

Supporting Information

Surface Nitriding to Improve the Catalytic Performance of FeNi₃ for Oxygen Evolution Reaction

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Experimental

Synthesis

The FeNi₃ precursor was synthesized by hydrothermal method. First, add (0.2703g) FeCl₃ 6H₂O, (0.7138g) NiCl₂ 6H₂O, (0.6362 g) urea and (0.9 g) ammonium fluoride into 80 mL of deionized water, stir until totally dissolved, then transfer the mixed solution to 100 mL Incubated at 180°C for 12 hours in a Teflon-lined stainless steel autoclave. The prepared precursors were washed three times with absolute ethanol and ultrapure water, respectively, and treated at 60°C overnight. Finally, (100 mg) of the FENI3 precursor was incubated in a tube furnace at 450 °C for 3 hours under a hydrogen environment to prepare FeNi₃. Furthermore, N-450, N-500 and N-525 samples were prepared by incubating the FeNi₃ precursor at 450, 500 and 525 °C for 3 hours in an ammonia atmosphere, respectively^{1,2}.

Characterization

Scanning electron microscopy (SEM; TESCAN MIRA3), transmission electron microscopy (TEM; Tecnai G2 F30, FEI) and EDX embedded in the SEM were utilized to observe the microscopic morphology of the samples and the elemental map of the catalyst. X-ray diffraction (X'pert Pro Philips) and Raman spectrometer (LabRAM HR Evolution) in the wavenumber of 100-900 cm⁻¹ was used to analyze the crystal structure of the catalyst and the elemental valence states of the samples analyzed by XPS (Kratos AXIS Ultra). Finally, the catalytic performance of the samples was tracked by the electrochemical workstation (CHI 660E).

Electrochemical measurements

The catalytic performance of the samples was tested at room temperature in an alkaline solution with a three-electrode system which were reference electrode (Ag/AgCl electrode), the counter electrode (platinum foil) and as the working

electrode (glassy carbon electrode). Electrode solution preparation is divided into two steps. First, 10 mg of carbon and 10 mg of catalyst were dispersed in 40 mL of petroleum ether for long-term ultrasonic treatment, and then 6 mg of the above dried sample was ultrasonically treated in 30 μL of Nafion and 1470 μL of DMF to synthesize electrode fluid. Finally, 9 μL of the slurry was loaded onto the glassy carbon electrode in three portions. The potential was calibrated to RHE through Nernst Eq: $E_{(\text{RHE})} = E_{\text{Ag/AgCl}} + 0.197 + 0.059 \cdot \text{pH}$. Electrochemical impedance spectroscopy (EIS) was evaluated at a potential bias of 1.48 V against the RHE with a frequency range of 10 kHz to 0.01 Hz. In addition, the overpotential was calculated by the difference between the potential and 1.23V.^{3,4}

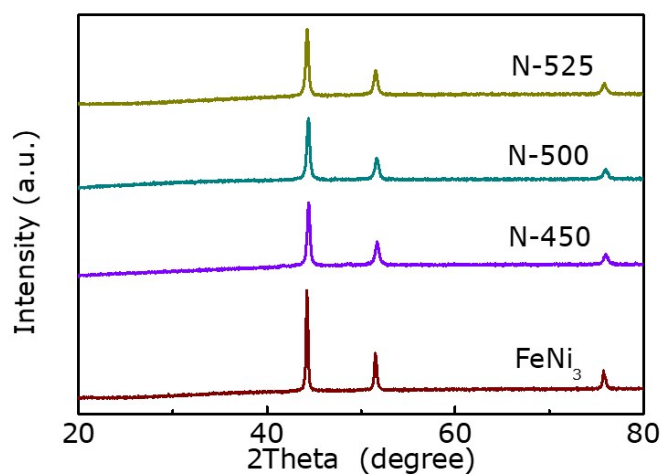


Figure S1. XRD patterns of all samples.

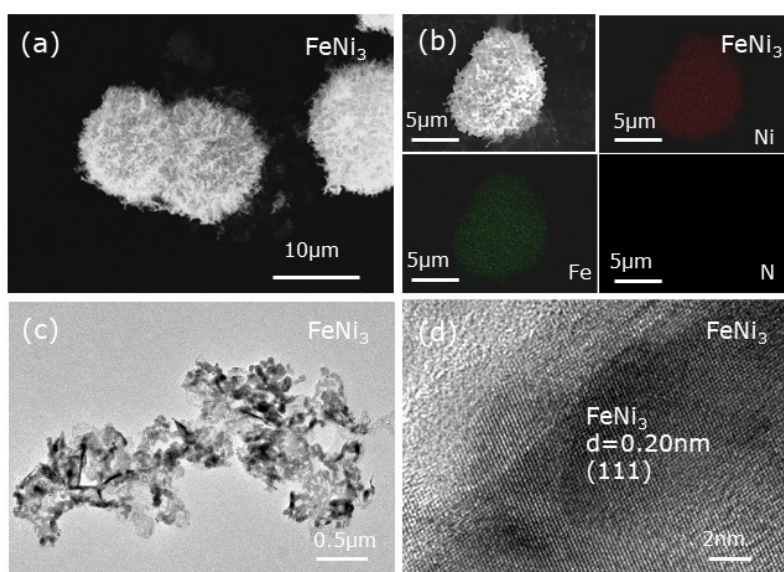


Figure S2 (a) SEM, (b) Element distributions, (c) TEM and (d) HRTEM images of FeNi₃,

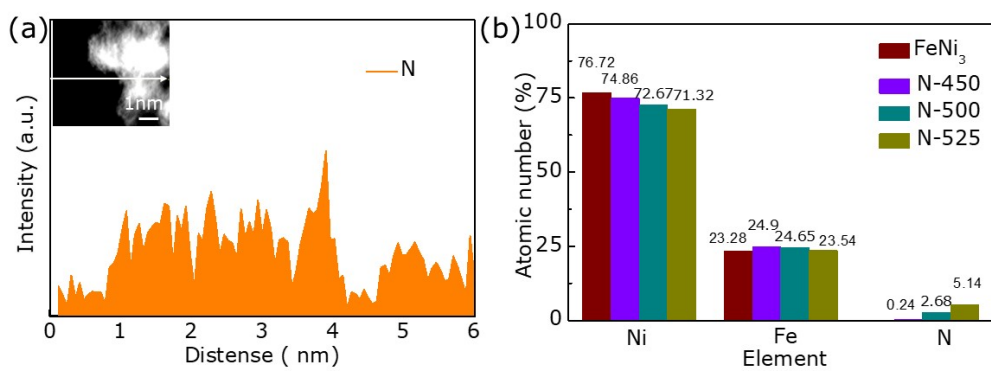


Figure S3 (a) EDX line scan of N element (the illustration is the scan area) and (b) EDS spectra of all samples.

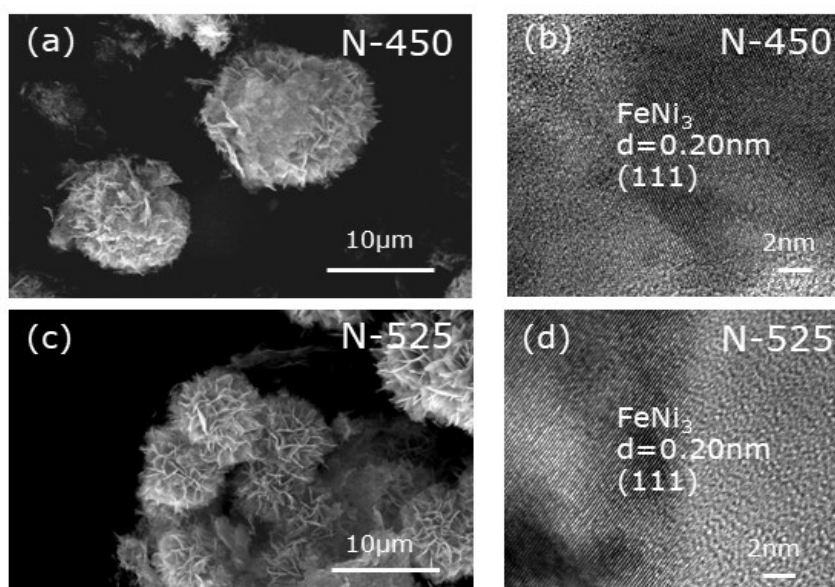


Figure S4 (a) SEM and (b) HRTEM images of N-450; (c) SEM and (d) HRTEM images of N-525.

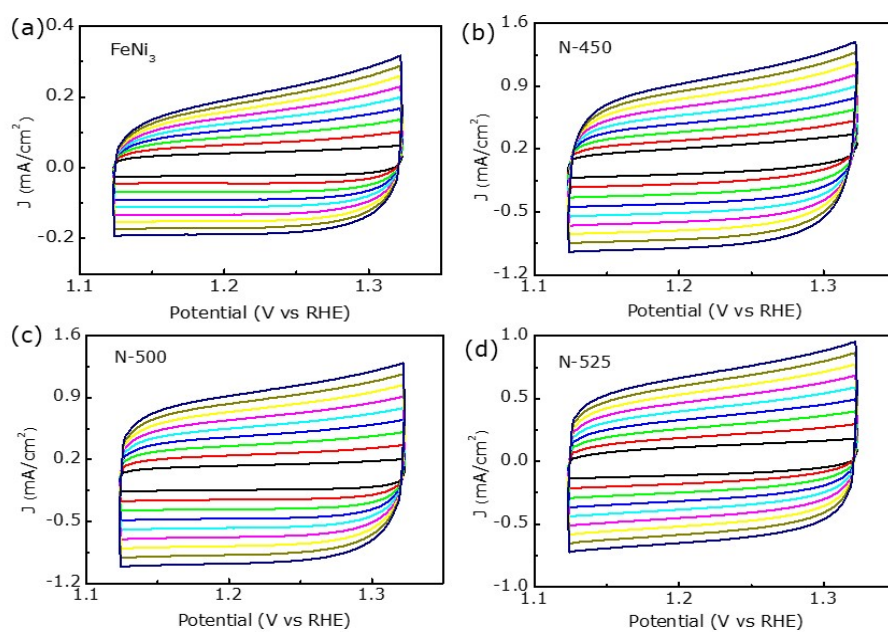


Figure S5. the cyclic voltammetry (CV) curve of (a) FeNi₃, (b) N-450, (c) N-500, and (d) N-525.

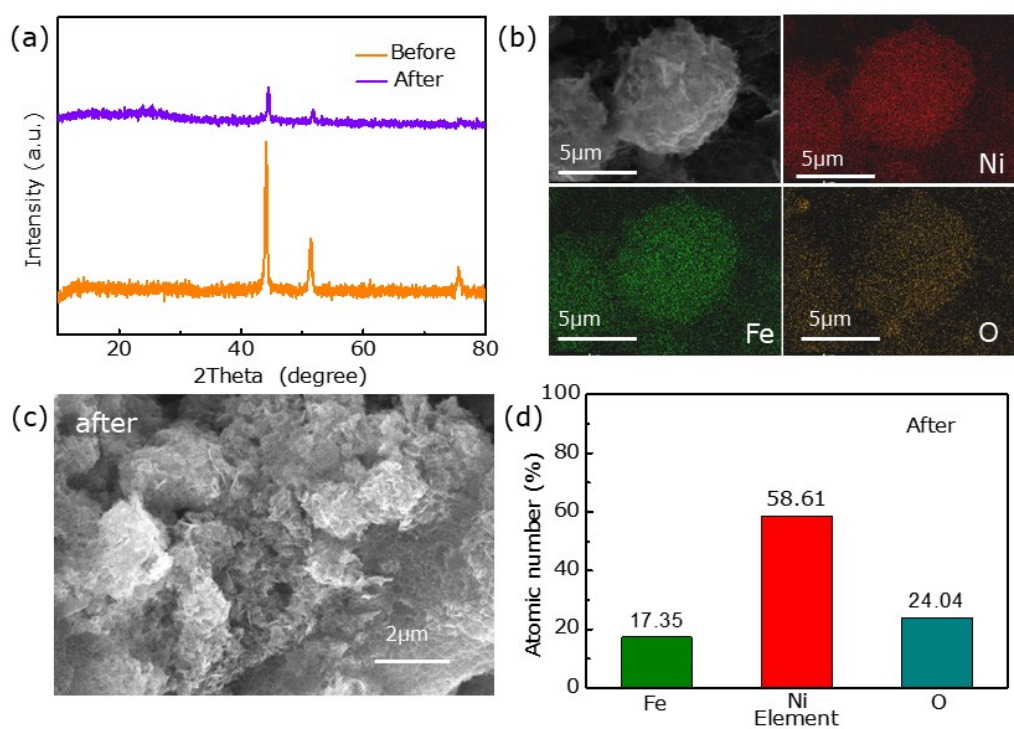


Figure S6 (a) XRD patterns before and after the stability test (b) Element distributions and (c) SEM image (d) EDX spectra after the stability test of N-500.

Table S1. Comparison of OER catalytic performance of Fe-Ni-based catalysts.

Catalyst	j (mA cm ⁻²)	η (mV)	Tafel slope
FeNi ₃ @NCNT5	10	264	58.5
FeNi ₃ @NC6	10	277	77
Fe-enriched-FeNi ₃ /NC7	10	260	82
NiFeC-800-58	10	269	72
FeNi ₃ N/FeNi ₃ 9	10	254	76
FeNi ₃ @GCDs-1010	10	238	48.7
FeNiNC-550-311	10	340	86.67
Fe ₃ O ₄ /Ni ₃ FeN2	10	261	43.6
Ni-FeO _x /FeNi ₃ /NF12	50	267	78.6
NiFeCoS _x @FeNi ₃ 13	10	210	45
Ni:Fe = 7.5:2.5/CF14	15	330	37
FeNi ₃ /Ni ₂ P@NCNTs15	10	298	81.82
Fe _{0.2} Ni _{0.8} /NC-600-a16	20	290	76
N-FeNi₃	10	251	34.63

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