## **Supporting Information**

CoMo-LDH ultrathin nanosheets as a highly active and bifunctional

electrocatalyst for overall water splitting

Jian Bao<sup>1,\*</sup>, Zhaolong Wang<sup>1</sup>, Junfeng Xie<sup>2</sup>, Li Xu<sup>1</sup>, Fengcai Lei<sup>2</sup>, Meili Guan<sup>1</sup>, Yunpeng Huang<sup>1</sup>, Yan Zhao<sup>1</sup>, Jiexiang Xia<sup>1</sup>, Huaming Li<sup>1,\*</sup>

1. Institute for Energy Research, School of Chemistry and Chemical Engineering, Jiangsu University, Zhenjiang, Jiangsu, 212013, P. R. China.

2. College of Chemistry, Chemical Engineering and Materials Science, Key Laboratory of Molecular and Nano Probes (Ministry of Education), Collaborative Innovation Center of Functionalized Probes for Chemical Imaging in Universities of Shandong, Institute of Molecular and Nano Science, Shandong Normal University, Jinan, Shandong, 250014, P. R. China.

Correspondence: baojian@ujs.edu.cn; lhm@ujs.edu.cn



Figure S1. XRD patterns of  $Co(OH)_2$  and CoMn-LDH samples.



Figure S2. XPS survey spectrum of CoMo-LDH.



Figure S3. (A) tapping mode AFM image of the CoMo-LDH (B) the corresponding height profile of the CoMo-LDH.









Table S1. ICP results of the CoMo-LDH



Figure S5. The CDL plots of the CoMo-LDH, CoMn-LDH and Co(OH)<sub>2</sub> samples.



Figure S6. The LSV curves of nickel foam (NF) for the (A) HER, (B) OER and (C) overall water splitting.



Figure S7. (A) Cyclic voltammetry (CV) curves for the CoMo-LDH deposited on ITO substrate in the OER process. Inset: Optical images of the CoMo-LDH prior to and after CV cycles. (B) The Co 2p XPS spectra for the CoMo LDH before and after the OER test.

In order to further investigate the role of the  $Mo^{6+}$ , the first three CV curves were tested and shown in Figure S7A. After the 1st cycle, the peak assigned to the oxidation of  $Co^{2+}$  to  $Co^{3+}$  reduced indicating that the catalyst becomes irreversibly oxidized to a higher oxidation state of  $Co<sup>3+</sup>$ . Furthermore, XPS data for CoMo-LDH before and after electrocatalysis were collected. As illustrated in Figure S7B, the XPS spectrum shows an obvious increase in intensity for the  $Co<sup>3+</sup>$  and the decrease for the  $Co<sup>2+</sup>$  after OER, thus confirming the oxidation of  $Co<sup>2+</sup>$  to  $Co<sup>3+</sup>$ . Benefiting from the prominent ability to draw electrons of high-valence  $Mo^{6+}$ , the Co would easily stay as their high-valence state, which is widely regarded as active sites for OER.

Material	Overpotential (mV)	Tafel slope	Ref.
	/10 mA $cm^{-2}$	mV / decade	
CoMo LDH	300	56	This work
NiCo LDH	330	42	
NiFe LDH	300	40	
<b>NiCoP</b>	300	80	$\overline{2}$
CoVOx	347	49	3
CoFe LDH	340	85	5
NiV LDH	310	50	4
NiCo <sub>2</sub> O <sub>4</sub>	320	32	5
CoMoO <sub>4</sub>	312	56	6
NiFe LDH/CR	256	50	7
NiFe LDH/ CF	260	55	8

Table S2. Comparision of the OER performance with various reported materials loaded on the glassy carbon.

Table S3. Comparision of the overall water splitting performance with various reported materials loaded on the nickel foam.



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