

Supporting Information for

Synthesis and Performance Optimization of Ultrathin Two-Dimensional CoFePt Alloy Materials via in-situ Topotactic Conversion for Hydrogen Evolution Reaction

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Experimental Section

Materials

Nickel foam, cobaltous nitrate ($\text{Co}(\text{NO}_3)_2 \cdot 6\text{H}_2\text{O}$), ferric nitrate ($\text{Fe}(\text{NO}_3)_3 \cdot 9\text{H}_2\text{O}$), urea ($\text{CO}(\text{NH}_2)_2$) and potassium chloroplatinate (K_2PtCl_6) were purchased from Aladdin Industrial Co., Ltd. Sodium hydroxide (NaOH) and ethylene glycol were purchased from Sinopharm Chemical Reagent Co., Ltd.

Material Synthesis

CoFe-LDHs arrays were synthesized by a hydrothermal method in Teflon-lined stainless-steel autoclaves. As a typical preparation processing, Ni foam was cleaned by HCl solution (~37 wt%) through ultrasound bath for 5 min, and then washed by deionized water and absolute ethanol respectively for 10 min to ensure that the nickel oxide layer on the surface was fully cleaned. $\text{Co}(\text{NO}_3)_2 \cdot 6\text{H}_2\text{O}$ (0.65 mmol) and $\text{Fe}(\text{NO}_3)_3 \cdot 9\text{H}_2\text{O}$ (0.2 mmol) and $\text{CO}(\text{NH}_2)_2$ (10 mmol) were dissolved in deionized water (36.0 mL) to form a homogenous solution. The solution and the pretreated Ni foam were transferred to a Teflon-lined stainless-steel autoclave (45-mL), which then maintained at 120°C for 12 hours. Finally, the brown CoFe LDHs precursor on the nickel foam- was washed by deionized water and dried in vacuum at 120°C for 6 hours.

CoFe alloy nanosheets were synthesized through an *in situ* topotactic reduction. Ethylene glycol (40 mL) and NaOH (1.1g) were firstly dissolved in a 45-mL Teflon-lined stainless-steel autoclave at room temperature and the CoFe-LDHs precursor was transferred into it. Then the autoclave was maintained at 160°C for 12 hours to obtain the dark CoFe alloy on the nickel foam. The obtained product was washed by absolute ethanol for two times, named as CoFe-NS. Later, K_2PtCl_6 aqueous solution (0.15mM) was preserved at zero degree, and then CoFe-NS was vertically put in the solution for 20 minutes to synthesize Pt doped CoFe catalyst. The as-synthesized catalyst was

entirely washed by deionized water to remove unreacted K_2PtCl_6 , followed by drying under vacuum for hours, and finally named as CoFePt-1%.

Measurement

Scanning electron microscopy (SEM) images were taken on a Zeiss SUPRA55 scanning electron microscope with accelerating voltages of 200 kV or 50 kV. High-resolution transmission electron microscope (HRTEM) images and EDS elemental mapping were obtained on a JEOL 2100 high-resolution TEM system with operating at 200 kV. The morphologies of samples were also characterized by a FEI G2 20 transmission electron microscopy (TEM). X-ray powder diffraction (XRD) patterns were recorded on an X-ray diffractometer (Bruker) at a scan rate of $10 (^{\circ})/\text{min}$ in the range from 5 to 90° . X-ray photoelectron spectra (XPS) were carried out by using a model of ESCALAB 250 and LabRAM Aramis. Elemental analysis was investigated by an inductively coupled plasma optical emission spectrometry (ICP-OES; Thermo Scientific iCAP 6300).

Electrochemical measurement

The electrochemical measurements were carried out at room temperature in three-electrode glass cell which connected to an electrochemical workstation (Princeton P4000), using saturated calomel electrode (SCE) as the reference electrode, 1 cm^2 Pt plate as the counter electrode and the sample with 1 cm^2 electrode area as the working electrode. Prior to the test measurements, H_2 was bubbled through the electrolyte solution to eliminate the dissolved oxygen and to maintain a fixed Nernst potential for the H^+/H_2 redox couple. Linear sweep voltammetry with a scan rate of 5 mV s^{-1} was conducted in 1 M KOH solution. AC impedance measurements were carried out in the same configuration under the frequency range of 10^{-1} to 10^5 Hz with an AC voltage of 5 mV . The electrochemical double layer capacitance was measured in 1 M KOH at different scan rates from 20 mV s^{-1} to 200 mV s^{-1} in the range of 0.1 V to 0.2 V vs. RHE. As a control sample, 1 mg Pt/C commercial catalyst was loaded on the Ni foam by mixing 2 ml absolute ethanol and $20 \mu\text{l}$ of 5 wt\% Nafion solution. Then the loaded Pt/C catalyst was dried under infrared lamp and then used in HER test.

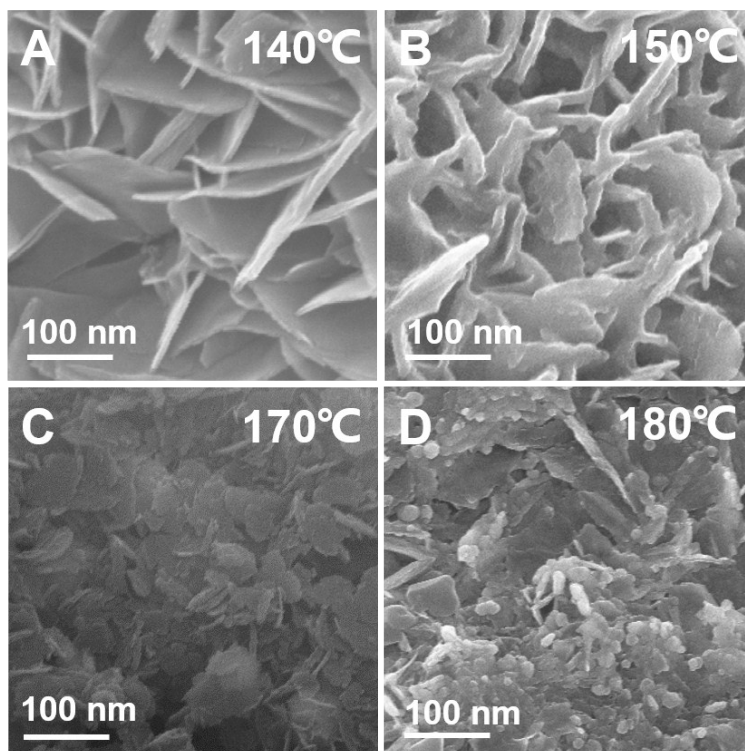


Figure S1 SEM images of CoFe alloy samples prepared at different temperatures.

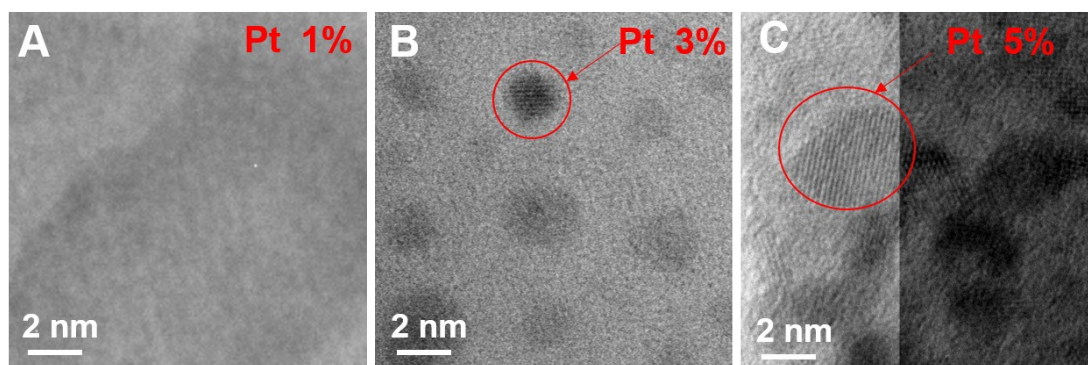


Figure S2 HRTEM images of CoFePt samples with different Pt loading contents.

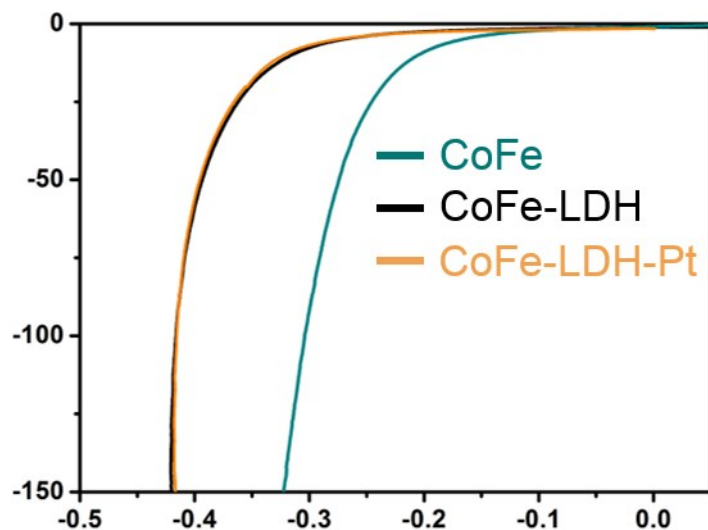


Figure S3 HER polarization curves of CoFe-LDHs and Pt loaded CoFe-LDHs (K_2PtCl_6 solution directly reacted with CoFe-LDHs).

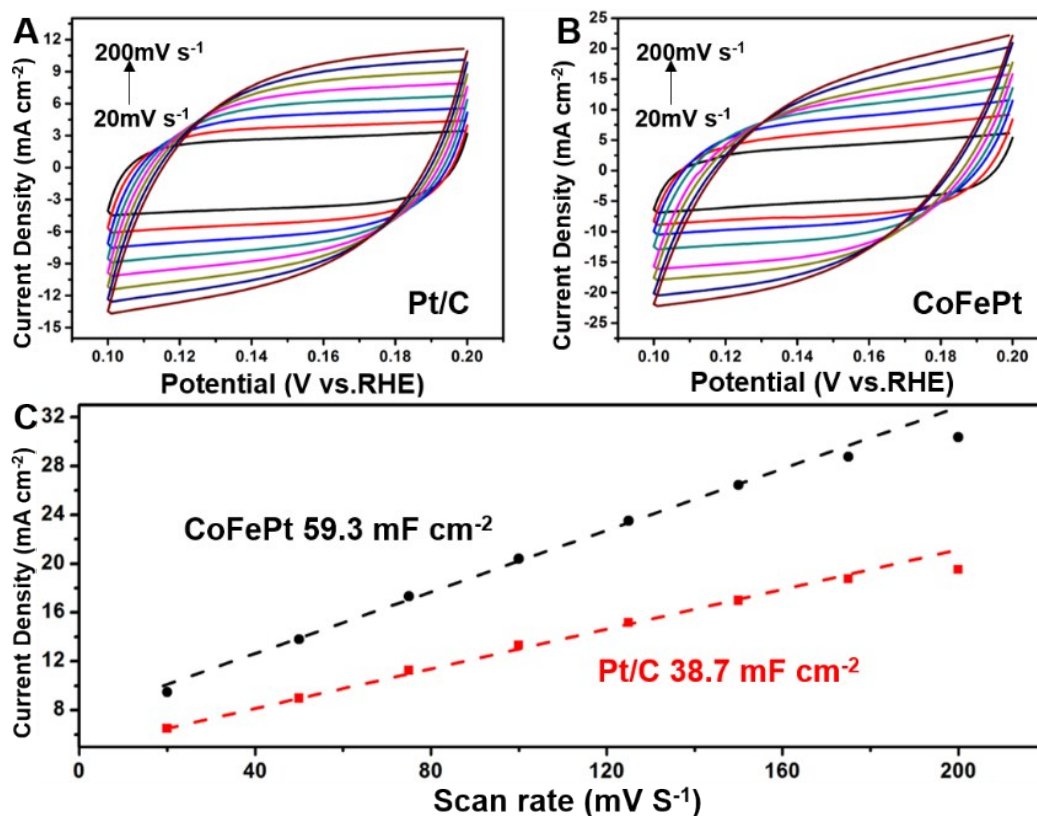


Figure S4 (A) Cyclic voltammetry curves of Pt/C at different scanning rates. (B) Cyclic voltammetry curves of CoFePt-1% at different scanning rates. (C) C_{dl} calculations of CoFePt-1% and Pt/C.

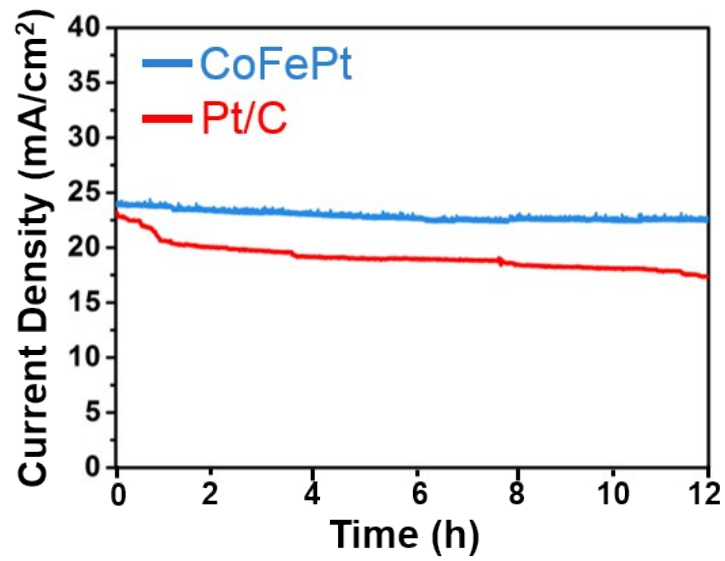


Figure S5 Long-term stability of CoFePt-1% and Pt/C at a constant voltage.

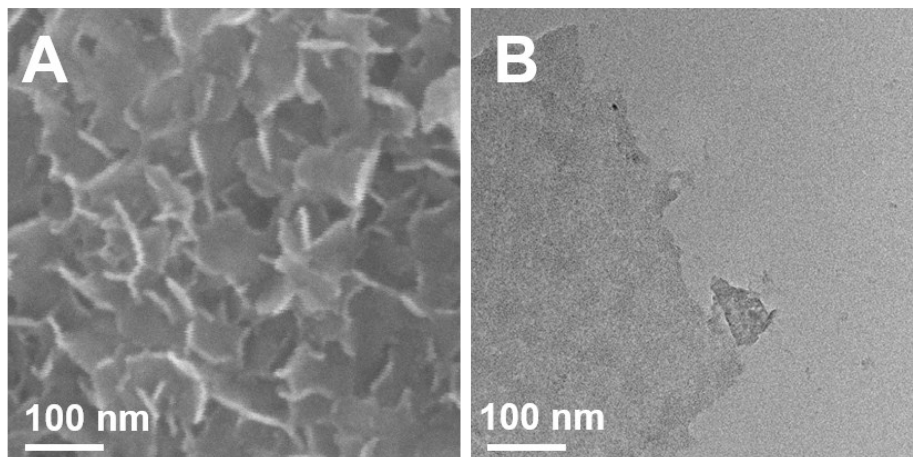


Figure S6 (A) SEM image and (B) HRTEM image of CoFePt-1% after long-term stability test.

Table S1 ICP-OES results of the samples with different Pt contents.

Sample	Co (atom%)	Fe (atom%)	Pt (atom%)
CoFe alloy	76.42	23.58	\
CoFePt-1%	76.19	22.63	1.18
CoFePt-3%	74.71	22.03	3.26
CoFePt-5%	73.04	21.85	5.11

Catalysts	Electrolyte	Overpotential at 10 mA cm ⁻² /mV	Tafel slope /mV dec ⁻¹	Ref.
N-rGO/ CoFe-CoFe ₂ O ₄	1 M KOH	190	100	<i>Carbon</i> 2018 , 139,35
CoFe@N-rGO	1 M KOH	116	78	<i>J. Mater. Chem. A</i> 2018 , 6, 32
NiCo ₂ S ₄ /Ni foam	1 M KOH	65	84.5	<i>Nano Energy</i> , 2016 , 24, 139
Co-Fe-P nanotubes	1 M KOH	86	59	<i>Nano Energy</i> 2019 , 56, 225
CoFe-NC	1 M KOH	202	67.96	<i>J. Power Sources</i> 2018 , 389, 249
Co/Co ₃ O ₄ nanosheets	1 M KOH	105	44	<i>Nano Lett.</i> 2015 , 15, 6015
Co-Fe-P nanosheets	0.5 M H ₂ SO ₄	41	31.5	<i>Appl. Surf. Sci.</i> 2019 , 1, 478
CoP nanosheets	1 M KOH	94	42	<i>Angew. Chem., Int. Ed.</i> 2015 , 127, 6349
CoFePt-1% nanosheets	1 M KOH	18	29	This work

Table S2 Hydrogen evolution reaction performance of CoFePt-1% nanosheets compare to other electrocatalysts.