# **Electronic Supplementary Information**

## **Experimental**

### Materials

Cobalt nitrate hexahydrate (Co(NO<sub>3</sub>)<sub>2</sub>·6H<sub>2</sub>O,  $\geq$  90%) and sodium hypophosphite (NaH<sub>2</sub>PO<sub>2</sub>,  $\geq$  99.0%) were obtained from Aladdin Industrial Corp. 2-Methylimidazole (C<sub>4</sub>H<sub>6</sub>N<sub>2</sub>,  $\geq$  99.0%) was received from Sinopharm Chemical Reagent Co. Ltd. Carbon cloth with hydrophilic surfaces (WOS 1002) was purchased from the CeTech Co. Ltd., Chinese Taipei. Deionized (DI) water was used throughout all experiments. All chemicals of analytical reagent grade were used directly without further purification during the experiments.

## Physical characterization

Scanning electron microscope (SEM) images, energy-dispersive spectrometry (EDS) results and corresponding elemental mappings were acquired on a Hitachi S-4800microscope at an accelerating voltage of 10 kV. Transmission electron microscopy (TEM), high-resolution TEM (HRTEM) and selected-area electron diffraction (SAED) images were obtained on a Hitachi H-8100 electron microscopy (Hitachi, Tokyo, Japan) with an accelerating voltage of 200 kV. X-ray diffraction (XRD) analyses were performed by a Bruker D8 Advanced Diffractometer System with a Cu K $\alpha$  (1.5418 Å) as the radiation source (40 kV, 40 mA) in transmission mode over the  $\theta$  range of 5-100°. X-ray photoelectron spectroscopy (XPS) measurements were taken on an ESCALABMK II X-ray photoelectron spectrometer using Mg as the exciting source.

#### **Preparation of Co-MOF NS/CC**

Co-MOF nanosheets array on carbon cloth (Co-MOF NS/CC) was synthesized by a simple precipitation method. The carbon cloth (CC) was firstly immersed in concentrated nitric acid and maintained at 100 °C for 3 h with the protection of a 50 mL Teflon-lined stainless steel autoclave to xxx. After this pretreatment, 0.582 g of  $Co(NO_3)_2$ •6H<sub>2</sub>O was dissolved in 40 mL deionized (DI) water to form solution A, and 1.312 g of 2-methylimidazole (C<sub>4</sub>H<sub>6</sub>N<sub>2</sub>) was dispersed in 40 mL of DI water to form

solution B. Then, solution B was quickly added into solution A, after that a piece of acid-treated CC ( $2 \times 3 \text{ cm}^2$ ) was immersed into the mixture solution. After reaction for 4 h at room temperature, the sample was taken out, cleaned with deionized water and dried overnight.

#### Preparation of Co<sub>3</sub>O<sub>4</sub> NS/CC and CoP NS/CC

The above-prepared Co-MOF NS/CC was calcined at 400 °C for 2 h with a heating rate of 2 °C min<sup>-1</sup> in air to form Co<sub>3</sub>O<sub>4</sub> nanosheets array on CC (CoP NS/CC). The loading mass of Co<sub>3</sub>O<sub>4</sub> on CC was calculated to be about 0.22 mg cm<sup>-2</sup>. In a typical synthesis of CoP phase, the Co<sub>3</sub>O<sub>4</sub> nanosheets precursor was phosphided by thermal decomposition of NaH<sub>2</sub>PO<sub>2</sub> under Ar flow. To be specific, a piece of Co<sub>3</sub>O<sub>4</sub> NS/CC ( $2 \times 1.5 \text{ cm}^2$ ) and 300 mg of NaH<sub>2</sub>PO<sub>2</sub> were put in two porcelain boats with NaH<sub>2</sub>PO<sub>2</sub> at the upstream side of tube furnace. Subsequently, the samples were annealed at 300 °C for 2 h with a heating rate of 2 °C min<sup>-1</sup> under Ar atmosphere. The CoP NS/CC was obtained after naturally cooling to ambient temperature under Ar. The loading mass of CoP on CC was calculated to be about 0.28 mg cm<sup>-2</sup>.

## Electrochemical characterization

Electrochemical tests were recorded on a computer-controlled electrochemical workstation (CHI660E, CH Instruments, Inc., Shanghai) with a home-made three-electrode configuration at room temperature. A mercuric oxide electrode (MOE) and a graphite plate were used as the reference and counter electrode in alkaline solution, respectively, while a saturated calomel electrode (SCE) served as the reference electrode in acidic solution. The as-prepared CoP NS/CC, Co<sub>3</sub>O<sub>4</sub> NS/CC and Co-MOF NS/CC were utilized as the working electrodes with a fixed catalytic area controlled as 0.25 cm<sup>2</sup> in all experiments. All the measured potentials should be finally converted to the reversible hydrogen electrode (RHE) according to the following equations: E (RHE) = E (MOE) + 0.9254 V in 1.0 M KOH and E (RHE) = E (SCE) + 0.259 V in 0.5 M H<sub>2</sub>SO<sub>4</sub>.

The HER and OER activities of CoP NS/CC were studied by steady-state linear sweep voltammetry at 2 mV s<sup>-1</sup>. Electrochemical impedance spectroscopy (EIS) measurements were carried out by applying an AC voltage of 0.60 V *vs*. MOE with 5

mV amplitude in the frequency range of 100 kHz to 0.005 Hz. Overall alkaline water electrolysis was performed in a single-compartment 1.0 M KOH cell from 2.0 to 1.0 V at 2 mV s<sup>-1</sup>, in which the obtained samples were served as both the anode and cathode. Tafel plots were expressed as the overpotential  $(\eta)$  vs. log current  $(\log |j|)$ . By fitting the linear portion of Tafel plots to the Tafel equation of  $[\eta = b \log (j) + a]$ , the Tafel slope (b) could be obtained. iR correction was conducted by the equation: E = $E_{\rm m}$  -  $iR_{\rm s}$  (where  $R_{\rm s}$  is the resistance of the solution, i is the current density, E is the corrected potential, and  $E_m$  is the measured potential). The electrochemical double-layer capacitances  $(C_{dl})$  of above samples were also compared by calculating the half values of slopes from the relationship curves of charging current density vs. scan rate derived from the cyclic voltammetric curves under different scan rates from 10-100 mV s<sup>-1</sup> in the potential window of 0.3-0.4 V vs. MOE without redox process in 1.0 M KOH. An acid base hybrid electrolyzer was constructed by this bifunctional CoP NS/CC electrode. This system was composed of two glass bottles connected with an open window using the Nafion 117 membrane as the proton exchange membrane to separate the anolyte and catholyte (1.0 M KOH and 0.5 M H<sub>2</sub>SO<sub>4</sub>, respectively). Linear sweep voltammetric curve of the CoP NS/CC (+)||CoP NS/CC (-) was recorded at a scan rate of 2 mV s<sup>-1</sup> from 1.7 V to 0.3 V to evaluate its overall acid|base hybrid water-splitting performance. Both the overall alkaline water-splitting and acid/base hybrid water-splitting devices were driven by the electrochemical workstation or a 2.0 V solar panel under the irradiation of a commercial 300 W xenon lamp (Perfectlight Technology, Beijing) under chopped AM 1.5 G illumination.



**Fig. S1**. (a) Optical photograph of Co-MOF NS/CC, Co<sub>3</sub>O<sub>4</sub> NS/CC, and CoP NS/CC (from left to right). (b-e) Optical photographs of the CoP NS/CC at different bending angles. (f-h) SEM images of the CoP nanoarray structures after bended at different angles shown in b-d, respectively.



**Fig. S2.** High-magnification top (left) and lateral (right) views of the CoP nanoflakes array structure.



**Fig. S3.** Energy-dispersive X-ray spectrum and elemental mapping images of the CoP NS/CC.



Fig. S4. XPS survey spectrum of the CoP NS/CC.



Fig. S5. BET surface areas of ZIF-L-Co, Co<sub>3</sub>O<sub>4</sub>, and CoP nanosheets powders.



Fig. S6. Mass-normalized alkaline (a) HER and (b) OER performances of the CoP NS/CC and  $Co_3O_4$  NS/CC.



**Fig. S7.** Long-term stability tests of the CoP NS/CC toward alkaline (a) HER and (b) OER.



Fig. S8. Mass-normalized acidic HER performance of this CoP NS/CC.



Fig. S9. Long-term stability test of this CoP NS/CC toward acidic HER.



Fig. S10. Pourbaix diagram of  $H_2/O_2$  at 25 °C and atmospheric pressure.



**Fig. S11.** The acid|base hybrid overall water-splitting performances of CoP NS/CC (+,-) recorded before and after 35 h electrolysis, as well as after electrolysis in new acid/base solutions.



**Fig. S12**. Low- and high-magnification SEM images of the CoP NS/CC after acidic (a-c) HER and (d-f) alkaline OER catalysis in the acid|base dual-electrolyte overall water-splitting system.



Fig. S13. XRD patterns of the CoP NS/CC after long-term water electrolysis.



Fig. S14. EDX spectra of the CoP NS/CC after long-term water electrolysis.



**Fig. S15.** (a) Co 2p and (b) P 2p XPS spectra recorded after long-term acidic HER and alkaline OER.

Movie S1. Solar-panel powered alkaline water-splitting device.

Movie S2. Enlarged view of H<sub>2</sub>/O<sub>2</sub> generation in the alkaline water-splitting device.

Movie S3. Solar-panel powered acid|base water-splitting device.

Movie S4. Enlarged view of H<sub>2</sub>/O<sub>2</sub> generation in the acid|base water-splitting device.

	Peak	Position (eV)	Area	FWHM (eV)	%GL (%)
Co 2p	0 (Sat.)	803.253	9364.524	5.033	80
	1 (Co <sup>2+</sup> )	798.554	9996.572	2.576	80
	2 (Co <sup>3+</sup> )	794.126	1747.299	0.862	80
	3 (Sat.)	784.582	15619.020	5.534	80
	4 (Co <sup>2+</sup> )	782.210	13988.300	2.187	80
	5 (Co <sup>3+</sup> )	779.210	9037.135	1.082	80

**Table S1.** Peak areas of different valence states in the Co 2p regions for CoP NS/CC.

Catalytic material	Current density (mA cm <sup>-2</sup> )	Overpotential (mV)	Ref.
CoP NS/CC	10	90	This work
CoP@NC	10	129	1
CoS <sub>2</sub> NTA/CC	10	193	2
Cu <sub>0.3</sub> Co <sub>2.7</sub> P/NC	10	220	3
Co-doped nickel selenide	10	90	4
Ni <sub>3</sub> S <sub>2</sub> -NGQDs/NF	10	218	5
Ni@CoO@CoNC	10	190	б
2D porous CoPO nanosheets	PO 10 158		7
Multi-shelled Ni <sub>2</sub> P	Ni <sub>2</sub> P 10 98		8
Ni-Co-P nanocubes	10	150	9
Mo <sub>2</sub> C/carbon microflowers	10 100		10
Ni <sub>2</sub> P film	10	183	11
Co/CoP	10	135	12
Co <sub>4</sub> Ni <sub>1</sub> P nanotubes	nanotubes 10 129		13
CNTs@Co-S/CP	10	190	14
Co <sub>1</sub> Mn <sub>1</sub> CH/NF	10	200	15
CoP/CC	oP/CC 10 209		16
Ni <sub>2</sub> P/CC	10	~205	17
2D-MoS <sub>2</sub> /Co(OH) <sub>2</sub>	10	128	18
MoB/g-C <sub>3</sub> N <sub>4</sub>	10	133	19

**Table S2.** Comparison of the alkaline HER efficiency of this CoP NS/CC with other reported advanced cathodic materials.

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Catalytic material	Current density (mA cm <sup>-2</sup> )	Overpotential (mV)	Ref.
CoP NS/CC	10	310	This work
CoP NR/C	10	320	1
CoP NP/C	10	340	1
CoP/rGO-400	10	340	2
MOF-CoP/NC	10	319	3
CoP/C	10	330	4
CoP@GC	10	345	5
Ni-CoP	10	360	6
Ag@Co <sub>x</sub> P	10	310	7
NiCoP/C	10	330	8
(Co <sub>0.54</sub> Fe <sub>0.46</sub> ) <sub>2</sub> P	10	370	9
Co <sub>2</sub> P/Co-Foil	10	319	10
Mn-Co oxyphosphide	10	320	11
CoMnP	10	330	12
Ni-Co mixed oxide	10	380	13
Ni <sub>x</sub> Co <sub>3-x</sub> O <sub>4</sub>	10	337	14
NiO@NF-6	10	405	15
Au@CoSe <sub>2</sub>	10	430	16
NiO/Ni	10	345	17
Ni(OH)2@O-MWCNTs	10	310	18
Cu <sub>x</sub> Co <sub>3-x</sub> O <sub>4</sub>	10	410	19
Ni <sub>x</sub> Co <sub>3-x</sub> O <sub>4</sub>	10	337	20
$Zn_{0.1}Co_{0.9}Se_2$	10	340	21
NiCo <sub>2</sub> O <sub>4</sub> rich in oxygen vacancies	10	320	22
Ni-Co <sub>2</sub> -O hollow nanosponges	10	362	23

**Table S3.** Comparison of the alkaline OER efficiency of this CoP NS/CC with other reported advanced anodic materials.

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Table S4.	Comparison	of the	alkaline	overall	water-splitting	efficiency	of this	CoP
NS/CC wit	th other repor	ted adv	anced bi	function	al catalysts.			

Catalytic material	Current density (mA cm <sup>-2</sup> )	Voltage (V)	Ref.	
CoP NS/CC	10	1.67	This work	
CoS <sub>2</sub> NTA/CC	10	1.67	1	
2D porous CoPO nanosheets	10	1.67	2	
amorphous Co <sub>2</sub> B	10	1.81	3	
Co-S/carbon tubes/CP	10	1.743	4	
Hierarchical NiCo <sub>2</sub> O <sub>4</sub> hollow microcuboids	10	1.65	5	
CoP/rGO-400	10	1.7	6	
PPy/FeTCPP/Co	10	1.81	7	
Co <sub>0.85</sub> Se/NiFe-LDH	10	1.67	8	
NiCo <sub>2</sub> S <sub>4</sub> NW/CC	10	1.68	9	
Ni <sub>3</sub> S <sub>2</sub> NS/NF	10	1.70	10	
CoS <sub>2</sub> NT/CC	10	1.67	11	
NiFe/NiCo <sub>2</sub> O <sub>4</sub> /NF	10	1.67	12	
Ni <sub>5</sub> P <sub>4</sub> /Ni foil	10	1.69	13	
SrNb <sub>0.1</sub> Co <sub>0.7</sub> Fe <sub>0.2</sub> O <sub>3-δ</sub>	10	1.68	14	

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**Table S5.** Comparison of the acidic HER efficiency of this CoP NS/CC with other reported advanced cathodic materials.

Catalytic material	Current density (mA cm <sup>-2</sup> )	Overpotential (mV)	Tafel slope (mV dec <sup>-1</sup> )	Ref.
CoP NS/CC	10	88.7	52.7	This work
Ni <sub>2</sub> P Ps-3	10	158	73	1
Ni <sub>2</sub> P-CoP	10	105	65	2
NiCo/NiCoO <sub>x</sub>	10	155	55	3
Zn <sub>0.1</sub> Co <sub>0.9</sub> Se <sub>2</sub>	10	140	49.9	4
Hollow Zn <sub>0.30</sub> Co <sub>2.70</sub> S <sub>4</sub>	10	80	50.4	5
CoP/NCNTs	10	383	49	6
CoP hollow polyhedron	10	159	59	7
Fe <sub>x</sub> P@NPC	10	227	81	8
CoP/CFP-H	10	128.1	49.7	9
CoP@BCN-1	10	87	54	10
MoP@PC	10	153	66	11
N-WC/CFP	10	113	75	12

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