Scheme.1. A schematic diagram to illustrate the operating principle of the HER based on CoP NS /CC. Electrons transport along CoP nanosheets and transfer to proton leading to hydrogen production.

Fig.S1. XRD patterns for $CoF_2 \cdot 4H_2O$ and CoP NS/CC.

Fig.S2. Optical photograph (From left to right) of dark gray CC, $CoF_2 \cdot 4H_2O$ NS/CC, and CoP NS/CC.

Fig.S3. SEM image of CoP NS /CC after sonication for 30 min.

Fig.S4. SAED pattern of CoP nanosheets.

Fig. S5. XRD of CoP NS/CC initially and after 3000 CV.

Fig.S6. SEM images of CoP NS /CC after 90 min continuous CV scanning at (a) pH 0, (b) pH 7, and (c) pH 14.

Table S1 Comparison of HER performance in acid media for CoP NS/CC with other HER electrocatalysts (*a* catalysts directly grown on current collectors).

Fig. S7. Calculated exchange current density of CoP NS/CC by applying extrapolation method to the Tafel plot.

Fig.S8. The amount of H_2 theoretically calculated (solid) and experimentally measured (sphere) versus time for CoP NS/CC at (a) pH 0 (b) pH 7 and (c) pH 14 under overpotentials of 250 mV for 60 min.

Table S2 Comparison of HER performance in neutral media for CoP NS/CC with other HER electrocatalysts (a catalysts directly grown on current collectors).

Table S3 Comparison of HER performance in alkaline media for CoP NS/CC with other HER electrocatalysts (a catalysts directly grown on current collectors)

Table S4. Summary of the Electrochemical Properties of the different Electrocatalysts in 0.5 M H_2SO_4.

Table S5. Summary of the Electrochemical Properties of CoP NS/CC at various HER overpotentials in 0.5 M H_2SO_4.

Scheme.1

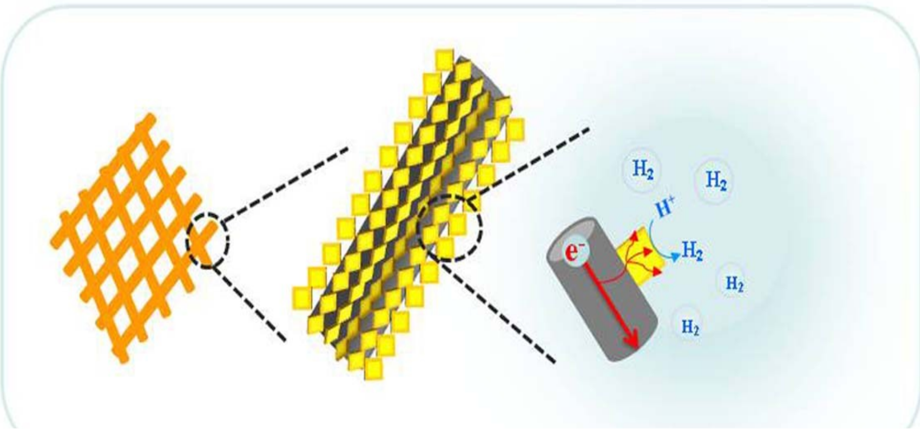


Fig.S1

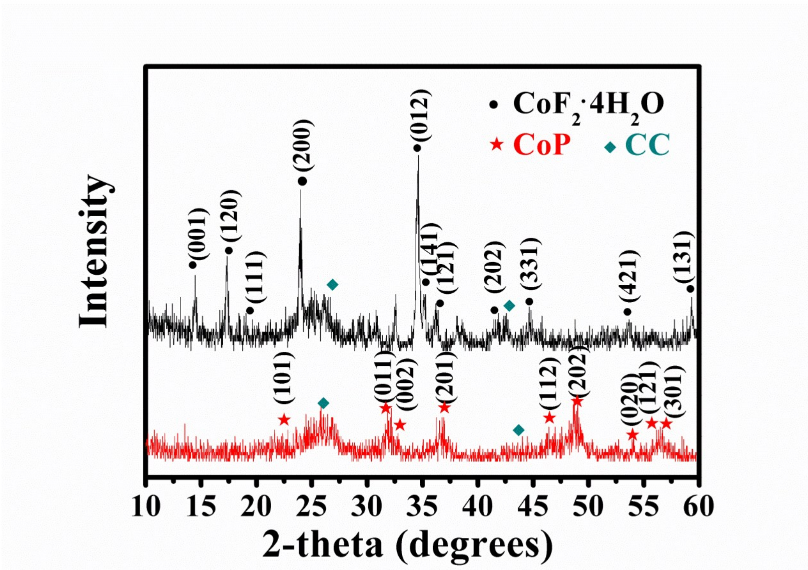


Fig.S2

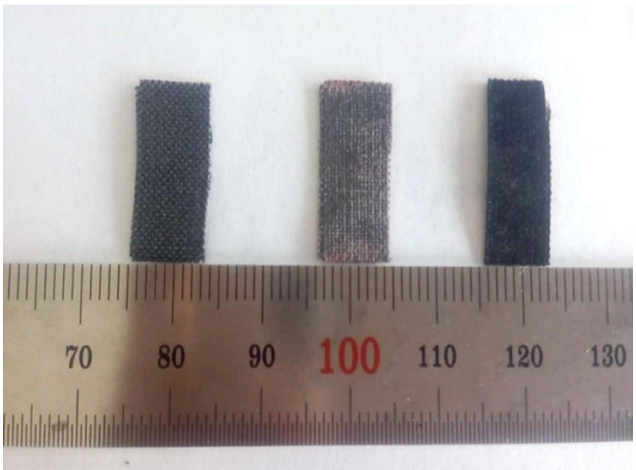
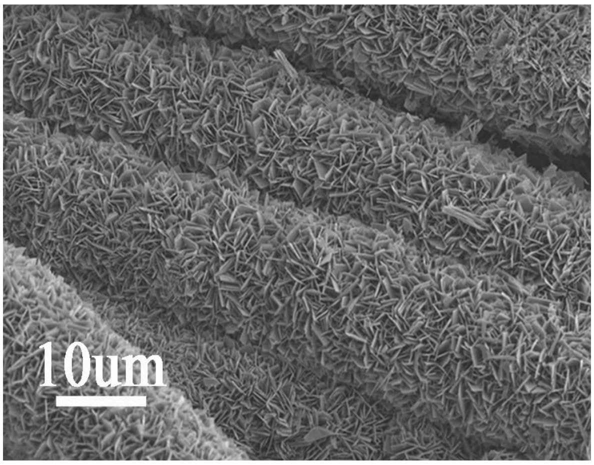


Fig.S3



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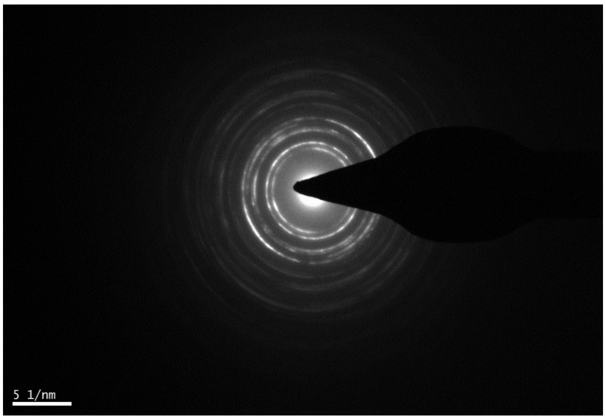

Fig.S5

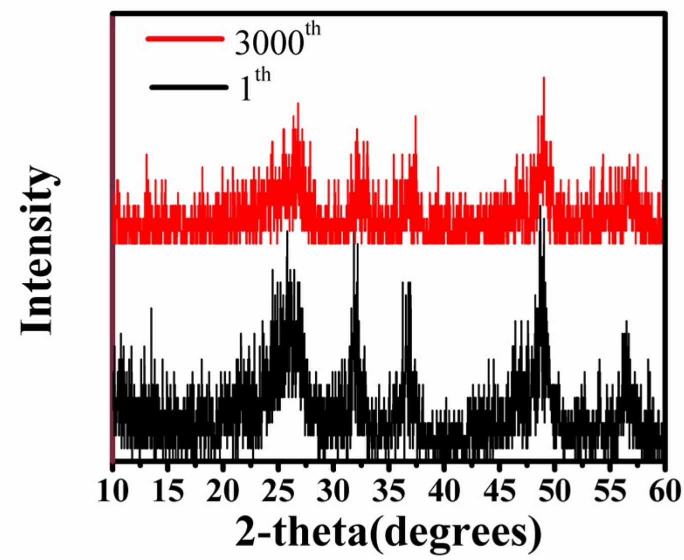

Fig.S6

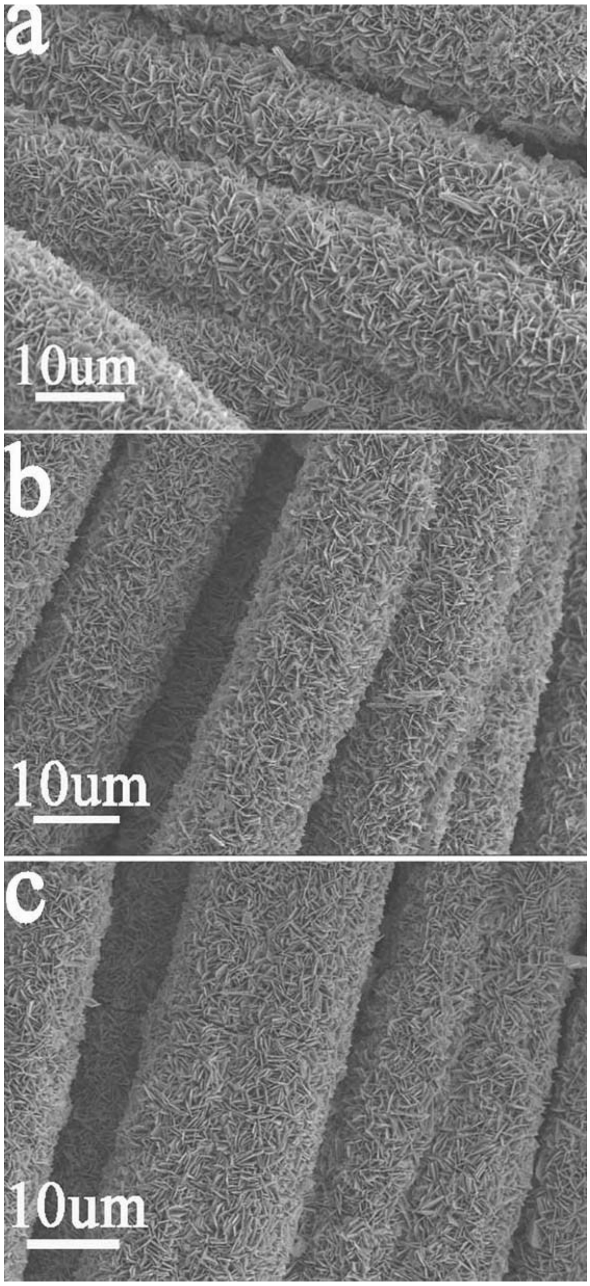

Table S1

Catalysts	Tafel slope (mVdec-1)	Current density (j, mA cm-2)	η at he corresponding j (mV)	Exchange current density (mA cm-2)	Ref.
Bulk Mo2C(2.0)	87.6	1	204	6.9 × 10-4	Energy Environ. Sci. 2013, 6, 943-951
Co-NRCNTs(0.28)	69	10	260	0.01	Angew.Chem.2014,126, 4461-4465
CoP/CNT(0.285)	54	10	122	0.13	Angew. Chem. 2014,126, 6828-6832
Nanoporous Mo2C NWs	53	60	200	------	Energy Environ. Sci. 2014, 7, 387-392
CoSe2 NP/CP	-----	10	139	(4.9±1.4) × 10-3	J. Am. Chem. Soc. 2014,136,4897-4900
Ni2P NPs	66	20	140	------	J. Am. Chem. Soc. 2013, 135, 9267-9270
MoP-CA2	40	10	1125	0.086	Adv. Mater. 2014, 26, 5702-5708
WS2 NSs	93	10	370	------	ACS Nano, 2015,9 , 5164-5179
NiP NPs/CNTs	53	10	124	0.537	J. Mater. Chem. A, 2015, 3, 13087-13094
MoS2@OMC	65	10	178	------	ACS Nano, 2015, 9, 3728-3739
CoSe2 NWs/CC	32	10	130	------	ACS Appl. Mater. Interfaces2015,7,877-388
NiMoNx/C	35.9	2	170	2.4×10-4	Angew. Chem. Int. Ed. 2012, 51, 6131
MoN/C	54.5	2	290	3.6×10-5	Angew. Chem. Int. Ed.2012, 51, 6131
Metallic MoS2 nanosheets	54	10	195	------	J. Am. Chem. Soc. 2013, 135, 10274
MoS2/graphene/ Ni foama	42.8	10	141	------	Adv. Mater. 2013, 25, 756
MoP/CF	56.4	10.1	200	------	Appl. Catal. B. 2014, 164, 144-150
This work	58	10	92	0.257	---------
		20	112		
		100	195		

Fig. S7

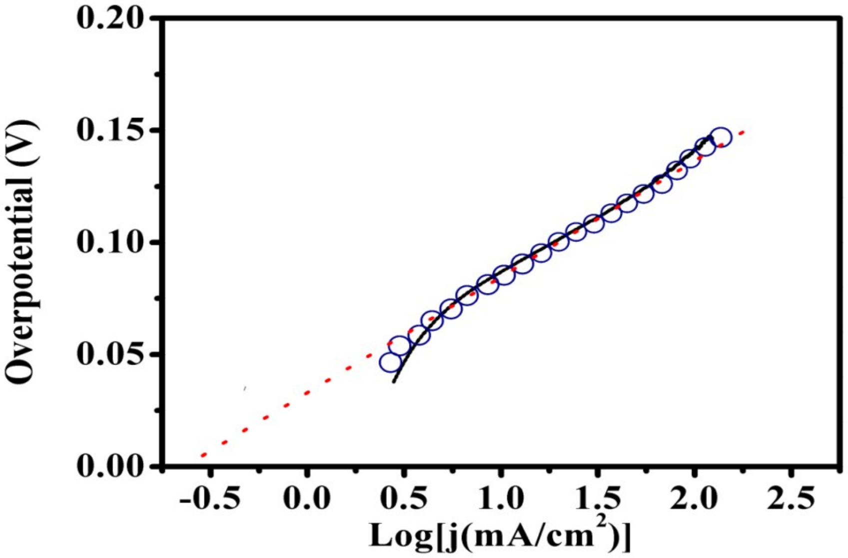
Overpotential (V)
Log[j(mA/cm^2)]
0.20
0.15
0.10
0.05
0.00
-0.5
0.0
0.5
1.0
1.5
2.0
2.5

Fig. S8

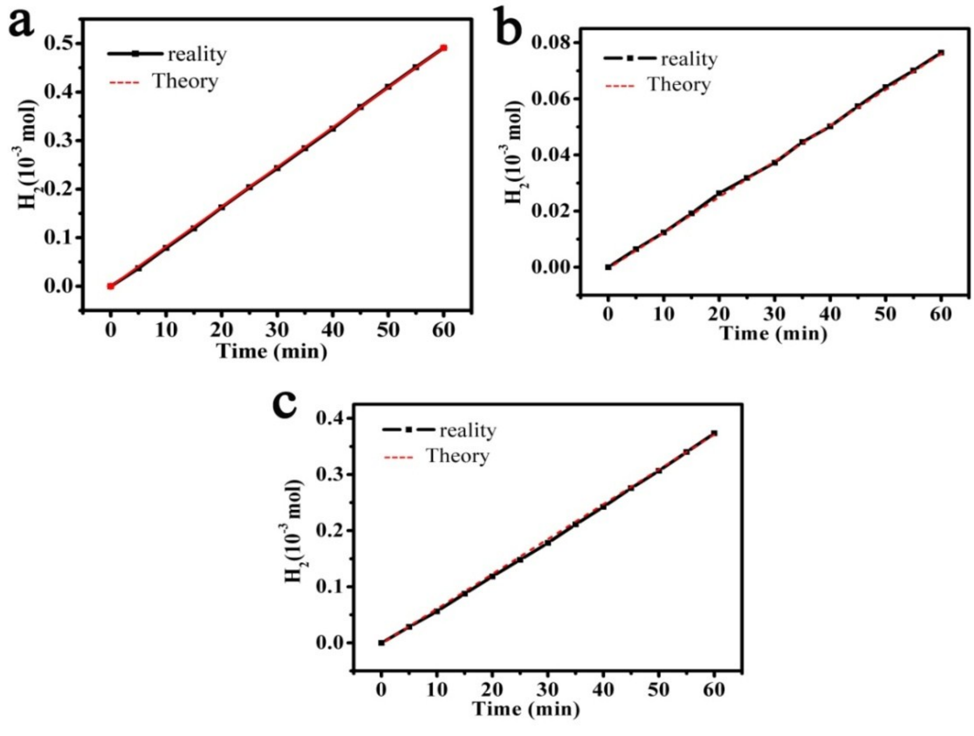
a
b
c
reality
Theory
H_2 (10^{-3} mol)
Time (min)

Table S2

Catalysts	Tafel slope ($mVdec^{-1}$)	Current density (j, mA cm^{-2})	η at the corresponding j (mV)	Ref.
H_2-CoCat/FTO[a]	140	2	385	Nat. Mater. 2012, 11, 802
Co-NRCNTs	—- -----	2	380	Angew. Chem. 2014, 126, 4461-4465
		10	540	
$CuMoS_4$ crystals	95	2	210	Energy Environ. Sci. 2014, 7, 387-392
MoS_3/FTO[a]	86	------	------	J. Am. Chem. Soc. 2014, 136, 4897-4900
Mo_2C	------	1	200	J. Am. Chem. Soc. 2013, 135, 9267-9270
Mo_2B	------	1	250	Adv. Mater. 2014, 26, 5702-5708
This work	93	1	64	---------
		2	94	
		10	160	

Table S3

Catalyst	Tafel slope (mV dec^{-1})	Current density (j, mA cm^{-2})	η at the corresponding j(mV)	Ref.
Ni	------	10	400	Angew. Chem. Int. Ed., 2012, 54, 12703
Co-NRCNTs	------	1	160	Angew. Chem. Int. Ed., 2014, 126, 4372
		10	370	
CoP/CC[a]	80	10	209	J. Am. Chem. Soc., 2014, 136, 7587
bulk MoB	59	10	225	Angew. Chem. Int. Ed., 2012, 54, 12703
NiP_2 NS/CC[a]	64	10	102	J. Am. Chem. Soc. 2014, 136, 4897-4900
Ni wire	------	20	350	ACS Catal., 2013, 3, 166
This work	68	10	92	-----
		20	96	
		100	142	

Tables S4

材料	R_s(Ω)	Q	n	R_{ct}
CC	2.83	2.522×10^{-5}	0.933	2.022×10^{-5}
CoP/CC	2.059	0.09663	0.854	366.1

Table S5

过电位（mV）	R_s	Q	n	R_{ct}
60	2	0.1856	0.6797	15.78
90	2.104	0.1892	0.6613	2.483
120	2.025	0.1798	0.6718	1.281
150	2.015	0.179	0.6295	0.8985
180	2.014	0.1979	0.6106	0.7436
210	2.026	0.1899	0.6096	0 .5941