

Supporting materials

Experimental Section

Experimental Reagents

Ti₂AlC (98%, 400 mesh) was purchased from Guangdong Foshan Xinxu Technology. Lithium fluoride (LiF, 99%), urea (CO(NH₂)₂, 99%), and potassium hydroxide (KOH, 99.99%) were purchased from Aladdin. Hydrogen chloride (HCl, 36wt%-38wt%), nickel nitrate hexahydrate (Ni(NO₃)₂ · 6H₂O, 98%), and ferric nitrate nonahydrate (Fe(NO₃)₃ · 9H₂O, 98%) were also purchased from Aladdin. Shanghai Guoyao Company provided Sodium chloride (NaCl, 99%), Sodium hypophosphite (NaH₂PO₂, 99%), Ethanol (C₂H₆O, 95%), Sodium Hydroxide (NaOH, 97%), and Hexadecyl trimethyl ammonium Bromide (CTAB, 99%). Yonghua Chemical Reagents supplied Ammonium fluoride (NH₄F, 96%) and Sodium borohydride (NaBH₄, 97%).

Preparation of single-layer Ti₂C MXene

To prepare single-layer Ti₂C MXene, 1.1g LiF and 20 mL 9M HCl are added to a 100 ml polytetrafluoroethylene reaction flask. Next, 1g of Ti₂AlC is slowly added to the flask in small portions over 10 minutes with the rotation speed adjusted to 500 rpm. The mixture is then reacted at 30° C for 2 hours. After the reaction, the product is poured into a 50 mL centrifuge tube and centrifuged at 5000 rpm for 5 minutes to remove the upper liquid layer. 30 ml of 2M HCl was added to the precipitate for a 2-hour reaction. After the reaction, the slurry was poured into a centrifuge tube and washed several times at 5000 rpm for 5 minutes until the upper liquid turned black and the bottom precipitate expanded. The slurry was then transferred to a gas-washing bottle, and after purging with N₂ for five minutes, the inlet and outlet were closed. The mixture undergoes ice bath sonication for 1.5 hours, followed by centrifugation at 2500 rpm for 0.5 hours. The upper liquid is collected and subjected to freeze-drying to obtain a single-layer of Ti₂C MXene.

Preparation of Ti₂C@NF

To prepare NF@MXene, foam nickel (1 × 5 cm²) was sequentially washed with 3M HCl, deionized water, and anhydrous ethanol for 15 minutes each using ultrasonic cleaning. The cleaned and dried foam nickel was then immersed in 50 mL of 2 mg mL⁻¹ CTAB for 30 minutes, followed by immersion in 50 mL of 1 mg mL⁻¹ single-layer Ti₂C MXene solution for 30 minutes. After immersion, the foam nickel was removed and dried at 60 °C in a vacuum oven for 12 hours to obtain NF@MXene.

Preparation of NiFe/Ti₂C@NF

A solution is prepared by mixing 1.2 mmol of Ni(NO₃)₂ · 6H₂O, 0.6 mmol of Fe(NO₃)₃ · 9H₂O, 3 mmol of NH₄F, and 12 mmol of urea in 60 mL of deionized water with stirring for 30 minutes. The solution is then poured into a 100 mL Teflon reactor, and 2 pieces of the NF@MXene are placed in the solution. The reactor is then placed in an oven at 100 °C for 7 hours. The product is washed repeatedly with deionized water and ethanol and then dried overnight at 60 °C in a vacuum oven to obtain NiFe/Ti₂C@NF.

Preparation of NiFeP/Ti₂C@NF

A piece of NiFe/Ti₂C @NF is immersed in a homogeneous solution prepared by stirring 0.1 mol NaBH₄ and 1 mmol NaOH for 30 minutes, followed by soaking for another 30 minutes. The sample is then dried overnight in a freeze dryer. Subsequently, the dried sample is placed in a ceramic boat downstream in a tube furnace, with another ceramic boat containing 10 mmol NaH₂PO₂ placed upstream. The system is then annealed at 350 °C under N₂ atmosphere for 2 hours to obtain NiFeP/Ti₂C@NF. In the same process, Ti₂C @NF is replaced with bare NF to obtain NiFe@NF and NiFeP@NF samples.

2.6 Material Characterization

The crystal phase of the materials is analysed qualitatively using X-ray powder diffraction (XRD) with a D/MAX2500 instrument, Cu K α radiation, and a scanning

range of 5-90° at 40 kV and 40 mA. The surface morphology of the catalysts is characterised using field emission scanning electron microscopy (FESEM) with a SUPRA-55 instrument from Zeiss. The phase, crystal structure, and grain size of the materials are characterised using transmission electron microscopy (TEM) with a JEM-2100F instrument from JEOL. Component identification and analysis are performed using X-ray photoelectron spectroscopy (XPS) with the ESCALAB 250XI from Thermo. The obtained spectra are calibrated using the C1s peak line at 284.8 eV, and the data are analysed using software.

Electrochemical Testing

The catalysts underwent electrochemical characterization using a CHI 760E electrochemical workstation in a three-electrode system at room temperature in 1 M KOH (pH = 14). The working electrode was a self-supporting electrode synthesized through the above steps, with a test area of $1 \times 1 \text{ cm}^2$. The counter electrode was a graphite rod, and the reference electrode was an Hg/HgO (saturated KCl) electrode. The equation is $E (\text{vs. RHE}) = E (\text{vs. Hg/HgO}) + 0.059 \text{ pH} + 0.098$, where pH = 14. To activate the catalyst and remove bubble effects, cyclic voltammetry (CV) is performed initially. This is done in the range of 0-1.5V at a scan rate of 50mV/s for 20 cycles until repeatable curves are observed. The Nernst equation is used to calculate the electrode potential (E) of a half-cell. Subsequently, linear sweep voltammetry (LSV) is conducted at a low scan rate of 1 mV/s in the same range to test the electrochemical performance of the samples. Tafel curves are fitted based on the LSV curves, using the Tafel equation: $\eta = b \times \log j + a$, where η , a, b, and j represent overpotential, Tafel constant, Tafel slope, and current density, respectively. The double-layer capacitance of the catalyst is determined by cyclic voltammetry in the non-Faradaic region at different scan rates (20~140mv/s) after 20 cycles, followed by linear fitting. Electrochemical impedance spectroscopy (EIS) testing was conducted using a PARSTAT3000 electrochemical workstation with a frequency range of 100 K to 0.1 Hz and an applied AC voltage of 5 mV. The multiple-step chronopotentiometry (CP) method was employed to investigate the variation of overpotential at different

current densities and to compare the deviation of linear sweep voltammetry (LSV) before and after 5000 cycles of cyclic voltammetry (CV) testing. The study investigates the catalyst's long-term stability at a current density of 50mA/cm for 20 hours using current-time curves. All electrochemical data, except for stability testing, are adjusted for 85% iR automatic compensation.

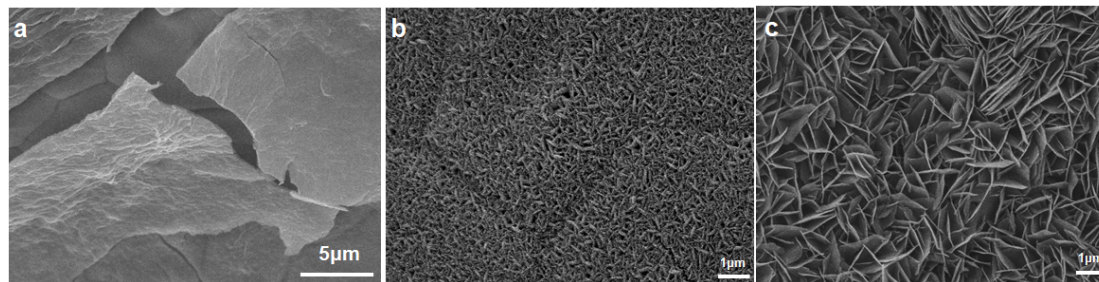


Figure S1 FSEM images of (a) $\text{Ti}_2\text{C@NF}$, (b) NiFe@NF and (c) NiFeP@NF .

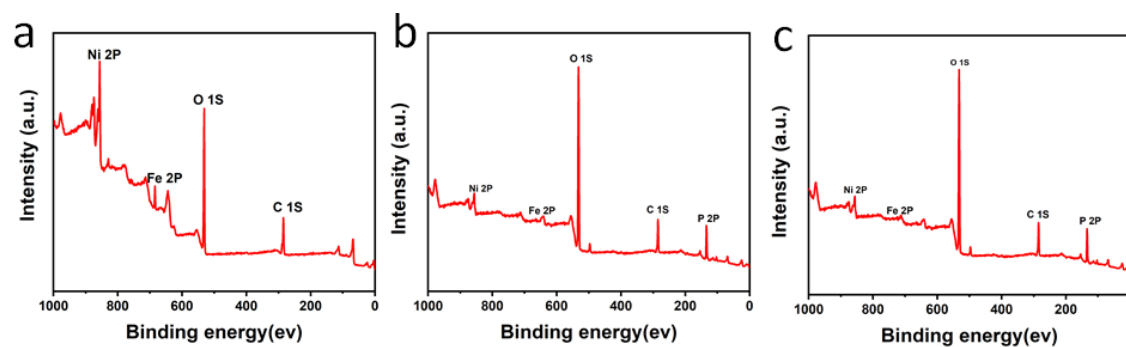


Figure S2. XPS full spectrum of (a) NiFe@NF , (b) NiFeP@NF , and (c) $\text{NiFeP/Ti}_2\text{C@NF}$ samples.

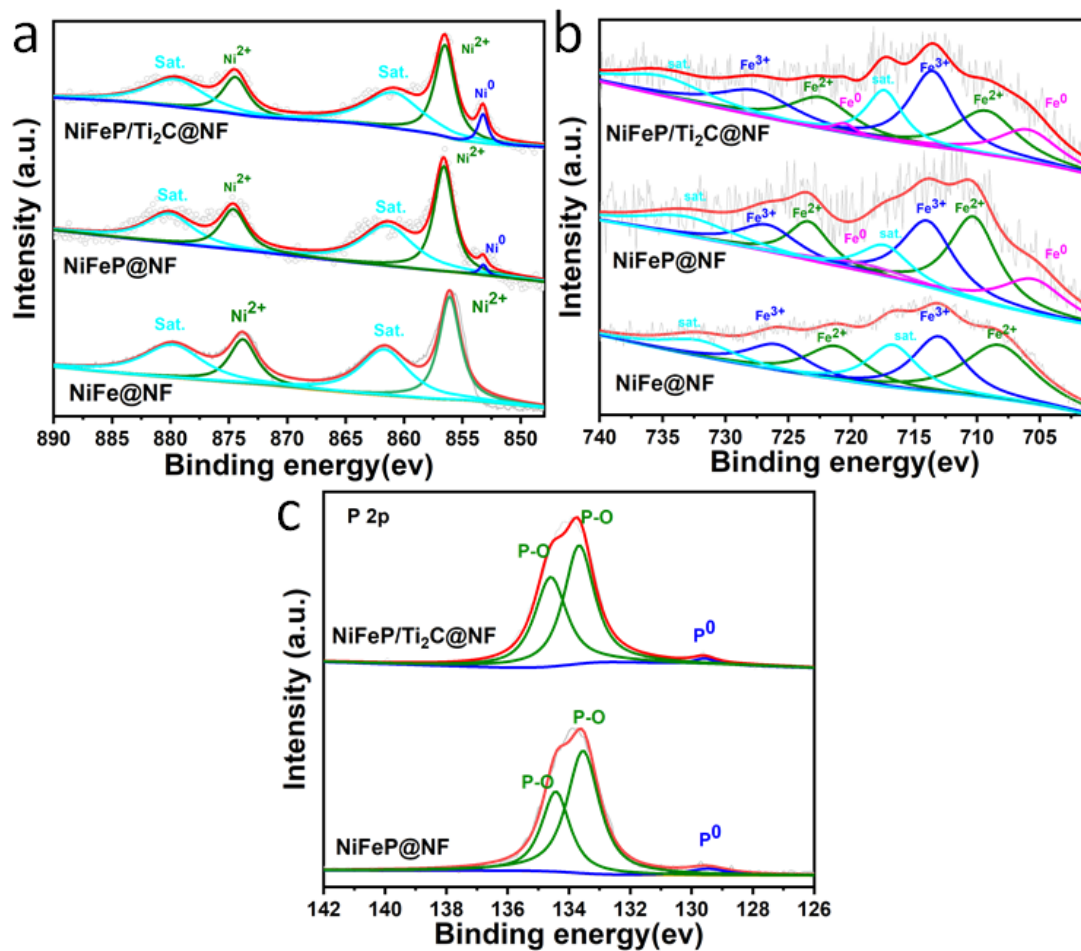


Fig. S3 (a) Ni 2p, (b) Fe 2p and (c) P 2p XPS spectra for as-prepared samples.

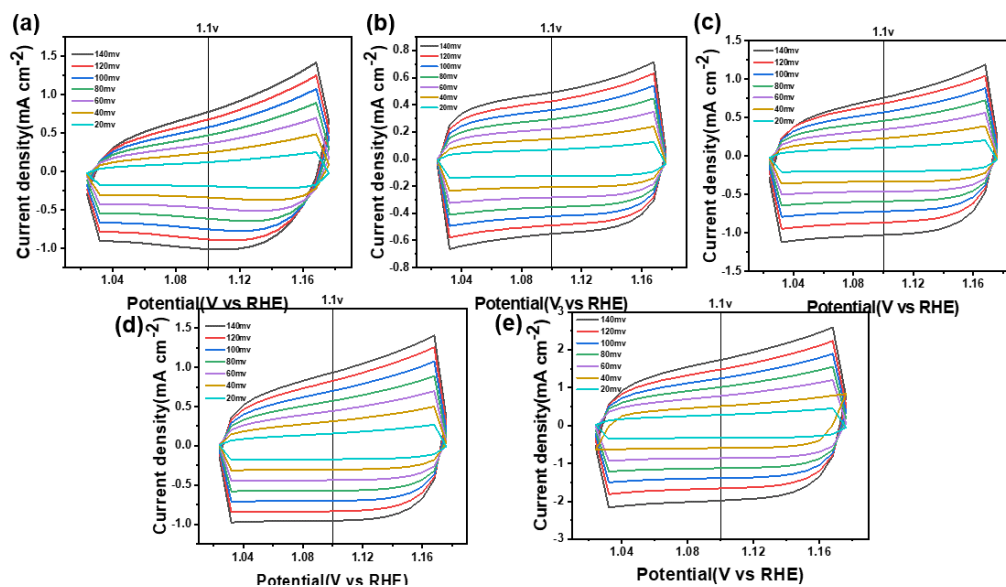


Figure S4. (a) CV of NF, (b) CV of NiFe@NF, (c) CV of NiFe/Ti₂C@NF, (d) CV of NiFeP@NF, and (e) CV of NiFeP/Ti₂C@NF at various scan rates.

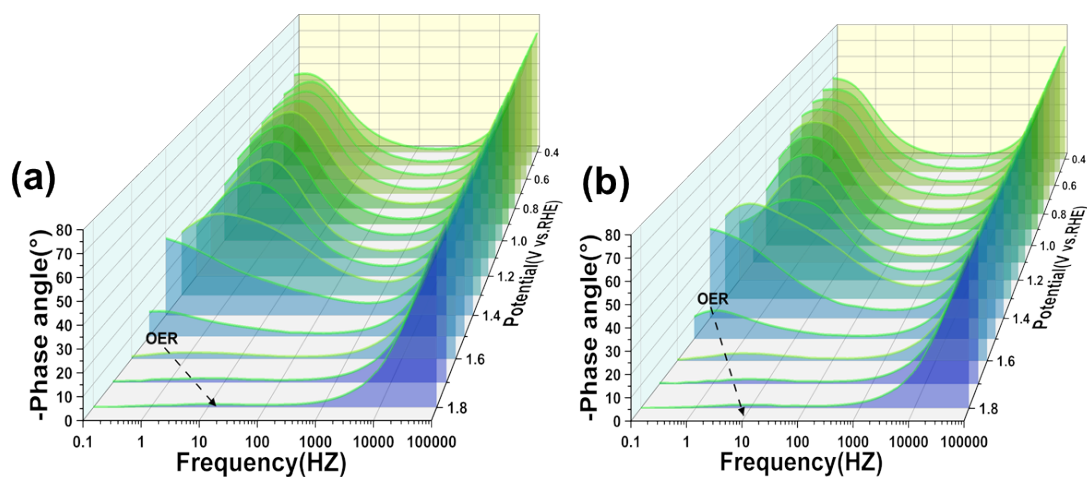


Fig. S5 Bode plots for (a) NiFeP@NF and (b) NiFeP/Ti₂C@NF samples.

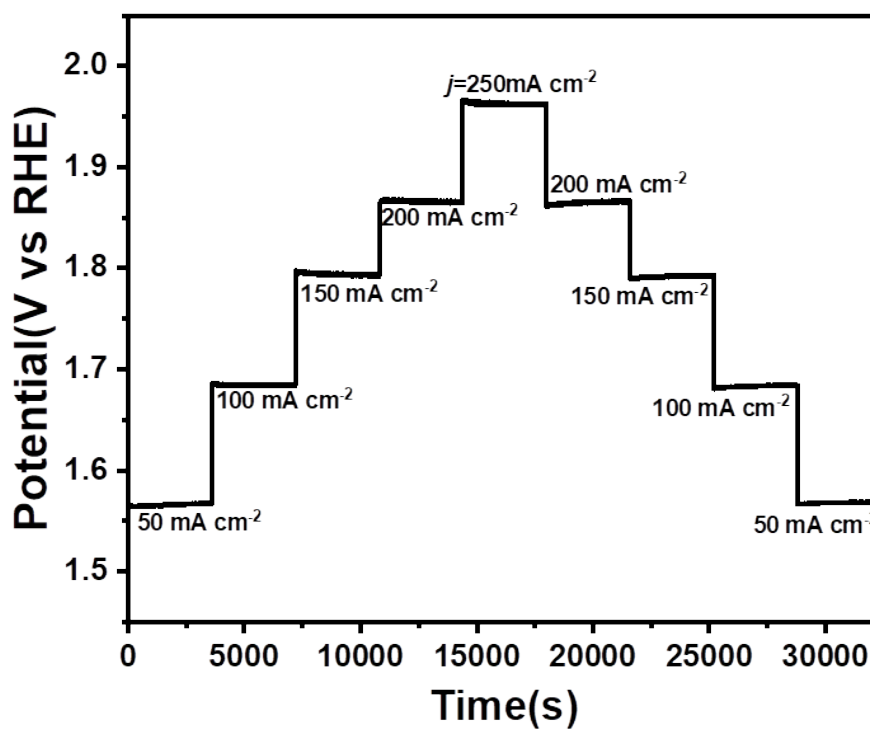


Fig. S6 The stability at different current density for NiFeP/Ti₂C@NF sample.

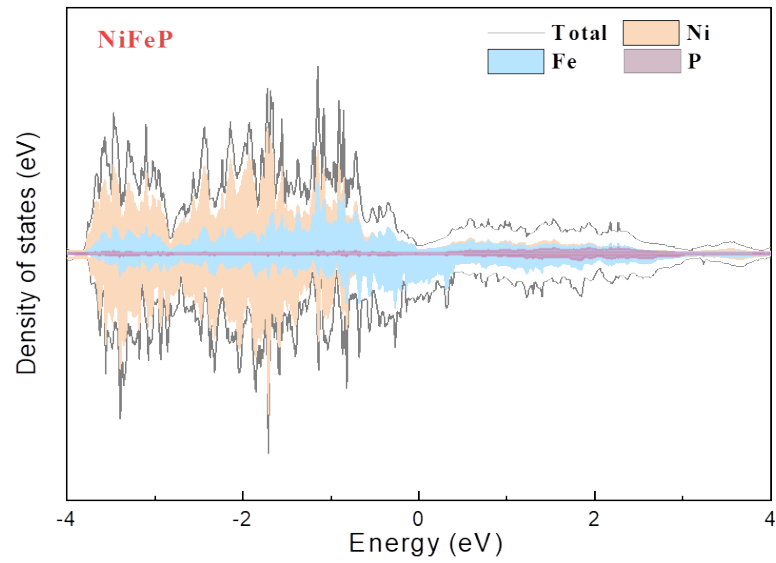


Fig. S7 The TDOS and PDOS of NiFeP.

Table S1. Specific values of EIS for different catalysts in full water electrolysis.

Catalysts	R_s (Ω)	Q	n	R_{ct} (Ω)
NF	1.969	0.0091	0.77	12.12
NiFe@NF	2.680	0.1372	0.8	1.43
NiFe/Ti ₂ C@NF	2.055	0.1194	0.63	1.28
NiFeP@NF	2.237	0.2897	0.8	1.0736
NiFeP/Ti ₂ C@NF	1.637	0.4079	0.64	0.7574

