Electronic Supplementary Material (ESI) for Nanoscale.

Electronic Supplementary Information

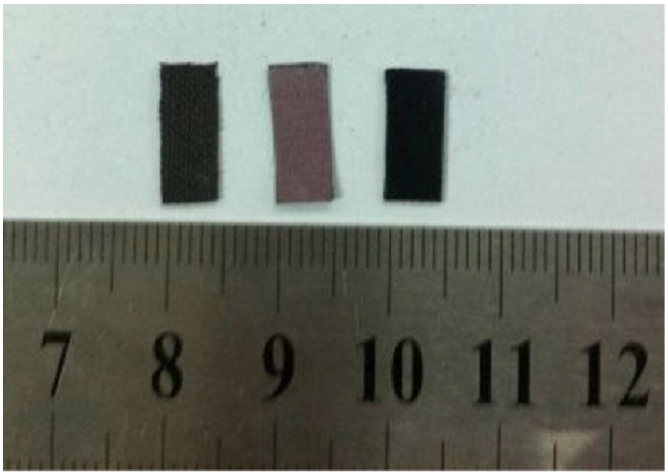

Fig. S1. Optical photograph for CC, CoAl-LDH/CC, and Al-CoP/CC (from left to right).

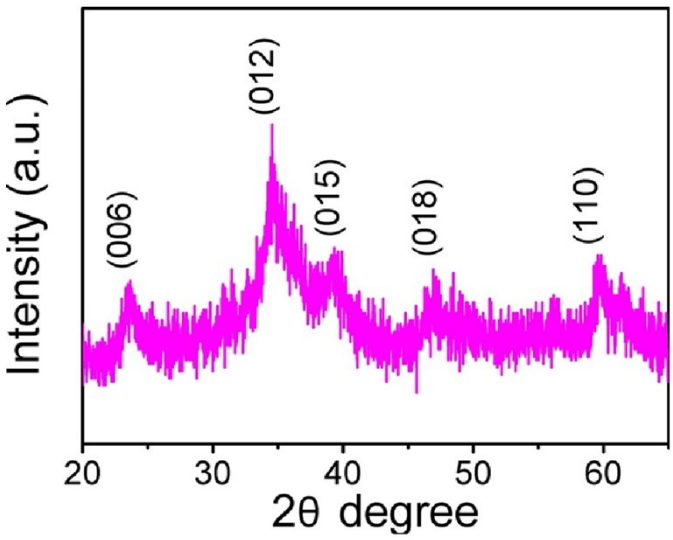

Fig. S2. XRD pattern of CoAl-LDH scratched down from CC.

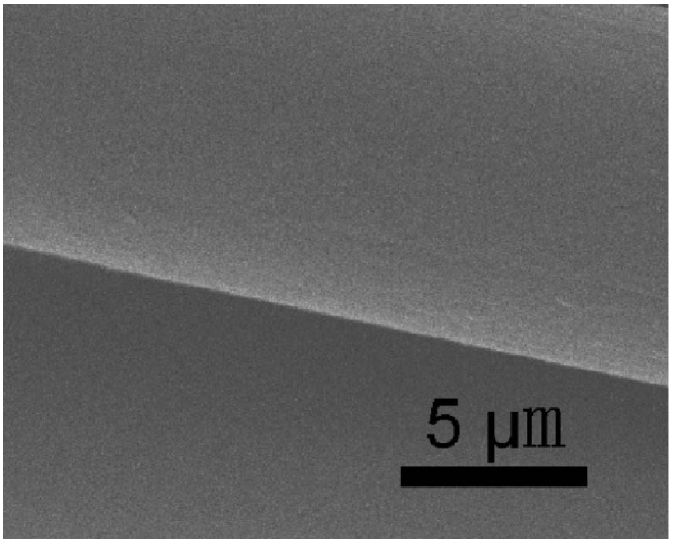

Fig. S3. SEM image for bare CC.

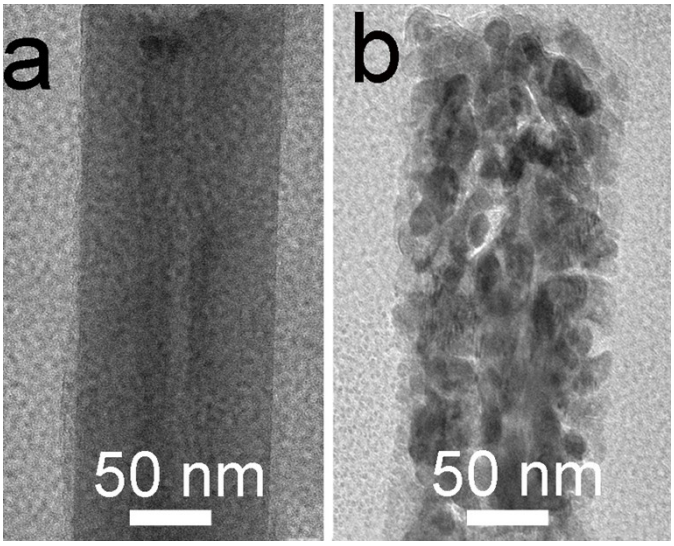

Fig. S4. TEM images for (a) AlCo-LDH and (b) Al-CoP.

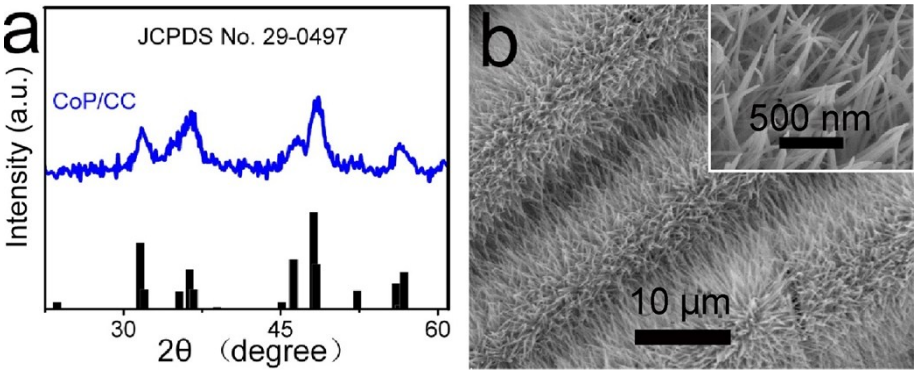

Fig. S5. (a) XRD pattern for CoP scratched down from CC. (b) SEM images for CoP/CC.

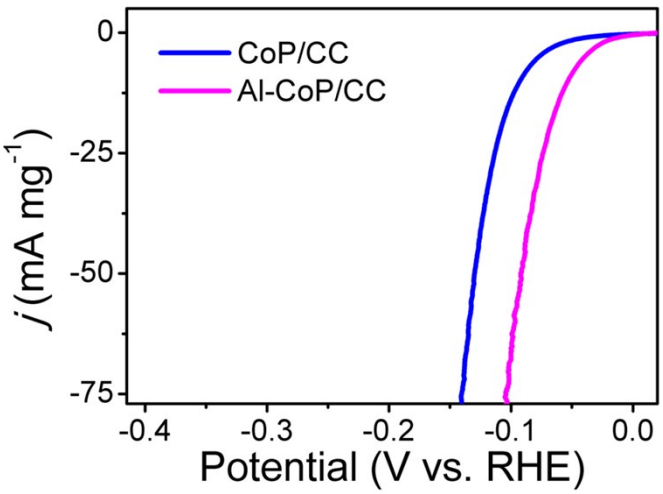

Fig. S6. Mass-normalized polarization curves for Al-CoP/CC and CoP/CC in 0.5 M H_2SO_4.

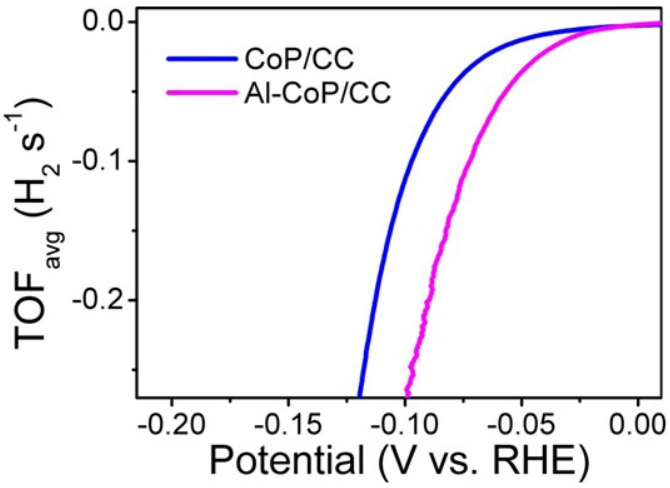

Fig. S7. TOF_{avg} calculation of Al-CoP/CC and CoP/CC.

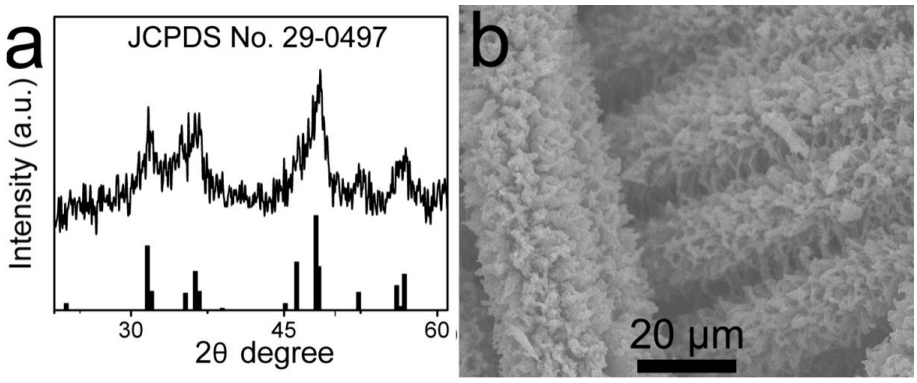

Fig. S8. (a) XRD pattern for Al-CoP and (b) SEM image for Al-CoP/CC after HER hydrolysis.

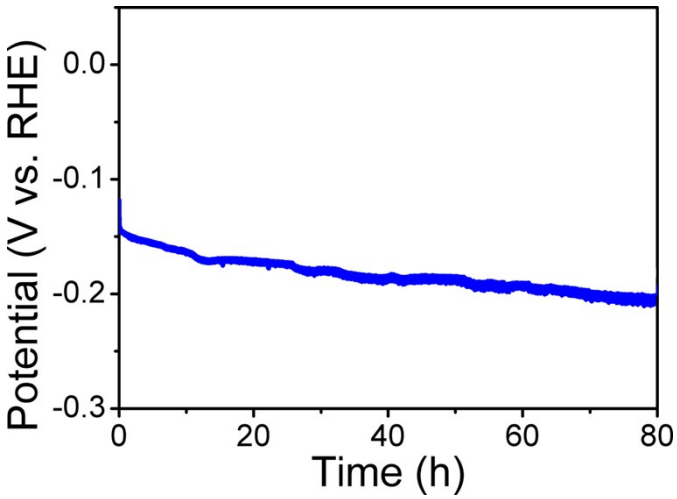

Fig. S9. Chronopotentiometric curve of CoP/CC with constant current density of 50 mA cm^{-2} for 80 h (without iR correction).

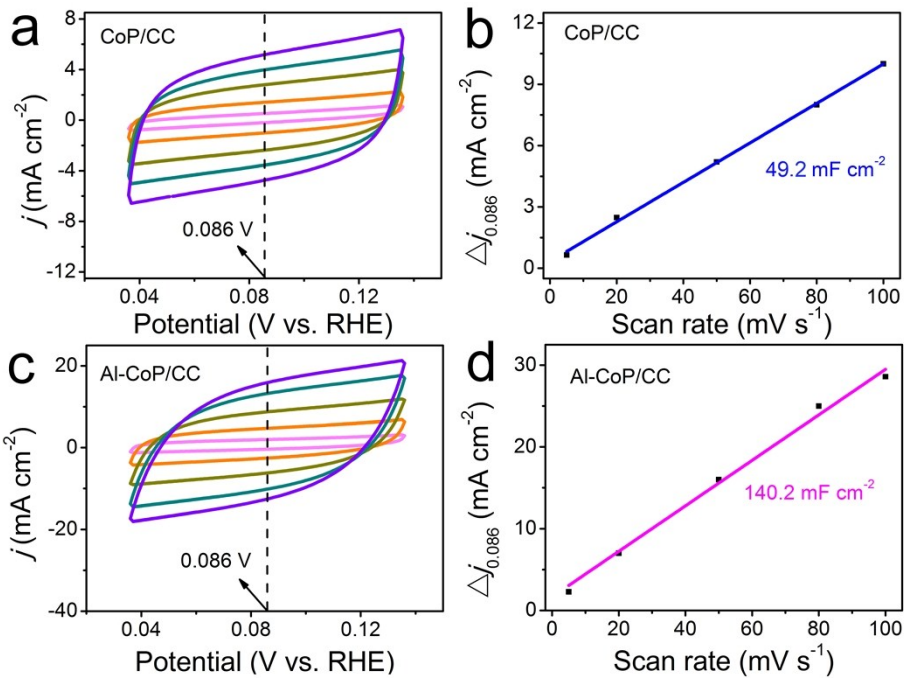

Fig. S10. CVs of (a) CoP/CC and (c) Al-CoP/CC after electrolysis with various scan rates (5–100 mV s^{-1}) in the region of 0.036 to 0.136 V. The capacitive current densities at 0.086 V as a function of scan rate for (b) CoP/CC and (d) Al-CoP/CC. All experiments were performed in 0.5 M H_2SO_4.

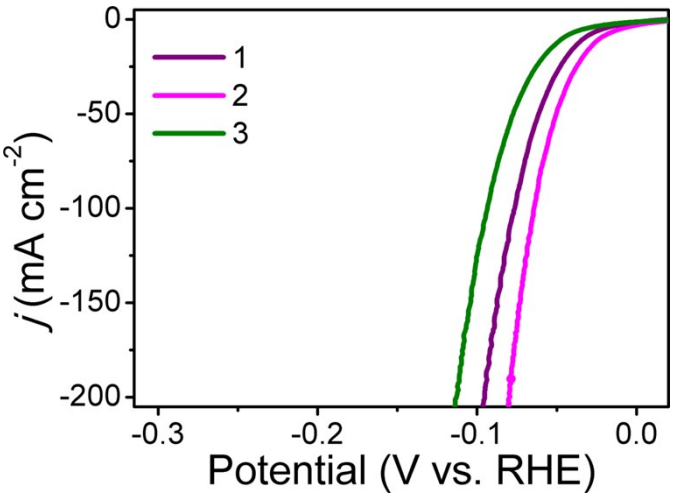

Fig. S11. Polarization curves for HER of Al-doped CoP nanoarray on CC with different Al doping degree: 4% (curve 1), 10 % (curve 2), and 16 % (curve 3). All experiments were performed in 0.5 M H_2SO_4 with a scan rate of 5 mV s^{-1}.

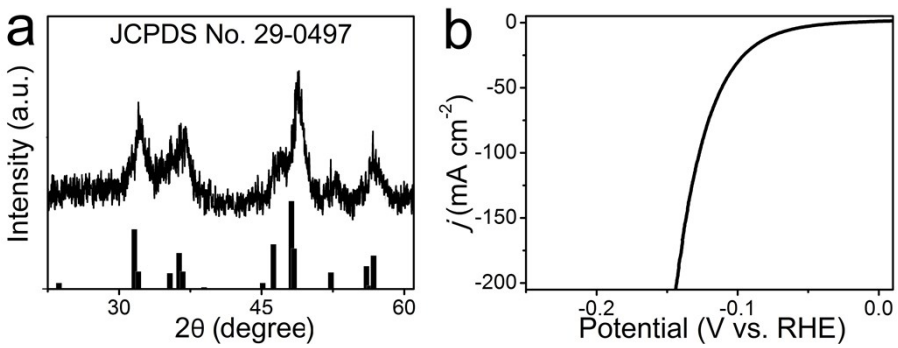

Fig. S12. (a) XRD pattern and (b) polarization curve for HER of Al-doped CoP nanoarray on CC with Al doping degree: 35%.

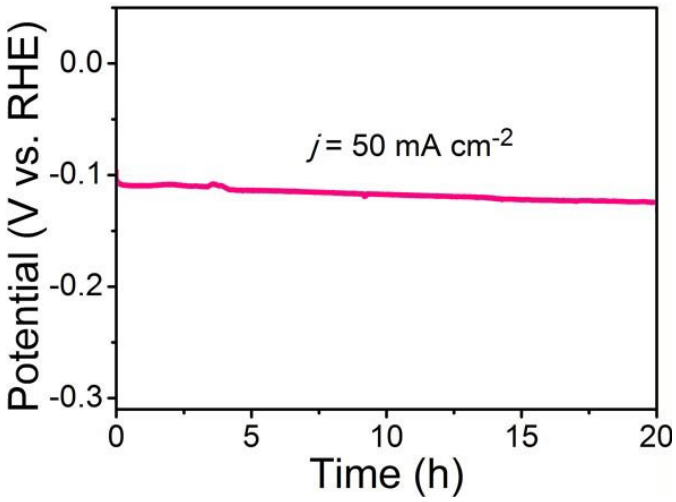

Fig. S13. Chronopotentiometric curve of Al-CoP/CC with constant current density of 50 mA cm^{-2} for HER in 1.0 M KOH (without iR correction).

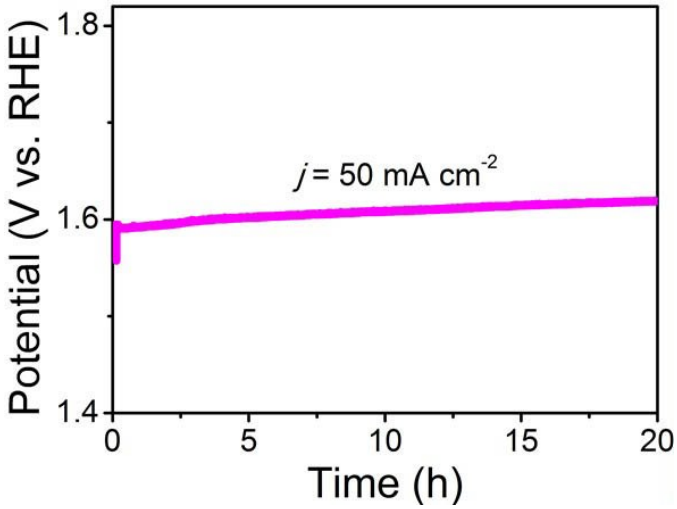

Fig. S14. Chronopotentiometric curve of Al-CoP/CC with constant current density of 50 mA cm^{-2} for OER in 1.0 M KOH (without iR correction).

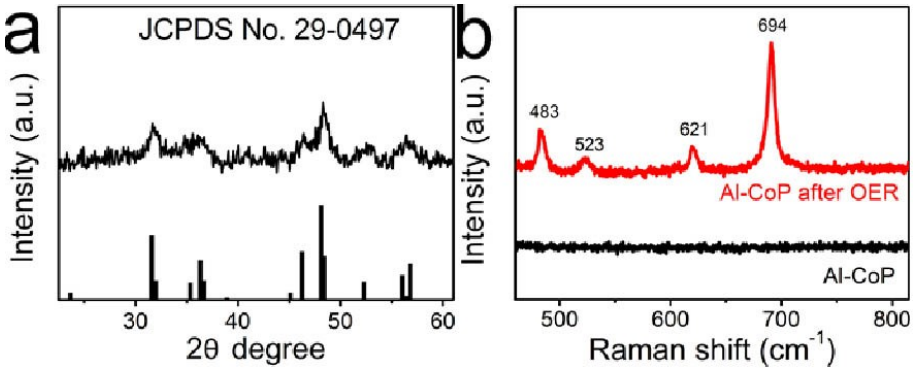

Fig. S15. (a)XRD pattern for Al-CoP after OER electrolysis and (b) Raman spectra for Al-CoP before and after OER electrolysis in 1.0 M KOH.

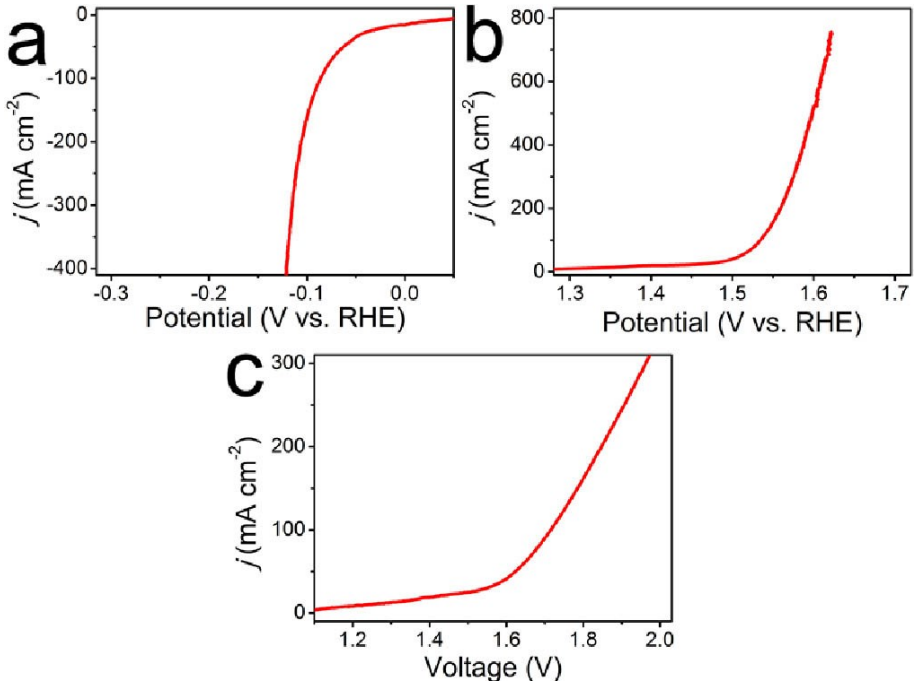

Fig. S16. Polarization curves for (a) HER and (b) OER with a scan rate of 5 mV s^{-1}. (c) Polarization curve of Al-CoP/NF‖Al-CoP/NF in a two-electrode setup for full water splitting with a scan rate of 5 mV s^{-1}. All experiments were performed in 1.0 M KOH.

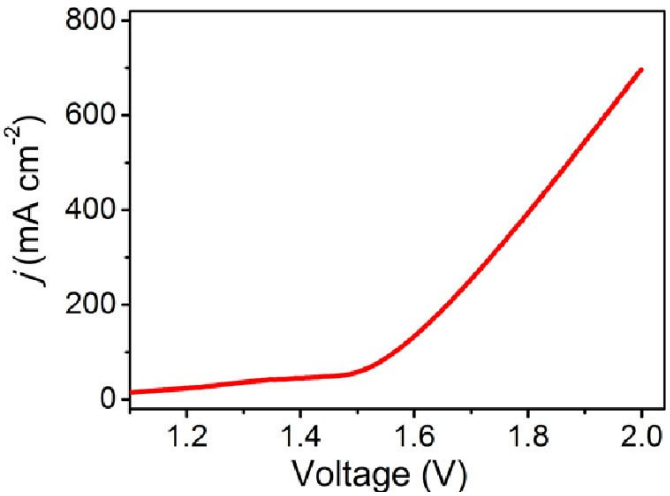

Fig. S17. (a) Polarization curve of Al-CoP/NF‖Al-CoP/NF in a two-electrode setup for full water splitting with a scan rate of 5 mV s^{-1} in 1.0 M KOH at 65 ^{o}C.

Table S1. Comparisons of HER performances of Al-CoP/CC with other Co-based phosphides HER catalysts in 0.5 M H_2SO_4.

Catalyst	*j* (mA cm^{-2})	Overpotential (mV)	Ref.
CoP nanowire/CC	10	67	1
CoP/CNT	10	122	2
CoP/Ti	10	90	3
hollow CoP nanoparticles	10	75	4
urchin-like CoP nanocrystals	10	~95	5
CoP nanosheet/CC	10	49	6
Co_2P nanopartical	10	95	7
u-CoP/Ti	10	45	8
CoP hollow polyhedron	10	159	9
Co_2P branched nanostructures	10	120	10
CoP_2/RGO	10	70	11
CoP/rGO-400	10	105	12
$Fe_{0.5}Co_{0.5}P$	10	130	13
$Co_{0.59}Fe_{0.41}P$ nanocubes	10	72	14
$Fe_{0.5}Co_{0.5}P$/CC	10	37	15
C@NiCoP	10	276	16
CoPS	10	48	17
CoS|P/CNT	10	48	18
Al-CoP/CC	10	23	This work

Table S2. Comparisons of HER performances of Al-CoP/CC and Al-CoP/NF with CoP catalysts and other non-precious metal HER catalysts in 1.0 M KOH.

Catalyst	*j* (mA cm^{-2})	Overpotential (mV)	Ref.
CoP nanowire/CC	10	209	1
u-CoP/Ti	10	72	8
CoP/rGO-400	10	340	12
CoP-MNA/NF	10	54	19
np-CoP NWs/Ti	10	100	20
Co-P	10	94	21
CoP_2/RGO	10	88	22
NiP_2 NS/CC	10	102	23
FeP NAs/CC	10	218	24
MoP_2/CC	10	67	25
a-CoSe/Ti	10	121	26
NiSe/NF	10	96	27
MoC_x	10	151	28
np-CuTi	10	50	29
NiMo HNRs/TiM	10	92	30
Ni/NiO-CNT	10	100	31
NiS nanoframes	10	94	32
CoO_x@CN	10	230	33
$NiCo_2O_4$	10	110	34
$Ni_{0.33}Co_{0.67}S_2$	10	73	35
NiCoP/NF	50	133	36
NiCoP/NF	10	32	37
Al-CoP/CC	10	38	This work
Al-CoP/NF	50	64	
	36	50	

Table S3. Comparisons of OER performances of Al-CoP/CC and Al-CoP/NF with CoP catalysts and other non-precious metal OER catalysts in 1.0 M KOH.

Catalyst	j (mA cm^{-2})	Overpotential (mV)	Ref.
CoP-MNA/NF	10	290	19
Co-P	10	345	21
a-CoSe/Ti	10	292	26
CoOx@CN	10	260	33
$NiCo_2O_4$	10	230	34
$Ni_{0.33}Co_{0.67}S_2$	10	330	35
NiCoP/NF	50	308	36
NiCoP/NF	10	280	37
Co/Co_2P@NF	10	190	38
NiFe-LDH/CNT	10	258	39
LNiFeP/rGO	10	247	40
PCPTF	10	300	41
Co-Bi NS/G	10	290	42
Ni_2P particles	10	290	43
$Zn_xCo_{3-x}O_4$	10	320	44
$Ni_xCo_{3-x}O_4$	10	370	45
Co_3O_4C-NA	10	290	46
Al-CoP/CC	10	265	This work
Al-CoP/NF	50	280	

Table S4. Comparisons of overall water-splitting performance of Al-CoP/CC and Al-CoP/NF with other non-nobel-metal bifunctional catalysts in 1.0 M KOH.

Catalyst	j (mA cm^{-2})	Voltage (V)	Ref.
CoP-MNA‖CoP-MNA	10	1.62	19
Co-P‖Co-P	10	1.65	21
CoP_2/RGO‖CoP_2/RGO	10	1.56	22
a-CoSe/Ti‖a-CoSe/Ti	10	1.65	26
NiSe/NF‖NiSe/NF	10	1.63	27
NiMo HNR/TiM‖NiMo HNR/TiM	10	1.64	30
$NiCo_2O_4$‖$NiCo_2O_4$	10	1.65	34
NiCoP/NF‖NiCoP/NF	50	1.77	36
NiCoP/NF‖NiCoP/NF	10	1.58	37
Ni_2P/NF‖Ni_2P/NF	10	1.63	43
Co_2B‖Co_2B	10	1.61	47
Ni_5P_4‖Ni_5P_4	10	below 1.70	48
$NiCo_2S_4$/CC‖$NiCo_2S_4$/CC	10	1.68	49
$Co_{0.13}Ni_{0.87}Se_2$‖$Co_{0.13}Ni_{0.87}Se_2$	10	1.62	50
$NiFe_2N_2$‖$NiFe_2N_2$	10	1.65	51
NiFe LDH/NF‖NiFe LDH/NF	10	1.70	52
Al-CoP/CC‖Al-CoP/CC	10	1.56	This work
	50	1.69	
Al-CoP/NF‖Al-CoP/NF	30	1.56	
	50	1.62	

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