Electronic Supplementary Material

Alcohol-Alkali Hydrolysis for High-Throughput PET Waste Electroreforming-Assisted Green Hydrogen Generation

Ying Li,[ab] Li Quan Lee,[c] Hu Zhao,[c] Yunxing Zhao,[b] Pingqi Gao,[*bd] and Hong Li[*c]

[a] College of Energy Engineering, Xi'an University of Science and Technology, Xi'an 710054, China

[b] School of Materials, Institute for Solar Energy Systems, Shenzhen Campus of Sun Yat-sen University, No. 66, Gongchang Road, Shenzhen, Guangdong 518107, China,

[c] School of Mechanical and Aerospace Engineering, Nanyang Technological University, 639798, Singapore.

[d] Sun Yat-sen University (Taizhou) Solar energy Joint R&D Center, Taizhou, 225300, China

* Corresponding authors.

E-mail addresses: gaopq3@mail.sysu.edu.cn (P. Gao), ehongli@ntu.edu.sg (H. Li),

1. Experimental

1.1 The synthesis of Fe, Co co-modified Ni_2P nanosheets on Ni foam (FeCo-Ni_2P/NF)

To produce FeCo-Ni_2P/NF nanosheets, 0.1 g cobaltous nitrate hexahydrate ($Co(NO_3)_2·6H_2O$), 0.02 g ferric nitrate nonahydrate ($Fe(NO_3)_3·9H_2O$), 0.5 g ammonium chloride (NH_4Cl) and 5 mL water were mixed to obtain a uniform solution. And then 1*1 cm nickel foam (NF) was immerged into the uniform solution and 1 g urea ($CO(NH_2)_2$) was added. The solution was then heated at 50 ℃ for 10 h, obtaining Fe, Co co-modified layered nickel hydroxide precursor on NF [termed as Fe_1Co_5-$Ni(OH)_2$/NF].

Next, a piece of Fe_1Co_5-$Ni(OH)_2$/NF was placed in one combustion boat and the other combustion boat containing 0.05 g sodium hypophosphite ($NaH_2PO_2·H_2O$) was located at upstream of a tube furnace and then heated at 300 °C for 2 h with a flow of argon. Finally, the Fe_1Co_5-Ni_2P/NF catalyst was obtained after being cooled down to room temperature naturally.

The Fe_1Co_10-Ni_2P/NF, Fe_1Co_2-Ni_2P/NF and Ni_2P/NF were obtained according to similar hydrothermal and phosphating treatment method, expect for adding different amounts of $Co(NO_3)_2·6H_2O$ and $Fe(NO_3)_3·9H_2O$.

1.2 Electrochemical characterizations

The electrooxidation process and HER measurement were performed with in a typical three-electrode system at 30 ± 1 °C. Thereinto, Ag/AgCl electrode acted as reference electrode, carbon rod acted as counter electrode, and the electrocatalyst

supported on NF was used as working electrode. According to the equation $E_{RHE} = E_{Ag/AgCl} + 0.197\ V + 0.0591\ pH$, all electrode potentials have been calibrated as reversible hydrogen electrode (RHE). Accurately measurement indicated that the loading of FeCo-Ni_2P/NF on NF was about 1.2 mg cm^{-2}.

In addition, the Faradaic efficiency (FE) for the PET electrooxidation to formic acid is determined by Equation: FE=(3Fn)/Q, where F is Faraday's constant (96485 C mol^{-1}), Q is the total charge passed across the electrode during electrolysis, and n represents the moles of the formic acid produced.

2. Supplementary Figures

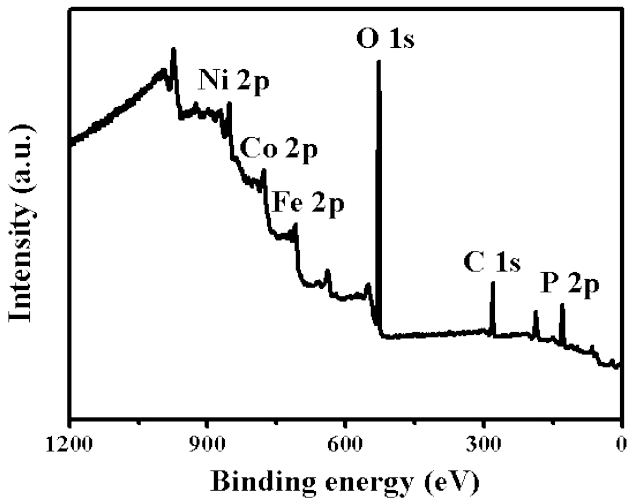

Fig. S1. The XPS spectra of FeCo-Ni_2P/NF.

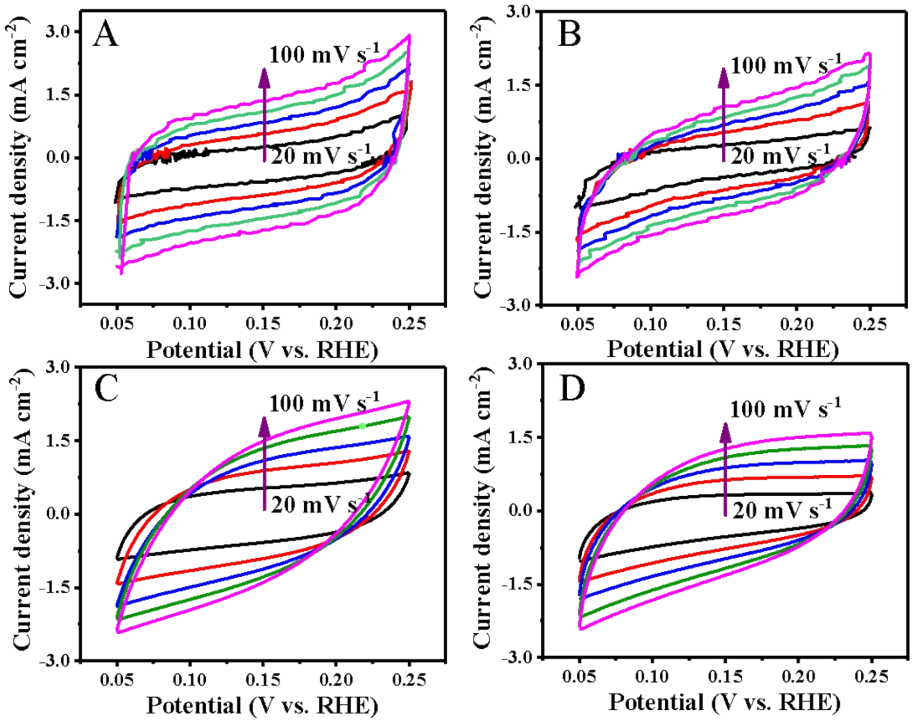

Fig. S2. curves of (A) Fe_1Co_5-Ni_2P/NF, (B) FeCo-$Ni(OH)_2$/NF, (C) Fe_1Co_2-Ni_2P/NF and (D) Fe_1Co_{10}-Ni_2P/NF in 1 M NaOH electrolyte at different scan rates.

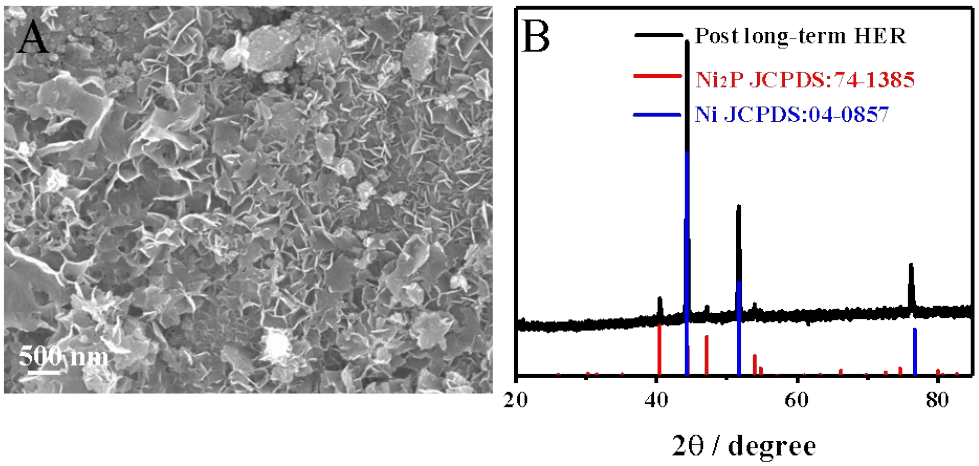

Fig. S3. (A) the SEM image, (B) XRD pattern of the Fe_1Co_5-Ni_2P/NF after a long-term HER reaction.

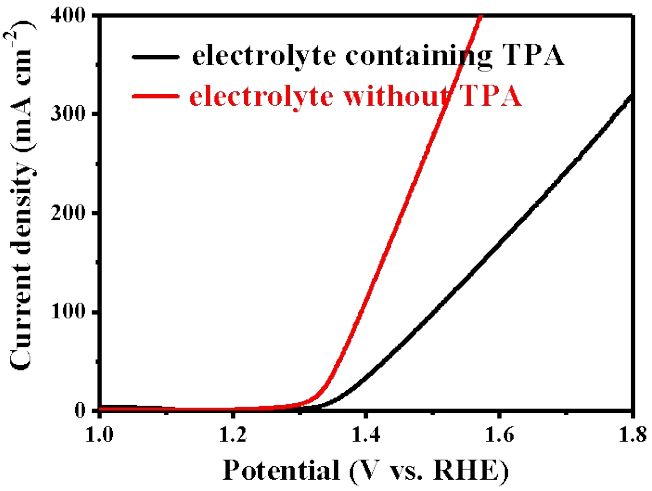

Fig. S4. Electrochemical activity tests of Fe_1Co_5-Ni_2P/NF in electrolytes with and without TPA.

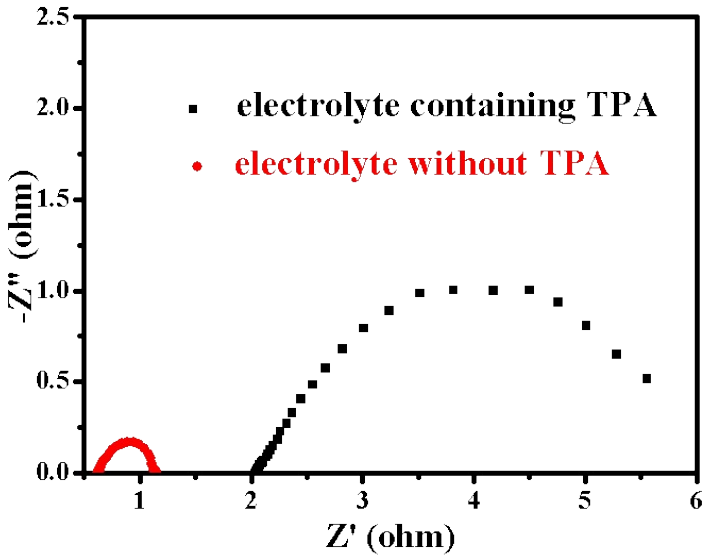

Fig. S5. Electrochemical impedance spectroscopy of Fe_1Co_5-Ni_2P/NF in electrolytes with and without TPA at 1.35 V vs. RHE.

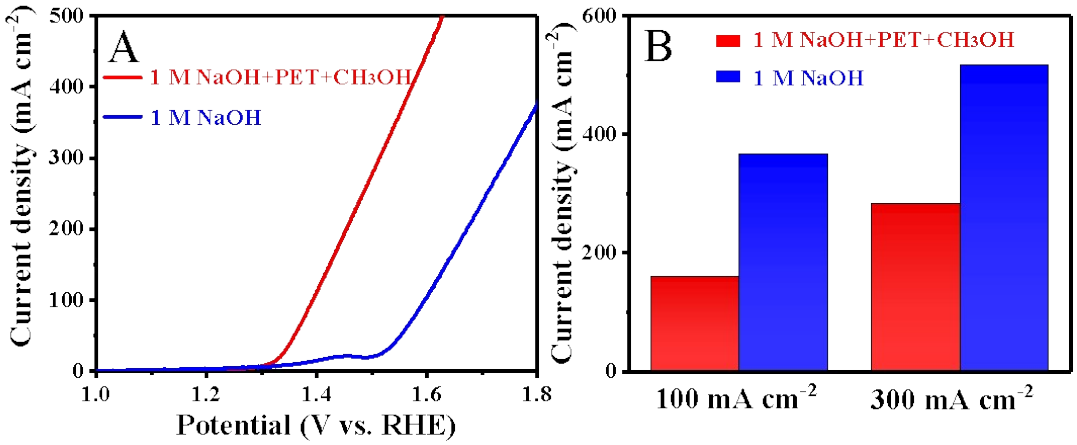

Fig. S6. (A) Electrochemical activity tests of Fe_1Co_5-Ni_2P/NF in different electrolytes. (B) Comparison of the overpotential of the Fe_1Co_5-Ni_2P/NF electrocatalyst at current densities of 100 and 300 mA cm^{-2}.

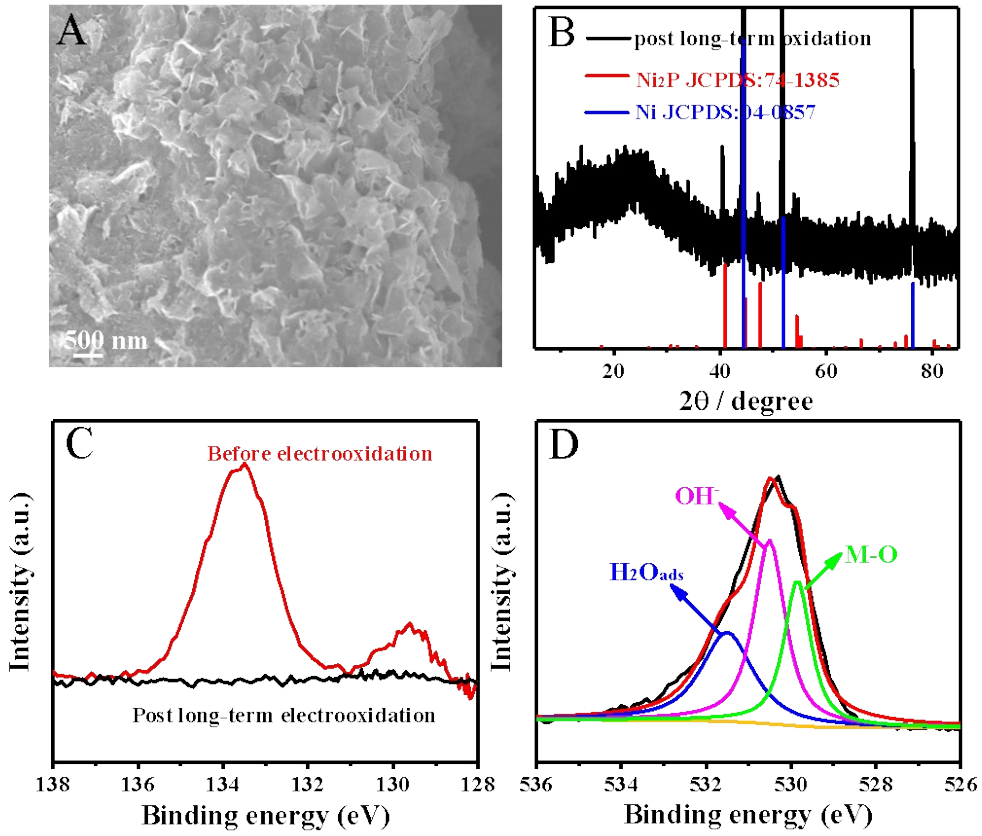

Fig. S7. (A) the SEM image, (B) XRD pattern, high-resolution XPS spectra of (C) P 2p and (D) O 1s peaks of the Fe_1Co_5-Ni_2P/NF post long-term electrooxidation.

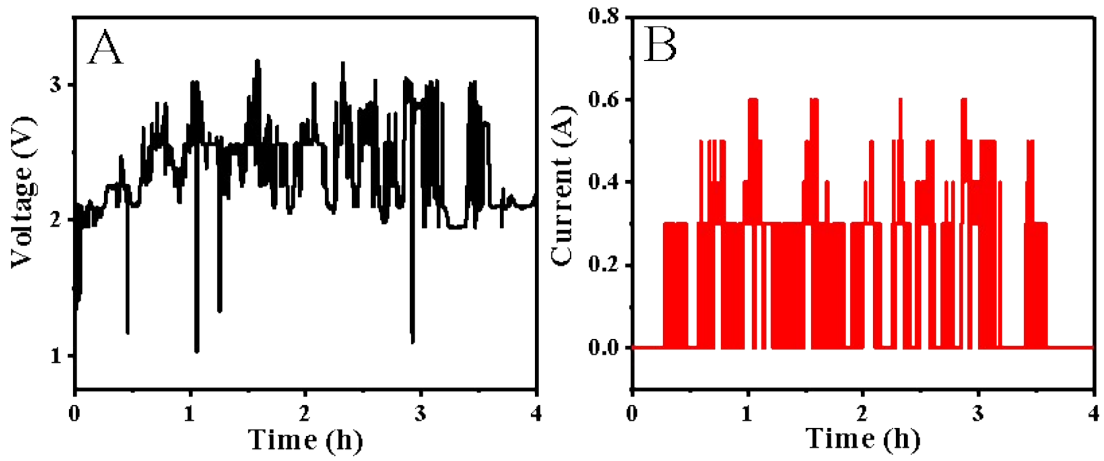

Fig. S8. (A) the voltage curve and (B) the current curve of Fe_1Co_5-Ni_2P/NF||Fe_1Co_5-Ni_2P/NF electrolytic cell under 4 hours photovoltaic drive.

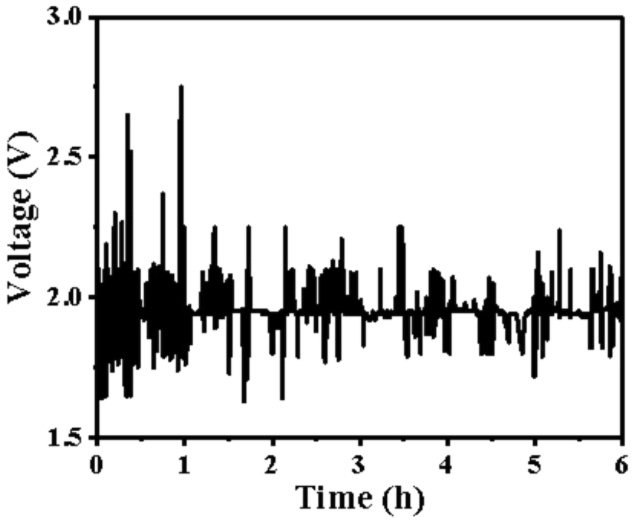

Fig. S9. The voltage curve of Fe_1Co_5-Ni_2P/NF||Fe_1Co_5-Ni_2P/NF electrolytic cell under 6 hours photovoltaic drive.

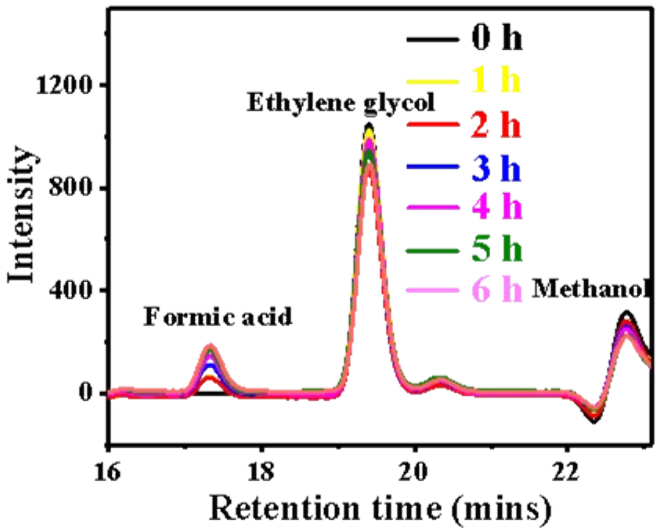

Fig. S10. The liquid chromatogram of the product after exposure to sunlight for different periods of time.