>P</b>	<b>NiFe foam</b>	<b>1.48 V</b>	<b>This work</b>
FeS	Fe foam	1.65 V	1
Ni/NiMnFe/CeO <sub>2</sub>	Carbon fiber	1.58 V	2
NiFe/NiCo <sub>2</sub> O <sub>4</sub>	Ni foam	1.67 V	3
NiFeOF	Ni foam	1.80 V	4
CoP	Ni foam	1.62 V	5
Ni-Pi	Carbon fiber	1.63 V	6

#### Reference

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