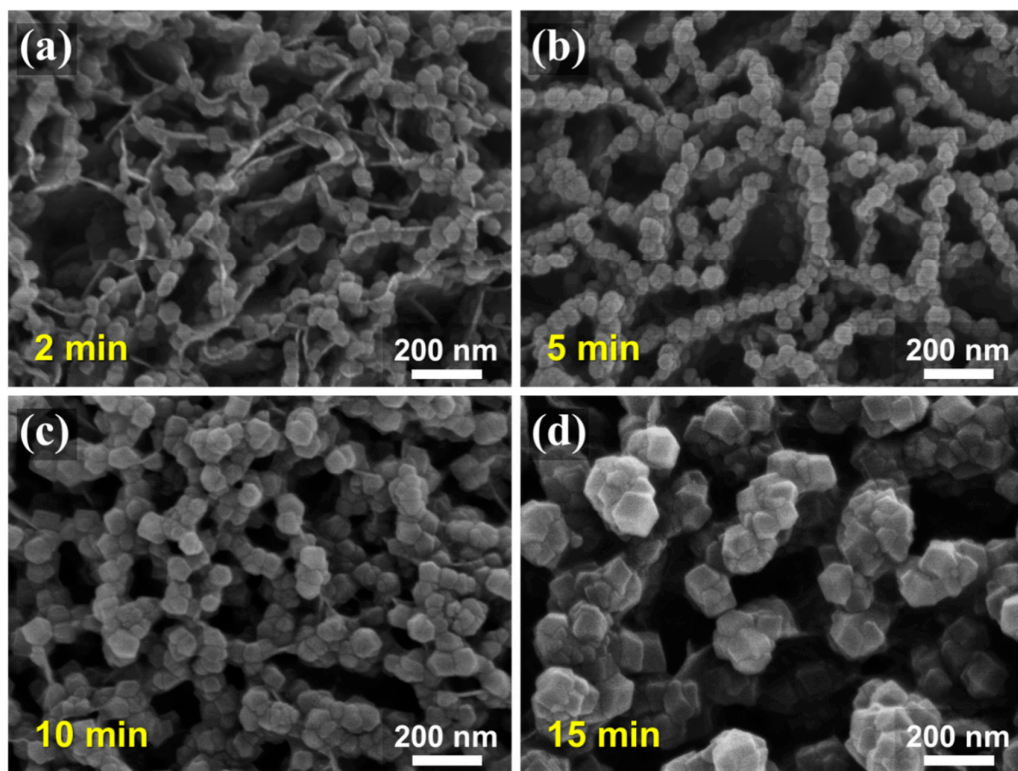


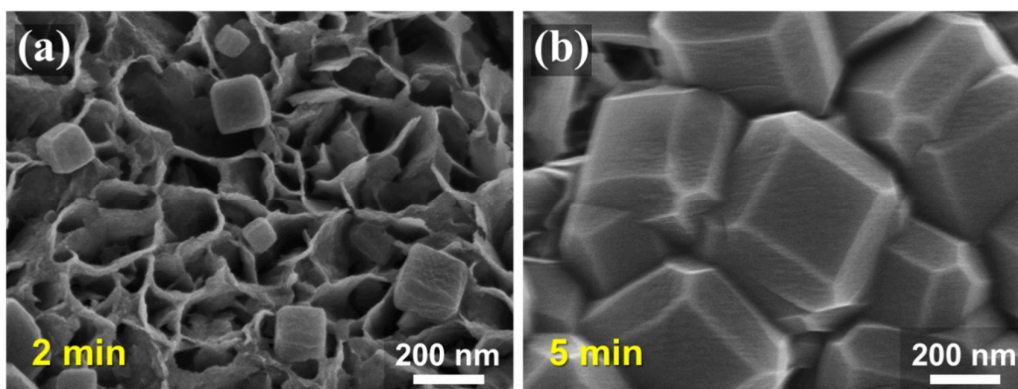
## Supporting Information

### **MOF Template-Derived Carbon Shell Embedded CoP Hierarchical Nanosheet as Bifunctional Catalyst for Overall Water Splitting**

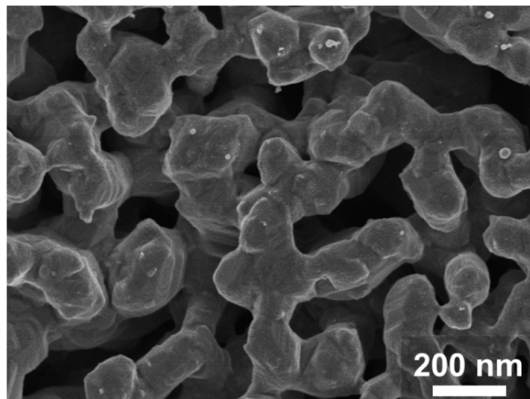
Meijun Liu<sup>a,b</sup>, Fuhao Yang<sup>a</sup>, Jicheng Mei<sup>a,b</sup>, Xu Guo<sup>a</sup>, Huayang Wang<sup>a</sup>, Mengyao He<sup>a</sup>,  
Yuang Yao<sup>a</sup>, Haifeng Zhang<sup>a,\*</sup>, Chengbin Liu<sup>b,\*</sup>



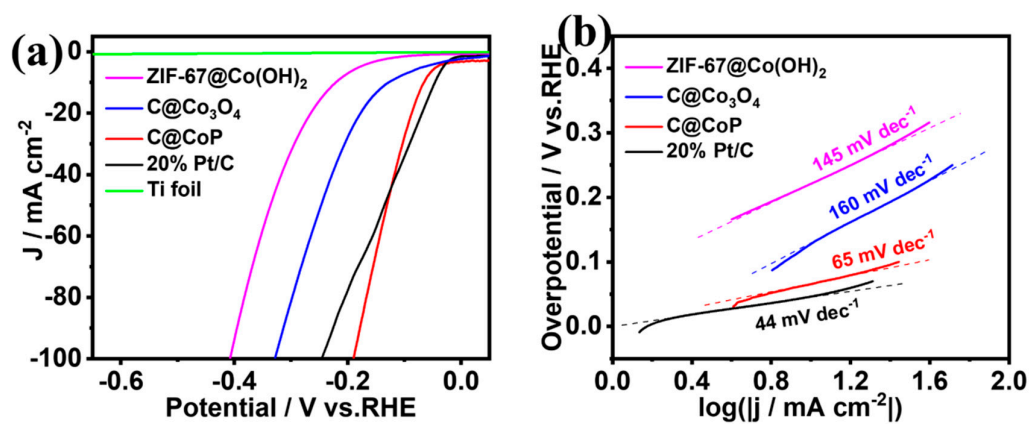
**Figure S1.** SEM images of ZIF-67 crystals grown on Co(OH)<sub>2</sub> nanosheets for (a) 2 min, (b) 5 min, (c) 10 min and (d) 15 min.



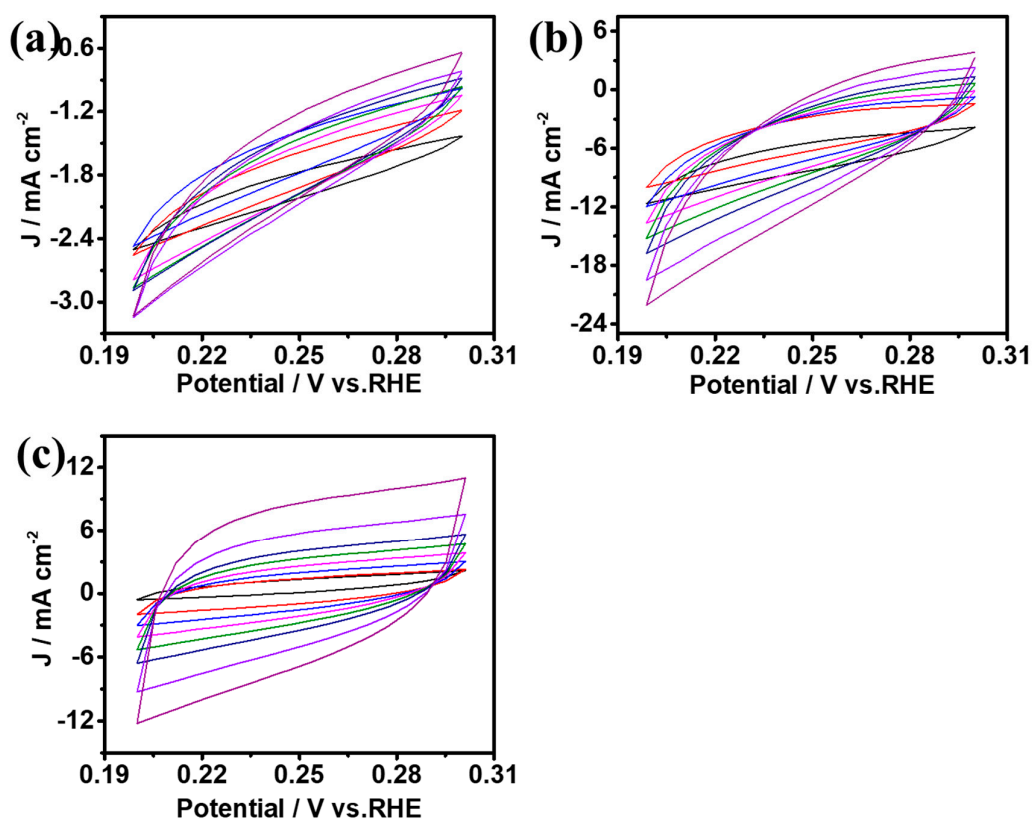
**Figure S2.** SEM images of ZIF-67 crystals grown on Co(OH)<sub>2</sub> nanosheets for (a) 2 min and (b) 5 min without the acceleration of TEA.



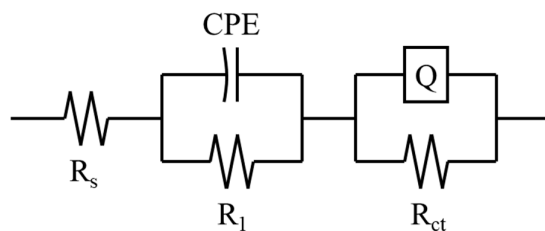
**Figure S3.** SEM image of ZIF-67@Co(OH)<sub>2</sub>-C prepared by directly pyrolytic carbonization of ZIF-67@Co(OH)<sub>2</sub>.



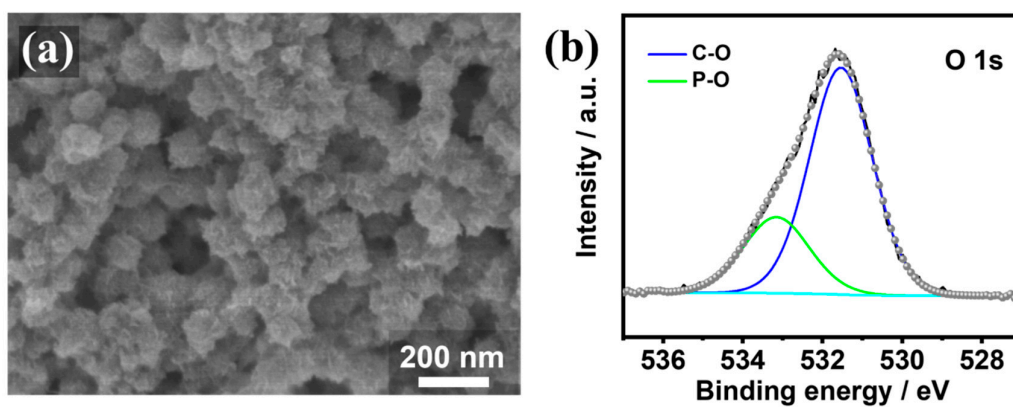
**Figure S4.** (a) HER Polarization curves and corresponding (b) Tafel plots for C@CoP and reference materials.



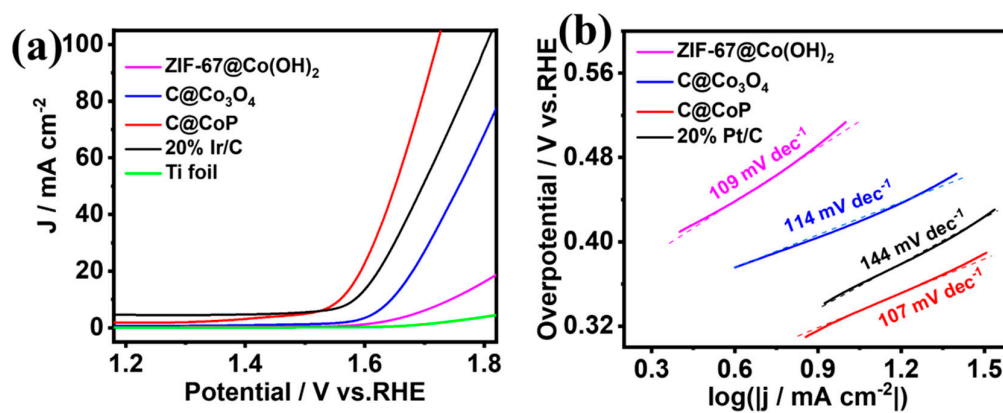
**Figure S5.** CV curves of (a) C@Co<sub>3</sub>O<sub>4</sub>, (b) CoP and (c) C@CoP in the range of 0.2–0.3 V vs. RHE. The curves from inside to outside correspond to the scanning rate of 10, 20, 30, 40, 50, 60, 80 and 100 mV s<sup>-1</sup>, respectively.



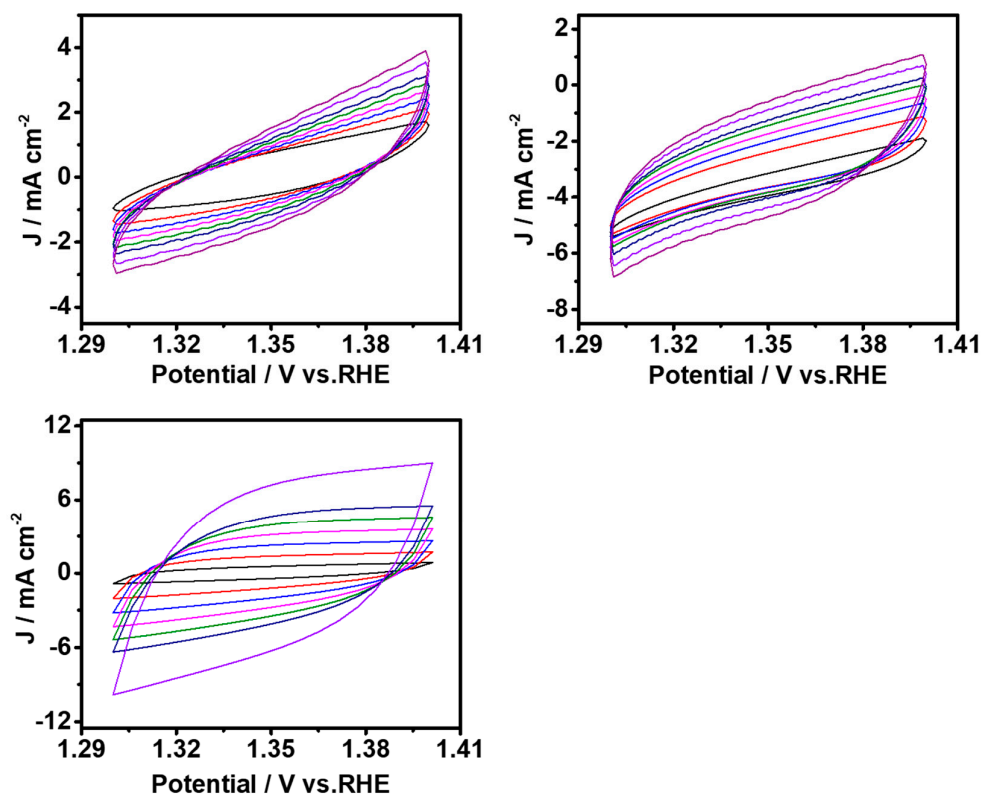
**Figure S6.** Equivalent circuit fitted according to the Nyquist diagram in Figure 5d and Figure 6d.



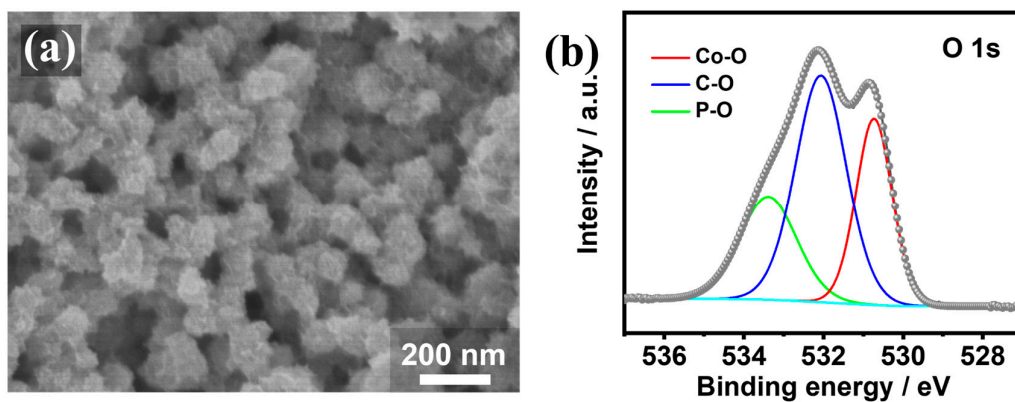
**Figure S7.** (a) SEM image and (b) high-resolution XPS spectra of O 1s of C@CoP after long term HER test.



**Figure S8.** (a) OER Polarization curves and corresponding (b) Tafel plots for C@CoP and reference materials.



**Figure S9.** CV curves of (a) C@Co<sub>3</sub>O<sub>4</sub>, (b) CoP and (c) C@CoP in the range of 1.3–1.4 V vs.RHE. The curves from inside to outside correspond to the scanning rate of 10, 20, 30, 40, 50, 60, 80 and 100 mV s<sup>-1</sup>, respectively.



**Figure S10.** (a) SEM image and (b) high-resolution XPS spectra of O 1s of C@CoP after long term OER test.



**Table S1.** Comparison of HER performance for C@CoP with other HER electrocatalysts

Catalyst	Overpotential (mV) at 10 mA/cm <sup>2</sup>	Tafel slope (mV/dec)	Electrolyte	Reference
C@CoP	72	65	1.0 M KOH	This work
Ni-CoP/HPFs	144	62	0.5 M H <sub>2</sub> SO <sub>4</sub>	1
	92	34	1.0 M KOH	
Mn <sub>2</sub> P-Mn <sub>2</sub> O <sub>3</sub> /PNCF	98	46	1.0 M KOH	2
FePx/ Fe-N-C /NPC	75	60	0.5 M H <sub>2</sub> SO <sub>4</sub>	3
	182	132	1.0 M KOH	
CoP/Co-MOF	27	43	0.5 M H <sub>2</sub> SO <sub>4</sub>	4
	34	56	1.0 M KOH	
	49	63	1.0M PBS	
PMA@ZIF-67-C-AT	570	222	0.2M PBS	5
Co <sub>5</sub> Fe <sub>5</sub> -C	165	70.7	1.0 M KOH	6
V-CoP <sub>2</sub> /CC	50	32	0.5 M H <sub>2</sub> SO <sub>4</sub>	7
VCoCOx@NF	63	93	1.0 M KOH	8
Co <sub>2</sub> P/CoP@Co@NCNT	118	46	1.0 M KOH	9
	136	49	0.5 M H <sub>2</sub> SO <sub>4</sub>	



**Table S2.** Electrochemical impedance parameters obtained by fitting the Nyquist plots of Figure 5d to the equivalent circuit model

Catalyst	$R_s$ ( $\Omega$ )	C (mF cm <sup>-2</sup> )	$R_1$ ( $\Omega$ )	Q		$R_{ct}$ ( $\Omega$ )
				$Y_1(\Omega^{-1}s^n)$	n	
C@CoP	1.01	3.89	4.30	$2.56 \times 10^{-3}$	0.480	4.30
CoP	1.17	5.43	5.43	$5.03 \times 10^{-3}$	0.254	5.43
C@Co <sub>3</sub> O <sub>4</sub>	1.22	0.966	22.40	$5.98 \times 10^{-3}$	0.872	22.40

**Table S3.** Comparison of OER performance of C@CoP with other reported electrocatalysts.

Catalyst	Overpotential (mV) at 10 mA/cm <sup>2</sup>	Tafel slope (mV/dec)	Electrolyte	Reference
<b>C@CoP</b>	329	107	1.0 M KOH	This work
<b>Co<sub>5</sub>Fe<sub>5</sub>-C</b>	245	58.2	1.0 M KOH	<sup>6</sup>
<b>W<sub>0.2</sub>Er<sub>0.1</sub>Ru<sub>0.7</sub>O<sub>2-δ</sub></b>	168	66.8	0.5M H <sub>2</sub> SO <sub>4</sub>	<sup>10</sup>
<b>np-Ir/NiFeO</b>	197	29.6	1.0 M KOH	<sup>11</sup>
<b>Mn<sub>2</sub>P-Mn<sub>2</sub>O<sub>3</sub>/PNCf</b>	370	86	1.0 M KOH	<sup>2</sup>
<b>FePx/ Fe-N-C /NPC</b>	325	79	1.0 M KOH	<sup>3</sup>
<b>NiMoP@NiFe-LDH</b>	299	23.3	1.0 M KOH	<sup>12</sup>
<b>V-CoP<sub>2</sub>/CC</b>	91	40	0.5 M H <sub>2</sub> SO <sub>4</sub>	<sup>7</sup>
<b>VCoCO<sub>x</sub>@NF</b>	240	65	1.0 M KOH	<sup>8</sup>
<b>Co<sub>2</sub>P/CoP@Co@NCNT</b>	256	46	1.0 M KOH	<sup>9</sup>

**Table S4.** Electrochemical impedance parameters obtained by fitting the Nyquist plots

of Figure 6d to the equivalent circuit model

Catalyst	$R_s$ ( $\Omega$ )	C (mF cm <sup>-2</sup> )	$R_1$ ( $\Omega$ )	Q		$R_{ct}$ ( $\Omega$ )
				$Y_1(\Omega^{-1}s^n)$	n	
C@CoP	2.17	4.90	9.66	$6.75 \times 10^{-3}$	0.604	0.59
CoP	2.68	6.75	8.55	$4.45 \times 10^{-3}$	0.968	3.81

**Table S5.** Comparison of overall water splitting performance of C@CoP with recent representative works.

Catalyst	Cell voltage (V) at 10 mA/cm <sup>2</sup>	Electrolyte	Reference
C@CoP	1.63	1.0 M KOH	This work
Mn <sub>2</sub> P-Mn <sub>2</sub> O <sub>3</sub> /PNCf	1.6	1.0 M KOH	<sup>2</sup>
FePx/ Fe-N-C /NPC	1.58	1.0 M KOH	<sup>3</sup>
Ir <sub>1</sub> @Co/NC	1.603	1.0 M KOH	<sup>13</sup>
Co-Fe NPs	1.92	1.0 M KOH	<sup>14</sup>
Fe-NiS <sub>2</sub> /CF	1.722	1.0 M KOH	<sup>15</sup>
O-Ni <sub>0.5</sub> W <sub>0.5</sub> Se <sub>2</sub>	1.56	1.0 M KOH	<sup>16</sup>
Co <sub>0.75</sub> Fe <sub>0.25</sub> P	1.63	1.0 M KOH	<sup>17</sup>
Co <sub>2</sub> P/CoP@Co@NCNT	1.6	1.0 M KOH	<sup>9</sup>
Co/Mo <sub>2</sub> C@NC800-2	1.67	1.0 M KOH	<sup>18</sup>

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