Supplementary Information

Cobalt Phosphide Supported by Two-Dimensional Molybdenum Carbide (MXene) for Hydrogen Evolution Reaction, Oxygen Evolution Reaction, and Overall Water Splitting

Shilong Liu,[a] Zongshan Lin,[a] Rendian Wan,[a] Yonggang Liu,[b] Zhe Liu,[b] Shuidong Zhang,[c] Xiaofeng Zhang,[d] Zhenghua Tang,[,b,c] Xiaoxing Lu,[*,a] and Yong Tian[*,a]*

[a.] School of Pharmacy, Guangdong Pharmaceutical University, Guangzhou, 510006, P. R. China. Emails: xxlu@gdpu.edu.cn, tian_yong_tian@163.com

[b.] Guangzhou Key Laboratory for Surface Chemistry of Energy Materials, New Energy Research Institute, School of Environment and Energy, South China University of Technology, Guangzhou Higher Education Mega Center, Guangzhou, Guangdong 510006, China. Email: zhht@scut.edu.cn

[c.] Guangdong Provincial Key Laboratory of Technique and Equipment for Macromolecular Advanced Manufacturing, South China University of Technology, China.

[d.] Institute of New Materials, Guangdong Academy of Science, Guangzhou, 510650, China.

List of Figures

Figure S19. SEM image of CoP/Mo_2CT_x (Post-OER sample) and corresponding elemental mapping, EDX pattern with elemental composition of CoP/Mo_2CT_x.

List of Tables

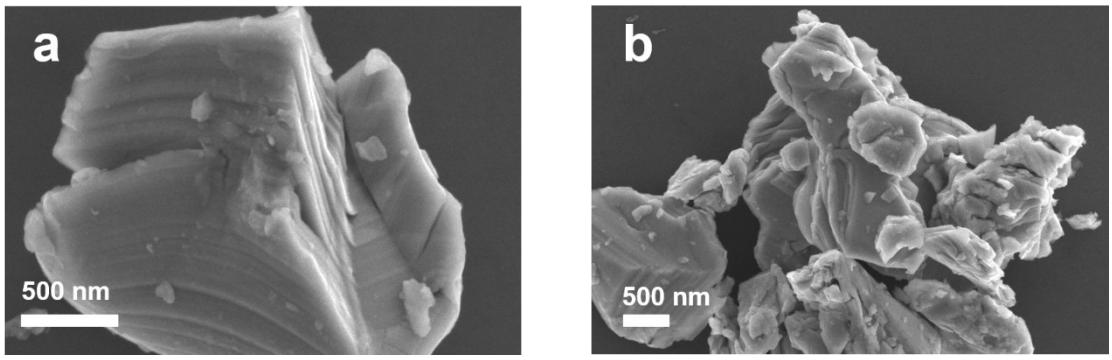


Figure S1. SEM image of the MAX precursor.

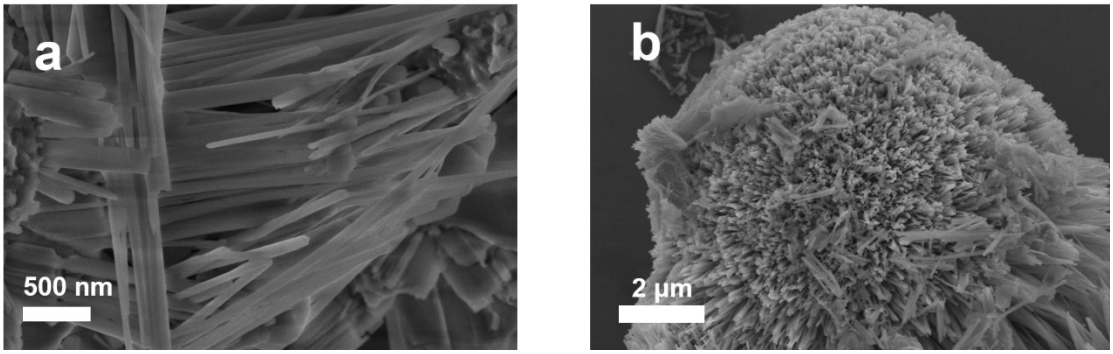


Figure S2. SEM image of the Co(OH)F (a) and Co(OH)F/Mo_2CT_x (b) precursor.

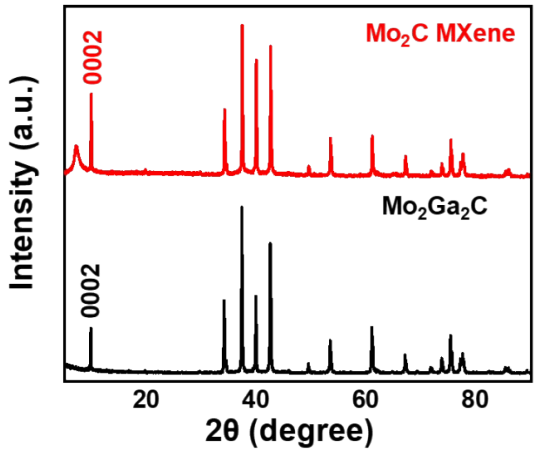


Figure S3. XRD pattern of the Mo_2Ga_2C phase and the Mo_2C MXene phase.

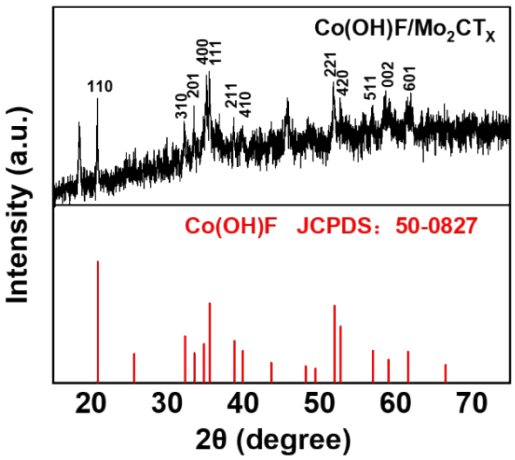


Figure S4. XRD pattern of Co(OH)F/Mo_2CT_x.

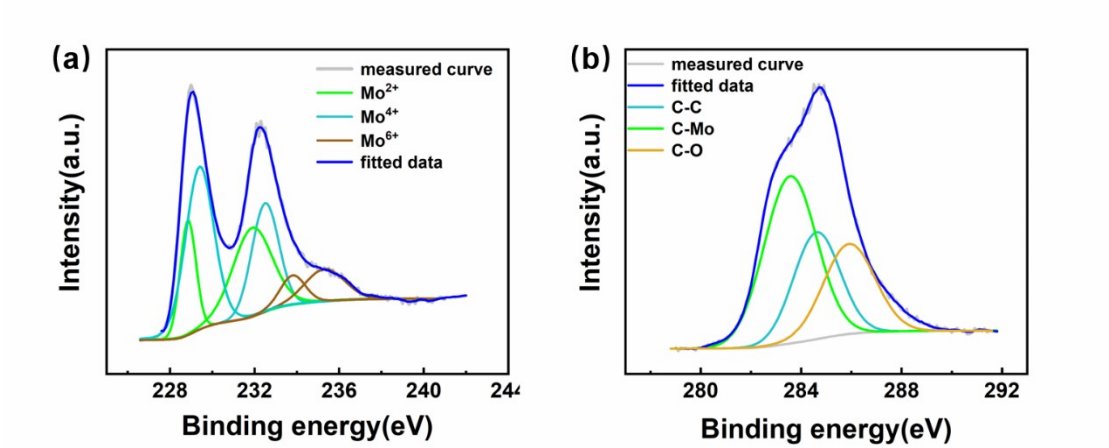


Figure S5. High resolution XPS spectra of the Mo 3d (a) and C 1s (b) electrons for CoP/Mo_2CT_x.

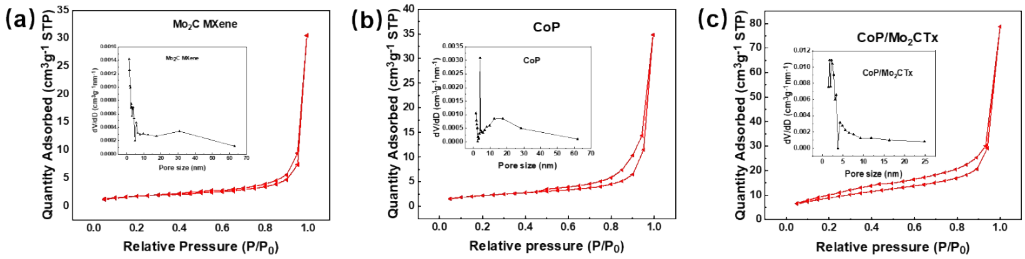


Figure S6. N_2 adsorption-desorption isotherm of various catalysts.

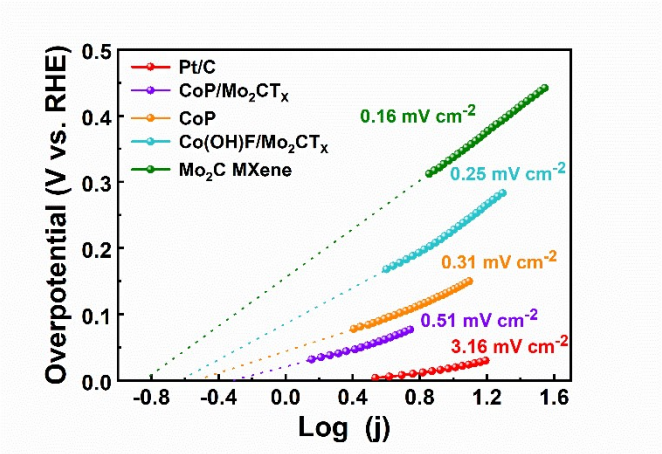


Figure S7. Exchange current densities of the electrocatalysts.

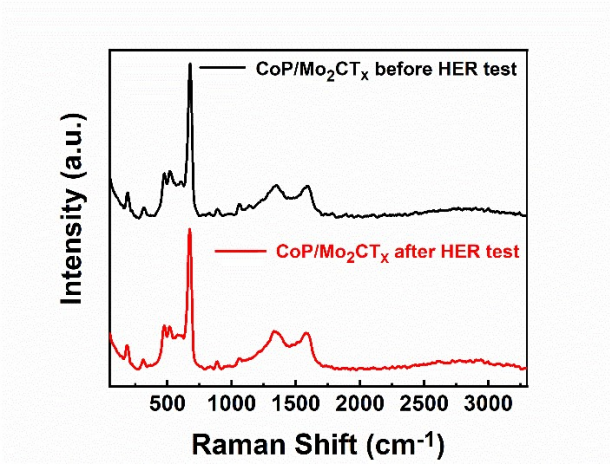


Figure S8. Raman spectra of CoP/Mo_2CT_x before and after HER test.

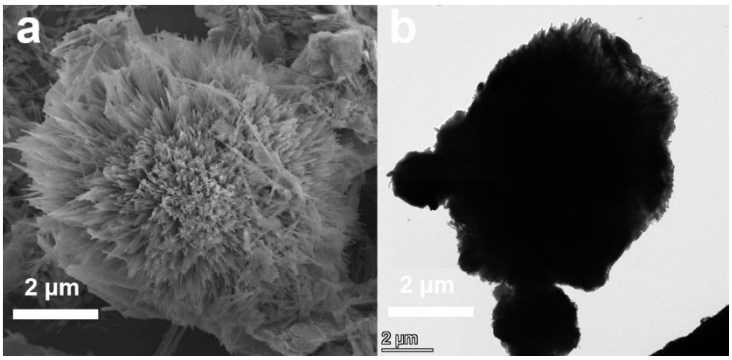


Figure S9. (a) SEM and (b) TEM image of CoP/Mo_2CT_x after the stability test of HER.

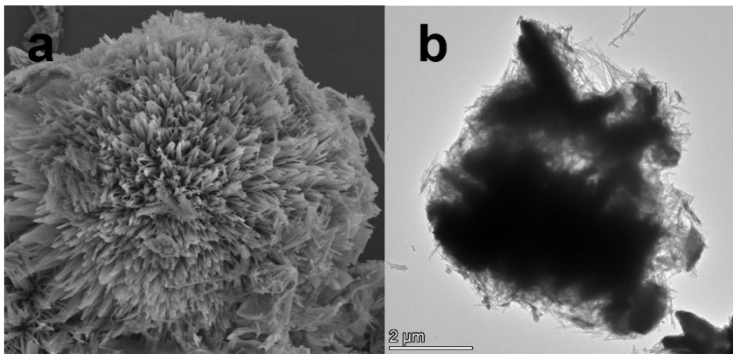


Figure S10. (a) SEM and (b) TEM image of CoP/Mo_2CT_x after the stability test of OER.

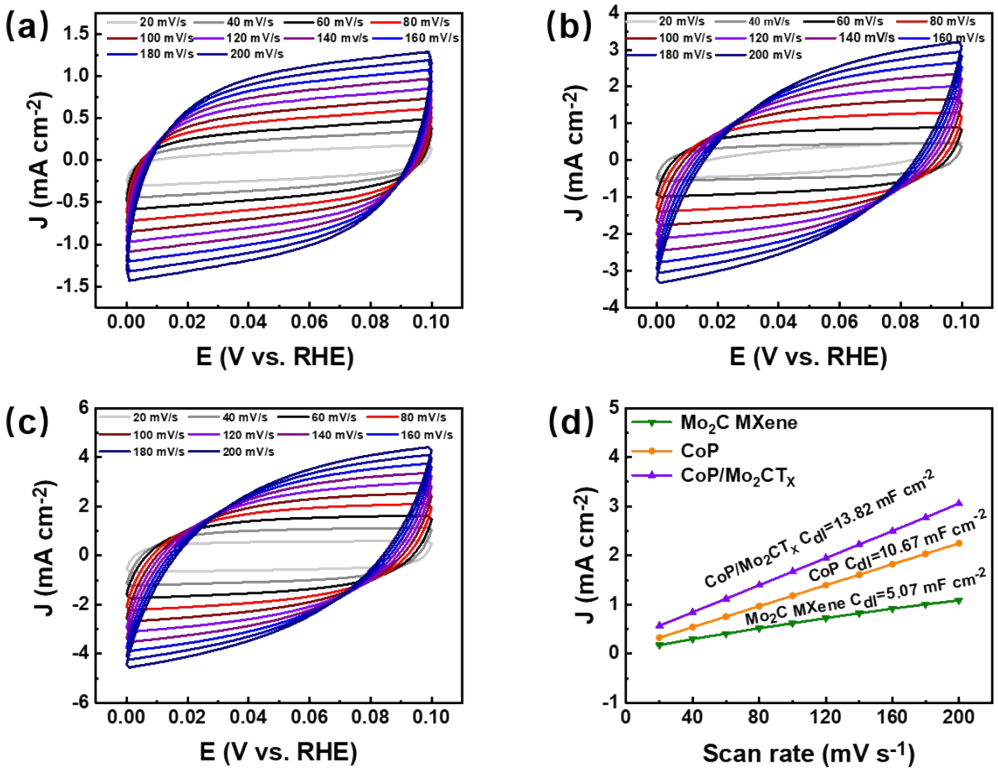


Figure S11. Cyclic voltammograms of (a) Mo_2C MXene, (b) CoP, and (c) CoP/Mo_2CT_x in the region of 0-0.1 V in 1.0 M KOH at various scan rates and (d) the plots of current density as a function of scan rate derived from (a-c), respectively.

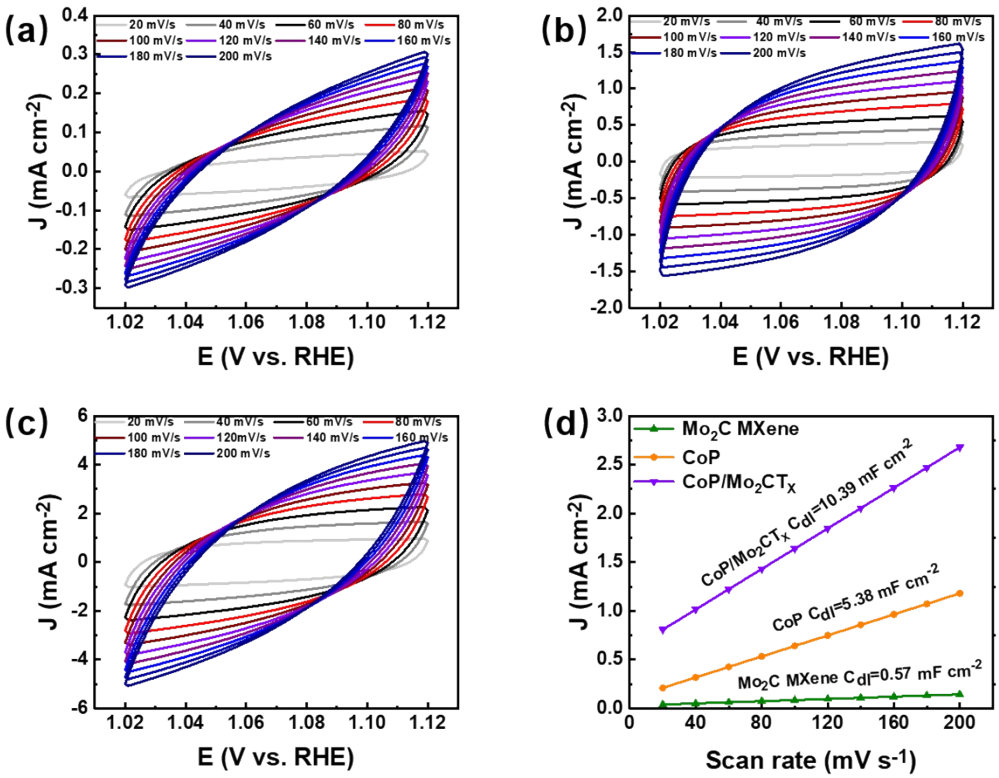


Figure S12. Cyclic voltammograms of (a) Mo_2C MXene, (b) CoP, and (c) CoP/Mo_2CT_x in the region of 1.02-1.12 V in 1.0 M KOH at various scan rates and (d) the plots of current density as a function of scan rate derived from (a-c), respectively.

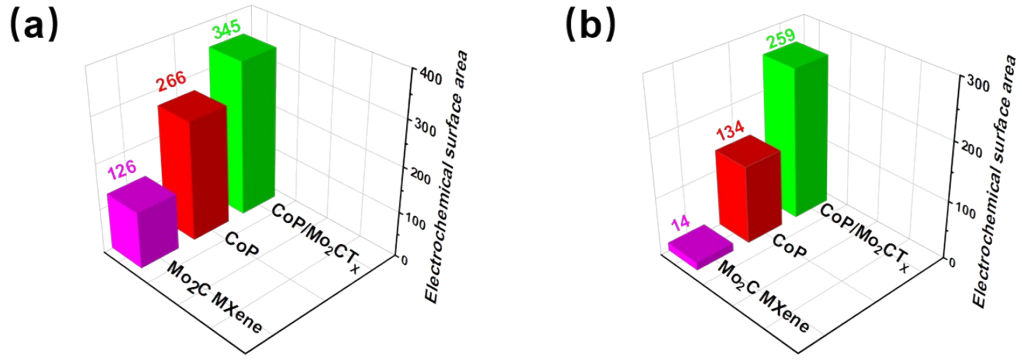


Figure S13. The electrochemically active surface area of different catalysts in HER (a) and OER (b) test.

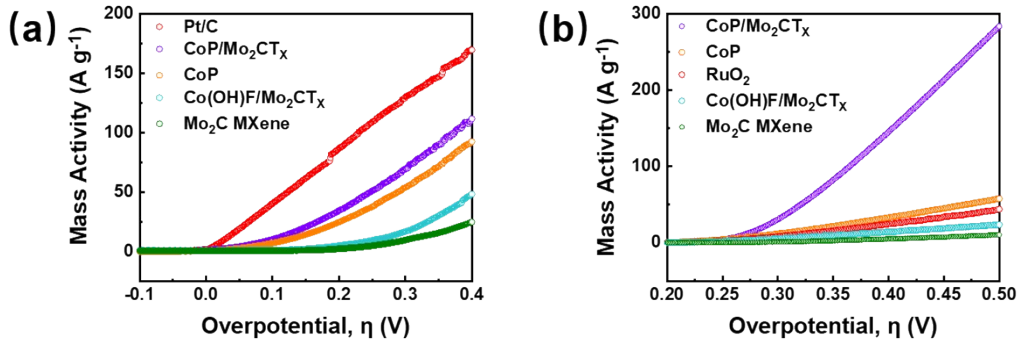


Figure S14. Mass activity for various catalysts in HER (a) and OER (b) test.

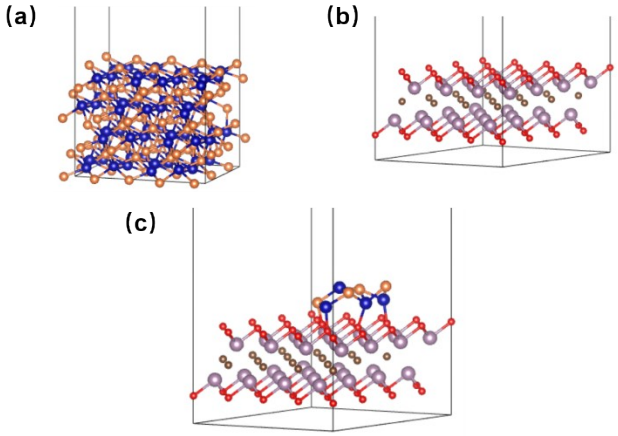


Figure S15. The calculation models of CoP (a), Mo_2C MXene (b), and CoP/Mo_2CT_x (c).

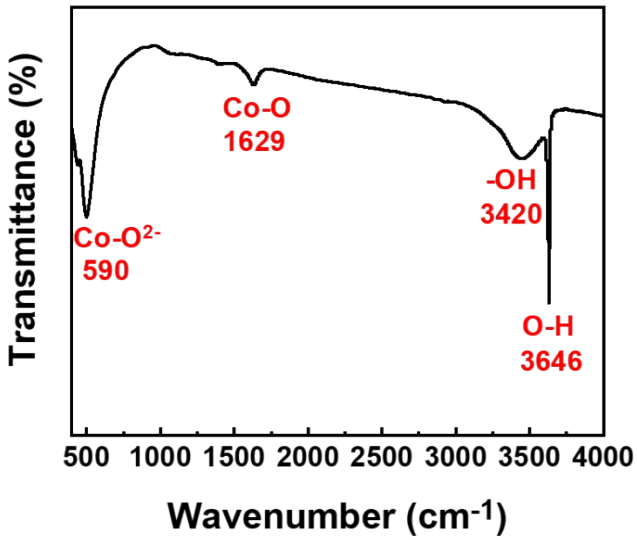


Figure S16. FTIR spectrum of CoP/Mo_2CT_x after OER test.

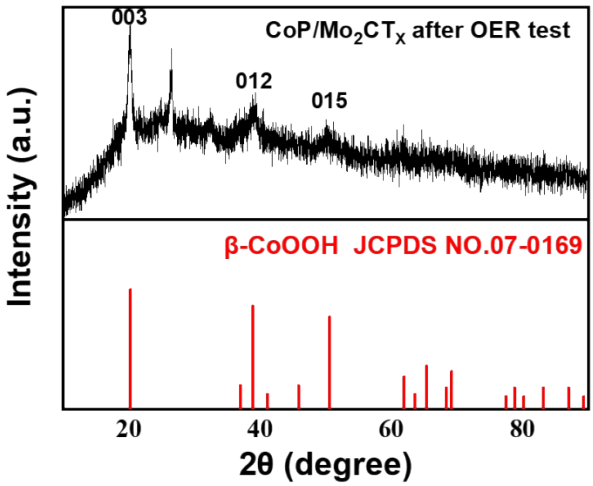
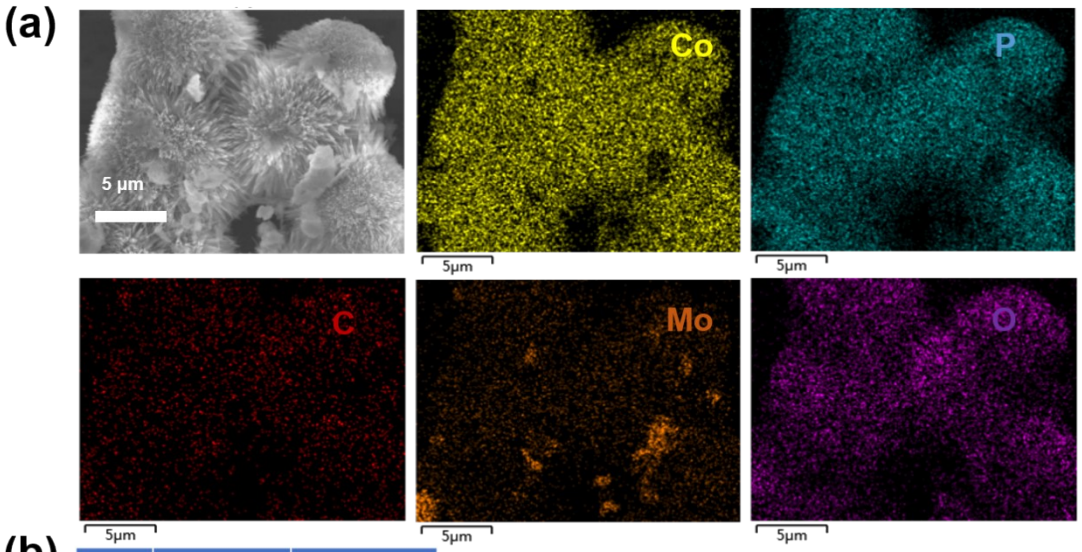


Figure S17. XRD patterns of CoP/Mo_2CT_x after OER test.



(b)

	wt%	at%
C	13.47	30.11
O	17.45	29.26
P	25.02	21.65
Co	37.84	17.20
Mo	6.21	1.77

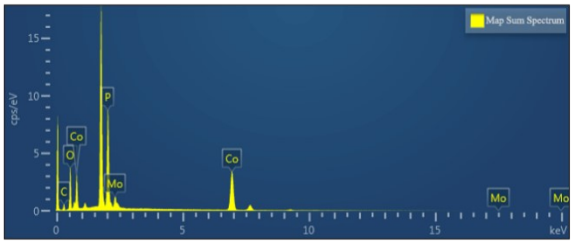
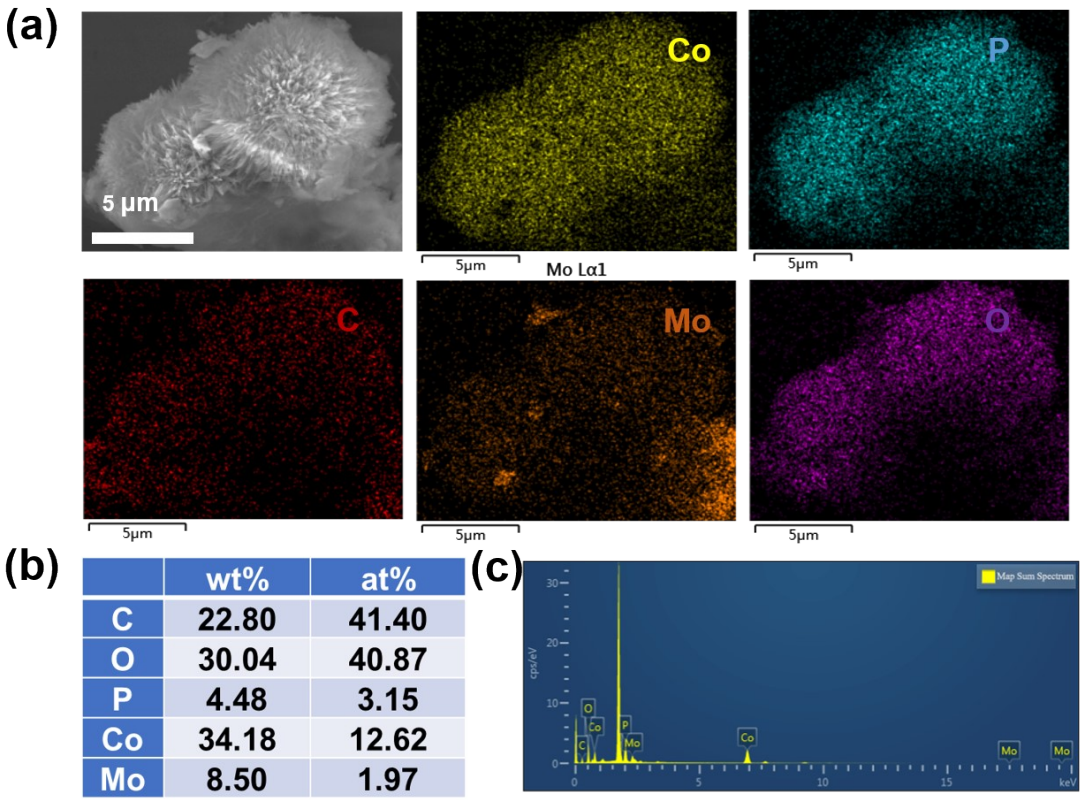


Figure S18. SEM image of CoP/Mo_2CT_x (fresh sample) and corresponding elemental mapping, EDX pattern with elemental composition of CoP/Mo_2CT_x.



	wt%	at%
C	22.80	41.40
O	30.04	40.87
P	4.48	3.15
Co	34.18	12.62
Mo	8.50	1.97

Figure S19. SEM image of CoP/Mo_2CT_x (Post-OER sample) and corresponding elemental mapping, EDX pattern with elemental composition of CoP/Mo_2CT_x.

Table S1. The content of Co, P and Mo elements percentage for all samples, measured by inductively coupled plasma- optical emission spectrometry (ICP-OES).

Catalyst	Co (wt. %)	P (wt. %)	Mo (wt. %)
CoP/Mo_2CT_x	31.34	25.23	17.52
After HER	34.18	18.45	16.04
After OER	38.64	3.62	16.55

Table S2. Comparison of HER performance for non-precious metal electrocatalysts in 1.0 M KOH.

Electrocatalysts	η@10mA cm^{-2} (mV)	Tafel Slope (mV dec^{-1})	Reference
CoP/Mo_2CT_x	**78**	**66**	**This work**
CoP/$Ti_3C_2T_x$	116	57	*J. Mater. Chem. A*, **2019**, *7*, 27383–27393
CoP@NC	129	58	*ACS Catal.***2017**, *7*, 3824.
$Ni_{0.9}Fe_{0.1}PS_3$@MXene	196	NA	*Adv. Energy Mater.* **2018**, *8*, 1801127
CoP/CC	209	129	*J. Am. Chem. Soc.* **2014**, *136*, 7587
CoP/NCNHP	115	66	*J. Am. Chem. Soc.* **2018**, *140*, 2610
NC-CNT/CoP	120	73	*J. Mater. Chem. A* **2018**, *6*, 9009
Co-P film/Cu	94	42	*Angew. Chem., Int. Ed.* **2015**, *54*, 625.
Co/N-doped carbon	260	91.2	*ACS Nano* **2016**, *10*, 684.
CoP NS/C	110	70.9	*Green Chem*. **2016**, *18*, 2287.
Co-MoS_2/Mo_2CT_x	112	82	*Nanoscale* **2019**, *11*, 10992
CoP NFs	136	56.2	*ACS Catal.* **2020**, *10*, 412
CoP@NF	155	96	*Nano Energy* **2020**, *67*, 104174
CoP/CC	87	72	*Appl. Catal. B Environ.* **2020**, *253*, 21
CoP/PC	76	NA	*Small* **2020**, *16*, 1900550
CoP film	94	42	*Angew. Chem., Int. Ed.* **2015**, *54*, 6251

Table S3. Fitting parameters obtained from the EIS data for the HER in 1 M KOH.

Catalysts	R_1(Ω)	R_2(Ω)	CPE_1-T(F)	CPE_1-P	R_3(Ω)	CPE_2-T(F)	CPE_2-P
CoP/Mo_2CT_x	2.567	1.347	0.11238	0.39366	3.373	0.24052	0.77702
CoP	2.434	2.466	0.20459	0.28379	3.479	0.24449	0.80125
Co(OH)F/Mo_2CT_x	2.486	1.271	0.39589	0.20214	3.841	0.23873	0.82258
Mo_2C MXene	3.509	5.737	0.13236	0.47809	4.666	0.07312	0.81123

R_1: electrolyte resistance.

R_2: charge-transfer resistance.

R_3: solid-electrolyte interface resistance.

CEP_1: capacitance generated from the Faradic process, and constant-phase element.

CEP_2: capacitance arisen from the solid-electrolyte interface process.

Table S4. Comparison of OER performance for non-precious metal electrocatalysts in 1.0 M KOH.

Electrocatalysts	η@10mA cm^{-2} (mV)	Tafel Slope (mV dec^{-1})	Reference
CoP/Mo_2CT_x	**260**	**51**	**This work**
CoP-MNA/NF	290	65	*Adv. Funct. Mater*. **2015**, *25*, 7337
Co-P film	345	47	*Angew. Chem., Int. Ed*. **2015**, *54*, 6251
CoP NR/C	320	71	*ACS Catal.* **2015**, *5*, 6874
CoP/rGO	340	66	*Chem. Sci.* **2016**, *7*, 1690
CoP/NCNHP	310	70	*J. Am. Chem. Soc.* **2018**, *140*, 2610
Ni0.7Fe0.3PS3@MXene	282	36.5	*Adv. Energy Mater.* **2018**, *8*, 1801127
CoP@NPMG	276	54	*Nanoscale* **2018**, *10*, 2603
CoP@PC-750	283	53	*Small* **2019**, *15*, 1900550
CoP/CoO	295	73	*Nano Energy* **2019**, *56*, 109
CoP/TiOx	337	72.1	*Small* **2020**, *16*, 1905075
Co2P NRs	372	111.8	*J. Am. Chem. Soc.* **2020**, *142*, 8490
CoP/CC	340	87	*Adv. Funct. Mater.* **2020**, *30*, 1909618
CoP NFs	323	49.6	*ACS Catal.* **2020**, *10*, 412
CoP/Graphene	340	66	*Chem. Sci.* **2016**, *7*, 1690.
CoP/CNT	330	40	*ACS Appl. Mater. Interfaces* **2015**, *7*, 28412.

Table S5. Fitting parameters obtained from the EIS data for the OER in 1 M KOH.

Catalysts	R_1(Ω)	R_2(Ω)	CPE_1-T(F)	CPE1-P	R_3(Ω)	CPE2-T(F)	CPE2-P
CoP/Mo_2CT_x	2.674	1.091	0.01291	0.53243	3.389	0.046454	0.70913
CoP	2.475	1.888	0.16482	0.33264	3.452	0.25667	0.81997
Co(OH)F/Mo_2CT_x	2.494	1.696	0.12766	0.36013	3.514	0.26314	0.82463
Mo_2C MXene	2.403	1.873	0.24933	0.19699	3.815	0.05575	0.74074

R_1: electrolyte resistance.

R_2: charge-transfer resistance.

R_3: solid-electrolyte interface resistance.

CEP_1: capacitance generated from the Faradic process, and constant-phase element.

CEP_2: capacitance arisen from the solid-electrolyte interface process.

Table S6. Comparison of the water splitting cell voltage of CoP/Mo_2CT_x with recently reported bifunctional electrocatalysts in alkaline medium.

Electrocatalyst	Potential (V) at 10 mA cm^{-2}	Reference
CoP/Mo_2CT_x	**1.56**	**This work**
NiCoP/rGO	1.59	*Adv. Funct. Mater.* **2016**, *26*, 6785.
Co-P film	1.65	*Angew. Chem. Int. Ed.* **2015**, *54*, 6251
CoP-MNA	1.62	*Adv. Funct. Mater.* **2015**, *25*, 7337
CoP NR	1.587	*ACS Catal.* **2015**, *5*, 6874
NiCoP/Ti	1.64	*Adv. Mater. Interfaces* **2016**, *3*, 1500454
CoP nanosheets	1.54	*Green Chem.* **2016**, *18*, 2287
CoP/GO	1.7	*Chem. Sci.* **2016**, *7*, 1690
CoP/CC	1.61	*ChemSusChem* **2016**, *9*, 472
Fe-CoP/Ti	1.60	*Adv. Mater*. **2017**, *29*, 1602441
CoP NA/CC	1.65	*ChemElectroChem* **2017**, *4*, 1840
Ni-Co-P HNBs	1.62	*Energy Environ. Sci*. **2018**, *11*, 872
S:CoP@NF	1.617	*Nano Energy* **2018**, *53*, 286
CoP-400	1.65	*Adv. Energy Mater*. **2018**, *8*, 1802445
CoP@a-CoOx	1.66	*Adv. Sci*. **2018**, *5*, 1800514
CoP/NCNHP	1.64	*J. Am. Chem. Soc*. **2018**, *140*, 2610