

Supporting information

Electrolyzing spend cupronickel to fabricate the superhydrophilic electrocatalyst enhanced water splitting

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Experimental details

Reagents and chemicals

SCN alloy was purchased from Dongguan Yang Chong metal material Co., Ltd. (Guangdong, China). 98 wt% concentrated H₂SO₄ was purchased from Chongqing Chuandong Chemical (Group) Co., Ltd. (Chongqing, China). 38 wt% concentrated HCl is purchased from Sichuan Xilong Science Co., Ltd. (Sichuan, China). The IF was purchased from Kunshan Guangjiayuan New Material Co., Ltd. (Jiangsu, China). Anhydrous ethanol was purchased from Chengdu Chron Chemicals Co., Ltd. (Sichuan, China).

Material synthesis

Preparation of NiCu/IF, NiMn/IF and CN/IF electrodes

Before the electrosynthesis process, the SCN (40*40*3 mm³) and porous IF (20*20*1 mm³) substrates were subjected to a series of pre-treatments, including mechanical polishing, ultrasonication in 3 M HCl solution, resining by deionized (DI) water and acetone, followed by vacuum drying at 60 °C. As for the preparation of NiCu/IF and NiMn/IF electrodes, a facile electrolysis method was employed using SCN and IF as anode and cathode, respectively, in 0.09 M NiSO₄ + 0.01 M CuSO₄ solution and 0.09 M NiSO₄ + 0.01 M MnSO₄ solution at 40 °C. During the electrolysis process, a current density of 100 mA cm⁻² was kept for 10 min. As a referential sample, Ni/IF and CN/IF electrodes were also prepared in 0.1 M NiSO₄ and 0.1 M H₂SO₄ solution using the above-mentioned processes, respectively.

Fabrication of IF-supported Pt/C and RuO₂

To prepare the RuO₂ and 20 wt% Pt/C electrodes, 40 mg active material (either iridium oxide or Pt on carbon) was uniformly dispersed in a mixture solution containing 90 μL Nafion solution, 810 μL ethanol and 600 μL DI water with the assistance of

vigorous stirring and sonication. A piece of cleaned IF electrode was then soaked in the solution for 1 h to let the active material coat the surface, followed by drying in air. ¹

Chemical and structural characterization

The phase compositions, chemical constitution, surface microstructure, element composition and chemical state of the synthetic composite electrode NiCu/IF were surveyed by X-ray diffraction (XRD, D/Max 2200) technology, Thermo Fischer DXR, field emission scanning electron microscopy (FE-SEM, Nova Nano SEM 450) and X-ray photoelectron spectroscopy (XPS, PHI 5500). Transmission Electron Microscope (TEM) measurements were executed by the JEM-2100 instrument.

Electrochemical measurements

The electrochemical measurements were performed by CHI 760E electrochemical station using a standard three-electrode system where a graphite rod, Ag/AgCl electrode, the as-prepared electrocatalyst were used as the counter electrode, reference electrode and working electrode, respectively. 1M KOH aqueous solution (pH=14) was used as electrolyte for all the tests unless otherwise specified. After 20 cyclic voltammetry (CV) cycles, the linear sweep voltammetry curve (LSV) of the activated sample was recorded without iR compensation at a scan rate of 5 mV s⁻¹. The recorded potential ($E_{\text{Ag/AgCl}}$) was calibrated to the reversible hydrogen electrode (RHE) using the Nernst equation: $E(\text{RHE}) = E(\text{Ag/Cl}) + 0.197 + 0.0591 \times 14$. Electrochemical impedance spectroscopy (EIS) was carried out at the onset potential from 100 kHz to 0.01 Hz and the transfer resistance (R_{ct}) values are calculated based on the equivalent circuit. To estimate the electrochemically active surface area (ECSA), CV cycling was performed at different scan rates. For the durability test, the i-t measurement at 100 mA cm⁻² were conducted for 120 hours. 10000 times CV test was performed for OER and

HER. Overall water splitting was carried out in an ordinary electrolyzer with NiCu/IF as the cathodic electrode and NiMn/IF as the anodic electrode.

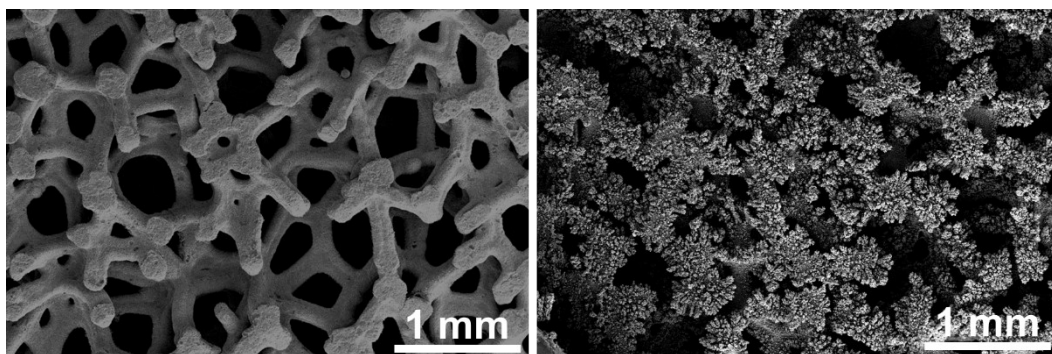


Fig. S1. SEM image of (a) IF and (b) NiMn/IF.

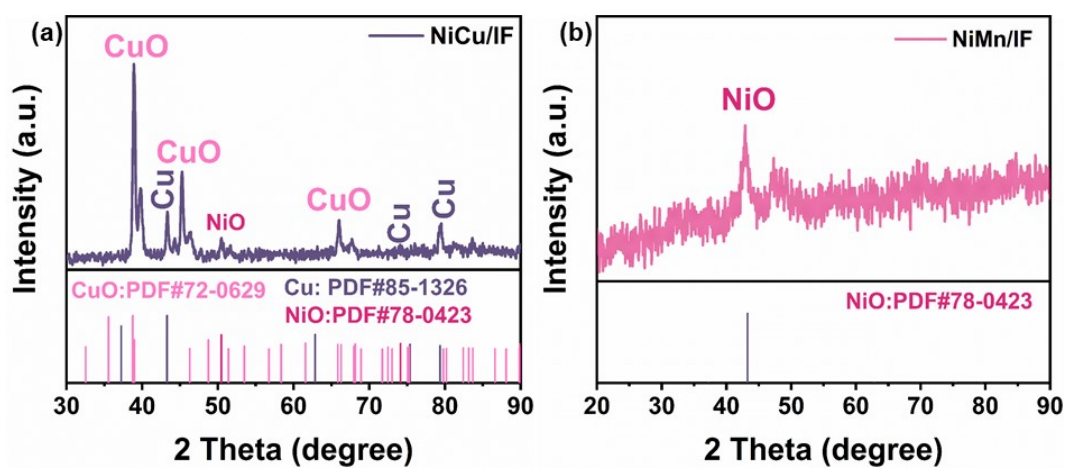


Fig. S2 High-Resolution Thin-Film X-Ray diffraction pattern of (a) NiCu/IF and (b) NiMn/IF.

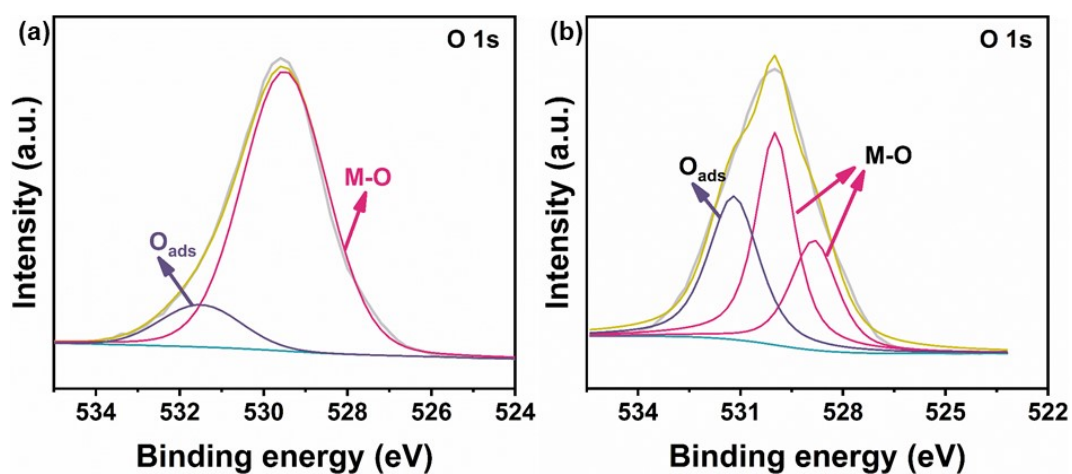


Fig. S3 O 1s XPS of (a) NiCu/IF and (b) NiMn/IF.

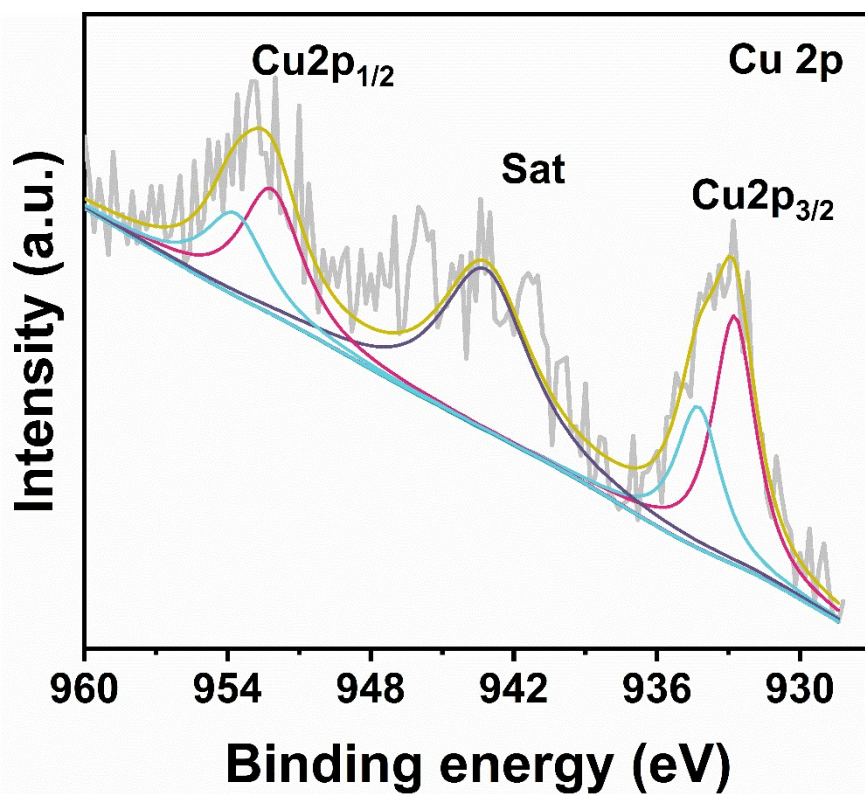


Fig. S4 Cu 2p XPS spectrum of NiMn/IF.

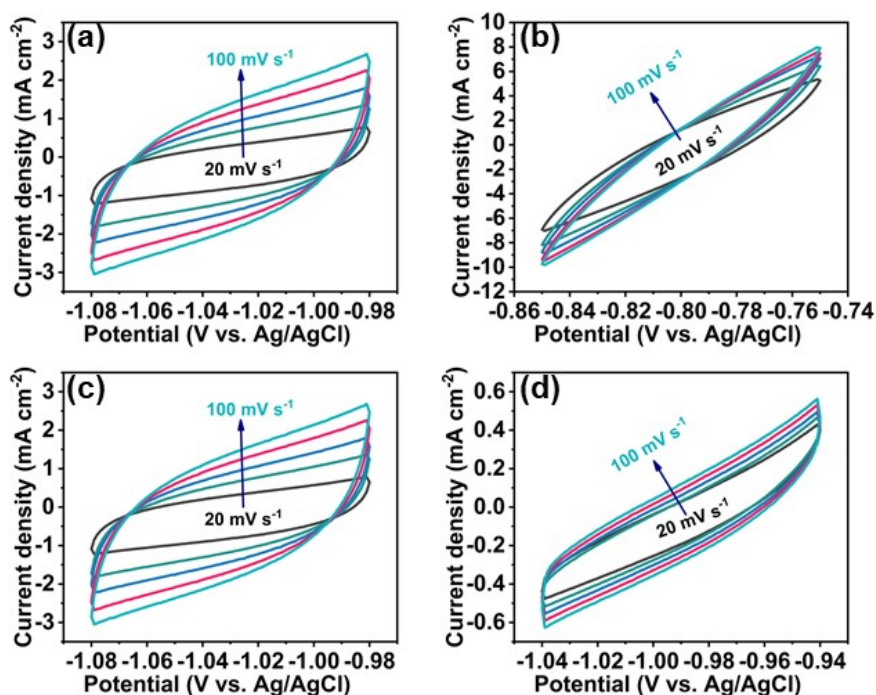


Fig. S5. CV recorded in a non-faraday region of NiCu/IF (a), Ni/IF(b), bare IF(c), and CN/IF(d).

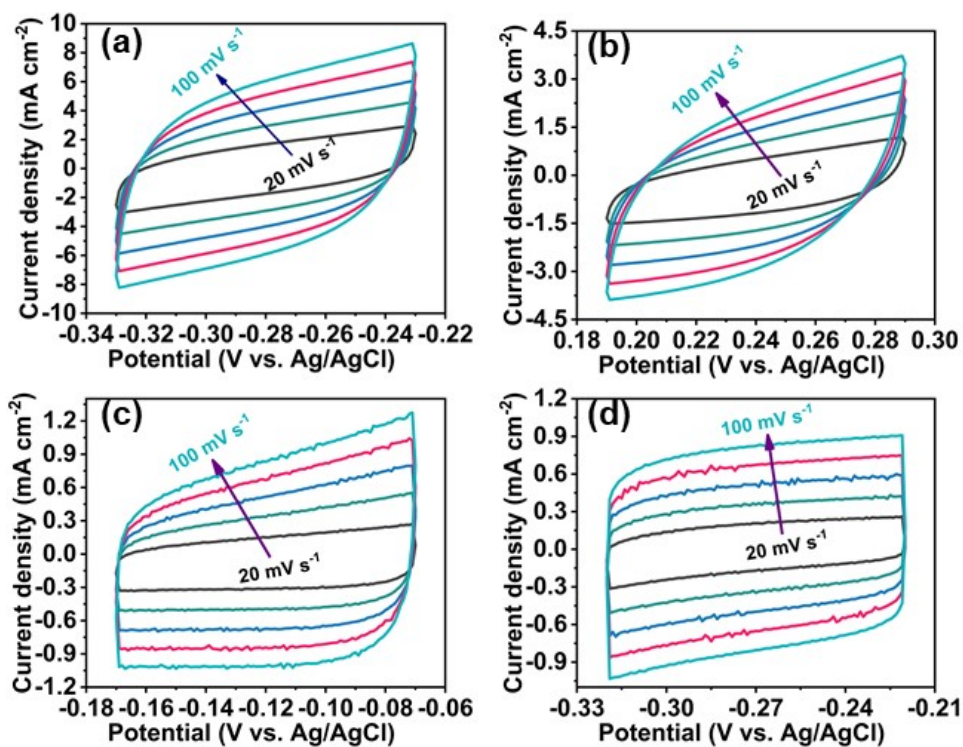


Fig. S6. CV recorded in a non-faraday region of NiMn/IF (a), Ni/IF (b), CN/IF (c), and bare IF (d).

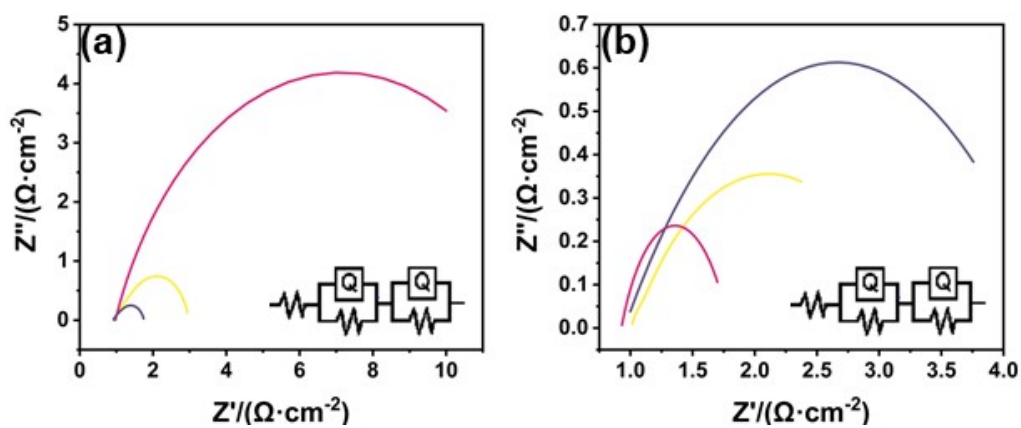


Fig. S7. Electrochemical impedance spectra of NiCu/IF (a) and NiMn/IF (b) in 1.0 M KOH at various potentials.

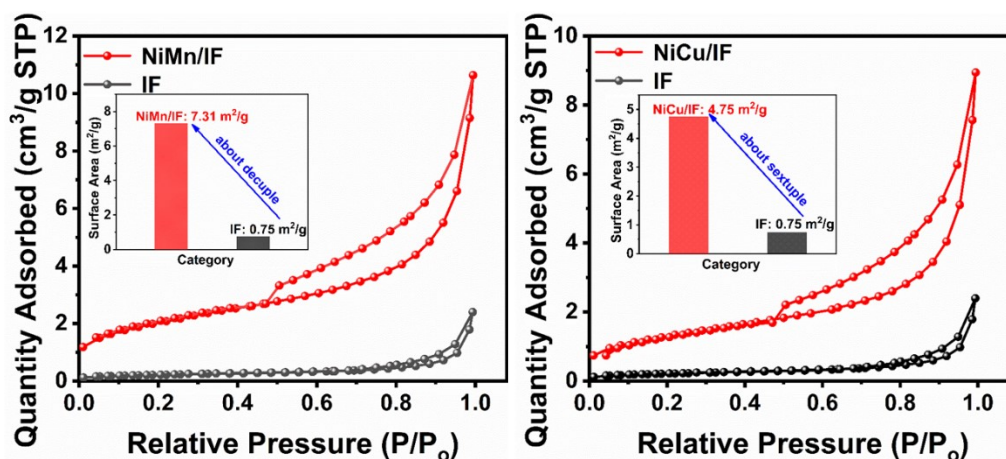


Fig. S8. N₂ adsorption–desorption isotherms and BET surface area of NiMn/IF, NiCu/IF and IF.

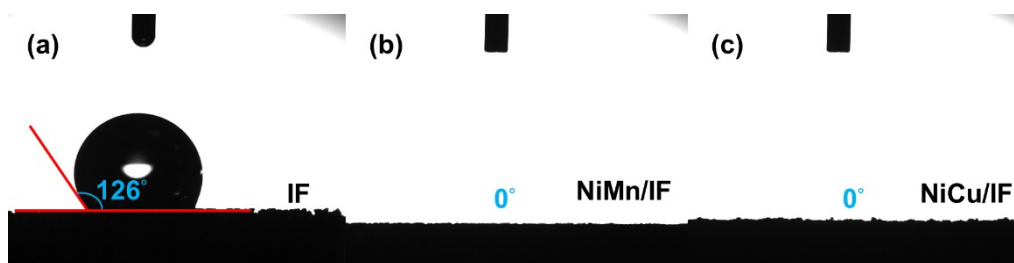


Fig. S9. Water contact angles of IF (a), NiMn/IF (b) and NiCu/IF (c).

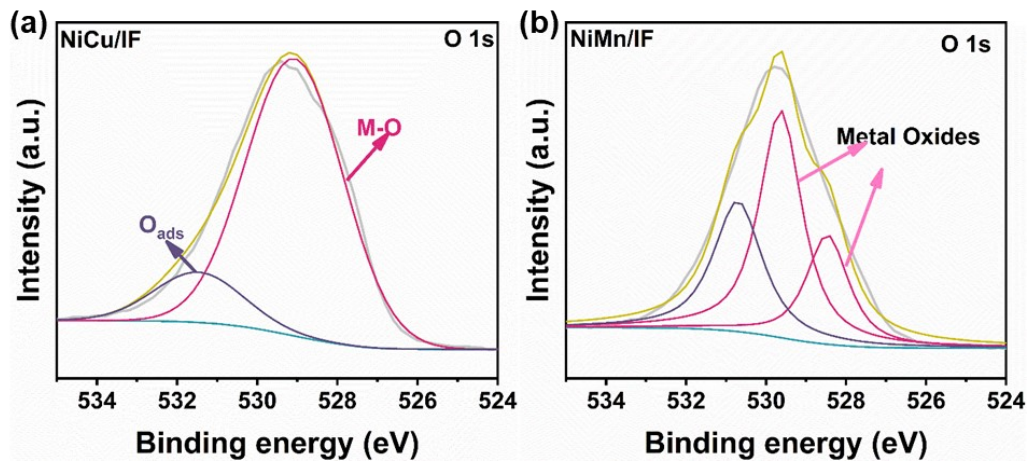


Fig. S10 O 1s XPS of (a) NiCu/IF and (b) NiMn/IF after i-t test.

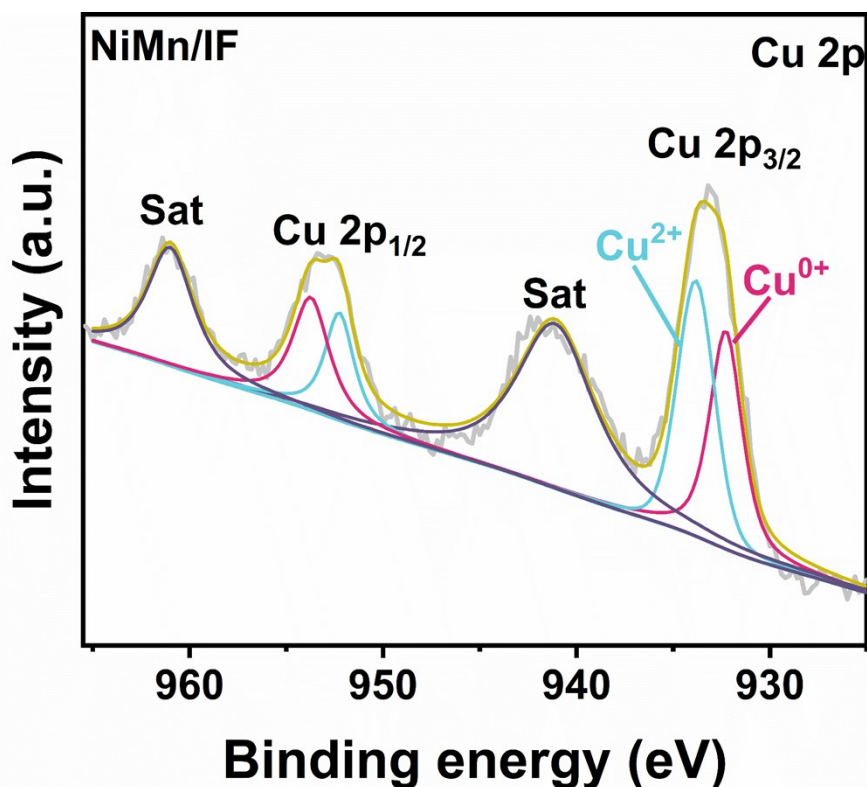


Fig. S11 Cu 2p XPS of NiMn/IF after i-t test.

Table S1. Overall water splitting performances of representative electrocatalysts in alkaline electrolyte

Electrocatalysts	Electrolyte	cell voltage(V) at 10 mA cm ⁻²	Ref.
NiMn/IF(+) and NiCu/IF(-)	1.0 M KOH	1.48 @ 10 mA cm⁻²	This work
CoFeP@C	1.0 M KOH	1.55	2
NiFeOOH	1.0 M KOH	1.23+0.36	3
CoN _x @GDY NS/NF	1.0 M KOH	1.48	4
1T-Fe/P-WS ₂ @CC	1.0 M KOH	1.53	5
NiFe-LDH-Vo@NiCu	1.0 M KOH	1.54	6
F-CoP NAs/CF	1.0 M KOH	1.48	7
V-CoP@a-CeO ₂	1.0 M KOH	1.56	8
Co@V2CTx	1.0 M KOH	1.60	9
POM@ZnCoS/NF	1.0 M KOH	1.56	10

meso-Fe-MoS ₂ /CoMo ₂ S ₄	1.0 M KOH	1.62	11
CoFeZr oxides/NF	1.0 M KOH	1.63	12
CoP	1.0 M KOH	1.65	13
Ni ₃ N/Ni _{0.2} Mo _{0.8} N	1.0 M KOH	1.487	14
CoP/NCNHP	1.0 M KOH	1.64	15
CoFe PBA@CoP	1.0 M KOH	1.542	16
CoP/CeO ₂ -FeOxH	1.0 M KOH	1.53	17

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