Supporting Information

Microporous 2D NiCoFe Phosphate Nanosheets Supported on Ni Foam for Efficient overall water splitting in Alkaline Media

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Experimental Section

Chemicals and materials.

The synthesis was carried out by hydrothermal method with commercially available reagents. Ni(acac)₂ (Alfa Aesar), Co(acac)₂ (Alfa Aesar), Fe(acac)₃ (Alfa Aesar), Urea (Sinopharm Chemicals Reagents), urea phosphate (Adamas-Beta), PVP (Sinopharm Chemicals Reagents), and Formamide (Sinopharm Chemicals Reagents).

Preparation of NiCoFe Phosphate Nanosheets

In a typical synthesis of NiCoFe phosphate nanosheets was carried out by hydrothermal method. 10 mg of Ni(acac)₂ (2.59 mM), 10 mg of Co(acac)₂ (2.59 mM), 15 mg of Fe(acac)₃(2.83 mM), 300 mg of urea,100 mg urea phosphate and 200 mg PVP were added in 15 ml Formamide solution with continuous stirring. Then the above homogenous mixture was transferred to Teflon-lined stainless steel autoclave, which was then sealed and kept in the oven at 150 °C for 4 hours. The autoclave was cooled down to room temperature and the black products were then centrifuged with ethanol for three times and dispersed in ethanol solvent for further characterization.

Preparation of porous NiCoFe Phosphate Nanoparticles

For the synthesis of NiCoFe phosphate nanoparticles, all above parameter kept same except the concentration of urea was decreased in the system.

Characterization technique

Transmission electron Microscopy (TEM) and high-resolution transmission electron microscope (HRTEM) were recorded on a HITACHI H-7700 TEM with an accelerating voltage of 100 kV and FEI Tecnai G2 F20 S-Twin high-resolution equipped with energy dispersive spectrometer (EDS) analyses at 200 kV. The sample for HRTEM was prepared by dropping ethanol dispersion on carbon-coated Cu grid. X-ray diffraction (XRD) pattern was obtained with a Bruker D8-advance X-ray powder diffractometer operated at 40 kV and instrument run at a scan rate of 0.02 deg/s in the angle range of 30° to 90° and the wavelength of incident radiation was $\lambda = 1.5418$ Å. XPS measurements were conducted on a scanning X-ray microprobe (Thermo Fisher Model ESCALAB 250Xi) operated at 250 kV, 55 eV with monochromated Al K α radiation. Elemental analysis of the samples was determined by ICP-OES. Fluorescence spectra were recorded on JASCO FP-8300 spectrofluorimeter equipped with a xenon discharge lamp, 1 cm quartz cell.

Electrochemical test

The electrochemical test was carried out by using a CHI650D electrochemical analyzer (CHI instrument, USA). A conventional three-electrode cell was used which include a graphite rod as a counter electrode, a saturated calomel as the reference electrode, and the nickel foam as the working electrode. The carbon-loaded catalyst was dispersed in 0.80 mL of H₂O, 0.20 mL of ethanol and 10 μ L Nafion solution (0.05 wt %) by sonication for more than one hours to form a homogenous catalyst ink. The catalyst ink was transferred to the surface of the nickel foam electrode and dried at room temperature. The electrochemical measurements were performed in 1M KOH at a scan rate of 2 mVs⁻¹. Tafel plot was obtained by taking advantage of Tafel plot function. The time-dependent curve was used to check the electrocatalytic stability of

the material. The electrochemical properties was studied after the activation by 50 CV cycles. The loading amount of catalyst on the nickel foam surface is 0.5 mg/cm^2 .



Figure SI 1. XPS spectrum of NiCoFe phosphate NSs.



Figure SI 2. TEM image of nanosheets by using different amount of urea. (a) 100 mg honey comb shape particle are formed, (b) 200 mg nanoplates are formed, (c) 250 mg nanosheets are formed (d) 300 mg regularly arranged nanosheets are formed, (e) 500 mg nanosheets are formed.



Figure SI 3. XPS spectrum NiCoFe phosphate NSs structure before and after OER. XPS spectra of P 2p binding energies before and after OER stability. The P 2p peak in the original samples, confirming the formation of PO₃ compounds, and no P signal is detected in post-OER samples, suggesting structure changes on the catalyst surface (d). XPS spectra of O 1s binding energies before and after OER stability tests. The original sample have one main components for PO₃ and for adsorbed H₂O, and post-

OER samples show negative shift in binding energy core-level features consisting of FeOOH and nickel, cobalt oxide, meaning that amorphous FeOOH is probably the dominant active site for water oxidation (e). XPS spectra of Fe binding energies before and after OER stability test, it is apparent that the binding energy Fe element is also negatively shifted (c).



Figure SI 4. Cyclic voltammograms (CV) curves of NiCoFe Phosphate NSs (a), and current density as a function of scan rate derived from (b) of NiCoFe Phosphate NSs (b).



Figure SI 5. Time-resolved Photoluminescence (TRPL) spectra of NiCoFe phosphate NSs was did to study the surface defect. The TRPL decay curve can be best fitted by a triexponential decay model indicating that the resultant PL is originating from three distinct channels with the life-times of 1.178 ns (35.42 %), 3.010 ns (49.59 %) and 9.273 ns (14.99 %). The fast PL decay time (1.31 ns) is attributed to the trapping of electrons in the shallow traps followed by their radiative recombination after escaping from these trap states. The second and 3rd time component 3. 010 ns (49.59 %), 9.273 ns (14.99%) can be attributed are due to the presence of defective states on the surface of NiCoFe phosphate NSs. ^[1, 2]

Materials	Electrolyte	Overpotential at 10 (mAcm ⁻²)	Reference
IrO ₂	1 M KOH	320	[3]
FeNiPi/NF	1 М КОН	220	[4]
NiCoFe	1 М КОН	240	This work
phosphate/NF			
CoP/rGO	1 М КОН	280	[5]
СоР	1 М КОН	330 (Overpotential	[6]
		at 30 (mAcm ⁻²))	
СоР	1 M KOH	340	[7]

Table 1. Comparison of OER of different catalyst

Table 2. Comparison of overall water splitting of different catalyst

Materials	Electrolyte	Overpotential at 10 (mAcm ⁻²)/potential	Reference
NiFe LDH	1 M KOH	470	[8]
CoFe LDH	1 M KOH	1.63 V	[9]

NixPx	1 М КОН	1.57 V	[10]
NiCoFe phosphate	1 М КОН	1.52 V	This work
NiFe phosphate/NF	1 М КОН	326	[11]
CoP/rGO	1 М КОН	1.60 V	[12]
Ni ₂ P/Ni/NF	1 М КОН	1.49 V	[13]
СоР	1 М КОН	330 (Overpotential	[6]
		at 30 (mAcm ⁻²))	

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