Efficient Interlayer Confined Nitrate Reduction Reaction and Oxygen Generation Enabled by Interlayer Expansion

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Fig. S1 AFM image of single $\alpha$-Ni$_{0.881}$Fe$_{0.119}$(OH)$_2$ nanosheet.
Fig. S2 (a,b) SEM images of α-Ni$_{0.881}$Fe$_{0.119}$(OH)$_2$ nanosheets.
Fig. S3 TEM images of $\alpha$-Ni$_{1-x}$Fe$_x$(OH)$_2$ nanosheets with different Fe doping levels: (a) $x=0$, (b) $x=0.053$, (c) $x=0.098$, (d) $x=0.119$, (e) $x=0.150$, (f) $x=0.171$. 
Fig. S4 Nitrogen adsorption-desorption curves and pore size distributions curves of (a,b) α-Ni$_{0.881}$Fe$_{0.119}$(OH)$_2$ nanosheets and (c,d) α-Ni(OH)$_2$ nanosheets.
Fig. S5 XPS survey of $\alpha$-Ni$_{0.881}$Fe$_{0.119}$(OH)$_2$ sample.
Fig. S6 (a) UV-vis absorption spectra based on spectrophotometry of Nessler’s reagent and (b) NH$_3$ concentration-absorbance curve at 420 nm of standard NH$_3$ solutions with a series of concentrations. (c) UV-vis absorption spectra for $\alpha$-Ni$_{1-x}$Fe$_x$(OH)$_2$ samples acquired at –0.6 V. (d) UV-vis absorption spectra for $\alpha$-Ni$_{0.881}$Fe$_{0.119}$(OH)$_2$ nanosheets acquired at –0.6 to –0.1 V.
Fig. S7 (a) UV-vis absorption spectra based on spectrophotometry of indophenol blue method and (b) NH$_3$ concentration-absorbance curve at 655 nm of standard NH$_3$ solutions with a series of concentrations. (c) NH$_3$ yield rates and NO$_3^-$-to-NH$_3$ FEs of $\alpha$-Ni$_{1-x}$Fe$_x$(OH)$_2$ nanosheets at $-0.6$ V. (d) Potential-dependent NH$_3$ yield rates and FEs of $\alpha$-Ni$_{0.881}$Fe$_{0.119}$(OH)$_2$ nanosheets.
Fig. S8 The comparison of LSV curves for $\alpha$-Ni$_{0.881}$Fe$_{0.119}$(OH)$_2$ and pristine $\alpha$-Ni(OH)$_2$ nanosheets recorded in KOH or the mixture of KNO$_3$ + KOH.
Fig. S9 (a) NH$_3$ yield rate and Faradaic efficiency for α-Ni$_{0.881}$Fe$_{0.119}$(OH)$_2$ sample recorded at –0.7 V. (b) XRD pattern of α-Ni$_{0.881}$Fe$_{0.119}$(OH)$_2$ after electrochemical catalysis at –0.7 V.
Fig. S10 Potential-dependent NH$_3$ production rates and FEs for α-Ni(OH)$_2$ nanosheets.
Fig. S11 (a) UV-vis absorption spectra and (b) concentration-absorbance curve of NO$_2^-$ solutions with a series of standard concentrations.
Fig. S12 Working curves for the determination of produced amount of (a) H$_2$ and (b) N$_2$. 

(a) $y = 7283.1 - V_{H_2} + 7.4283$ 
$R^2 = 0.9994$

(b) $y = 2635.7 - V_{N_2} + 56.118$ 
$R^2 = 0.9992$
Fig. S13 (a) TEM image and (b) XRD pattern of $\alpha$-Ni$_{0.881}$Fe$_{0.119}$(OH)$_2$ nanosheets after catalysis.
Fig. S14 (a) NH$_3$ yield rates and FEs of α-Ni$_{0.881}$Fe$_{0.119}$(OH)$_2$ nanosheets at (a) different concentrations of NO$_3^-$ and (b) pH.
Fig. S15 (a) XRD pattern and (b) SEM image of $\alpha$-Ni$_{0.881}$Fe$_{0.119}$(OH)$_2$ nanosheets arrays grown on nickel foam.
Fig. S16 (a) the NTRR and (b) OER performance of $\alpha$-Ni$_{0.881}$Fe$_{0.119}$(OH)$_2$ nanosheet arrays on nickel foam.
Fig. S17 $I-t$ curves using $\alpha$-Ni$_{0.881}$Fe$_{0.119}$(OH)$_2$ nanosheets as (a) cathodic catalyst and (b) bifunctional catalyst.
**Fig. S18** $I$-$t$ curves using $\alpha$-Ni$_{0.881}$Fe$_{0.119}$(OH)$_2$ nanosheets as bifunctional catalyst or cathodic catalyst and the corresponding energy efficiencies.
Fig. 19 Cyclic voltammetry curves recorded at different scan rates (20-140 mV s\(^{-1}\)):
(a) \(\alpha\text{-Ni}_{0.881}\text{Fe}_{0.119}(OH)_2\) nanosheets, (b) pristine \(\alpha\text{-Ni(OH)}_2\) nanosheets. (c) Comparative \(C_{dl}\) for \(\alpha\text{-Ni}_{0.881}\text{Fe}_{0.119}(OH)_2\) nanosheets and \(\alpha\text{-Ni(OH)}_2\) nanosheets derived from CV curves with different scan rates.
Fig. S20 EIS Nyquist plots for $\alpha$-Ni$_{1-x}$Fe$_x$(OH)$_2$ nanosheets with different Fe doping levels ($x=0, 0.053, 0.098, 0.119, 0.150, 0.171$).
Fig. S21 Schematic illustration of atomic layer deposition to block the outmost layers of the layered $\alpha$-Ni$_{0.881}$Fe$_{0.119}$(OH)$_2$ nanosheets.
Fig. S22 K 2p spectrum of $\alpha$-Ni$_{0.881}$Fe$_{0.119}$(OH)$_2$ samples after NTRR test.
Fig S23. Zeta potentials of $\alpha$-Ni$_{0.881}$Fe$_{0.119}$(OH)$_2$ nanosheets before or after catalysis.
Table S1. The mole ratios of Ni/Fe in the precursors and the corresponding x in the final $\alpha$-Ni$_{1-x}$Fe$_x$(OH)$_2$ samples.

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<thead>
<tr>
<th>Ni/Fe ratios</th>
<th>x</th>
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<tbody>
<tr>
<td>1:0.1</td>
<td>0.053</td>
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<tr>
<td>1:0.2</td>
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<td>0.119</td>
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<td>1:0.4</td>
<td>0.150</td>
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<td>1:0.5</td>
<td>0.171</td>
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