**Supporting Information**

# Heterointerface Engineering Constructs Microenvironment Enhancing Catalytic Kinetics of Fe/Ni oxyhydroxide@FeNi alloy for Overall Water Splitting

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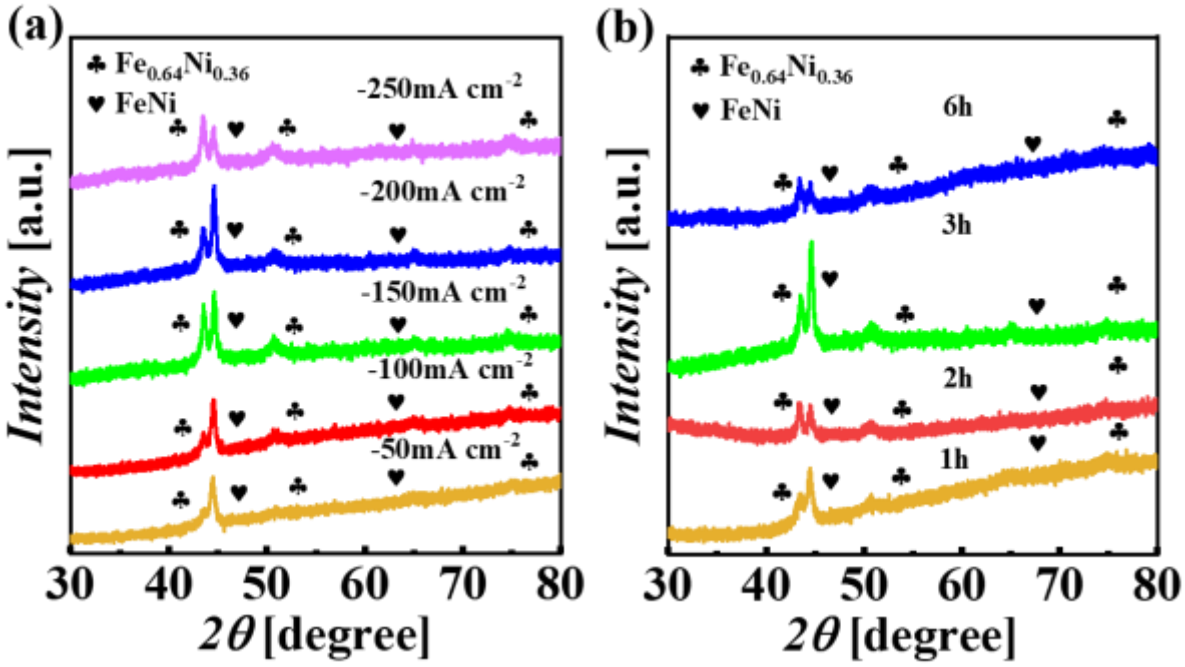


**Figure S1.** XRD patterns of FeNi-(1:1) $O_x(OH)_y$ at different deposition currents and deposition time.

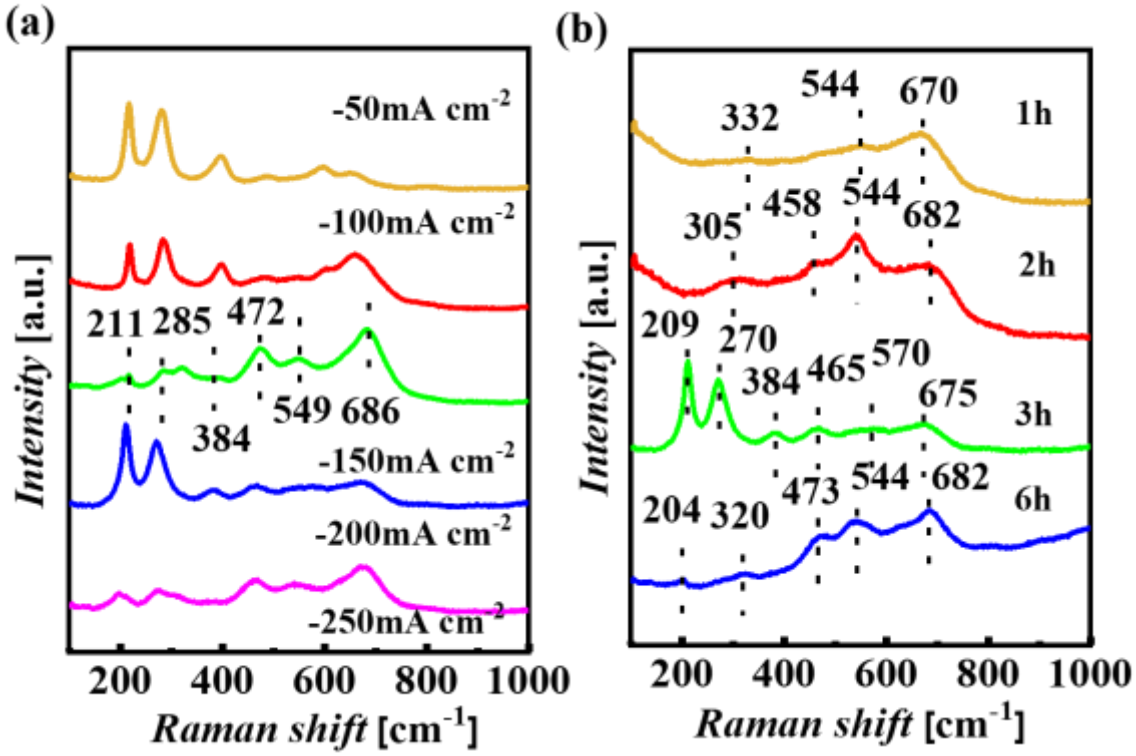


**Figure S2.** Raman spectra of FeNi-(1:1) $O_x(OH)_y$ at different deposition currents and deposition time.

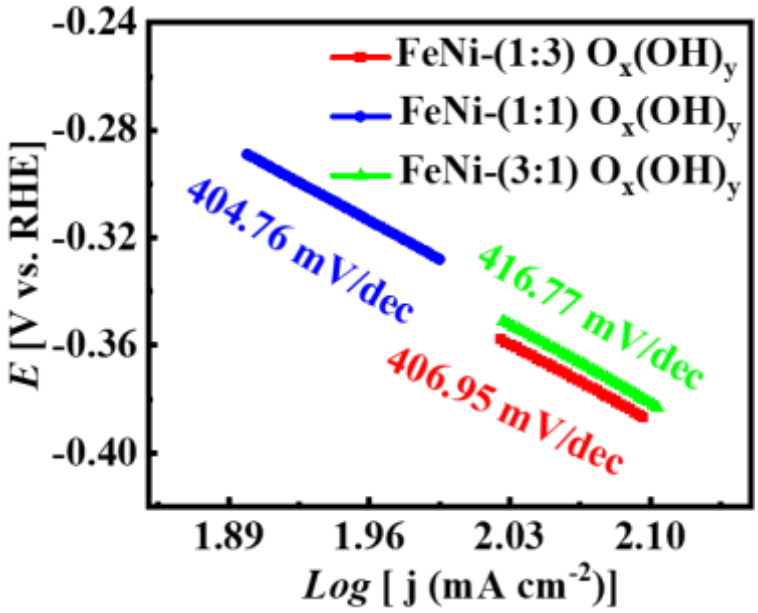


**Figure S3.** The Tafel slope was calculated at the current density of 100 mA $cm^{-2}$ for HER.

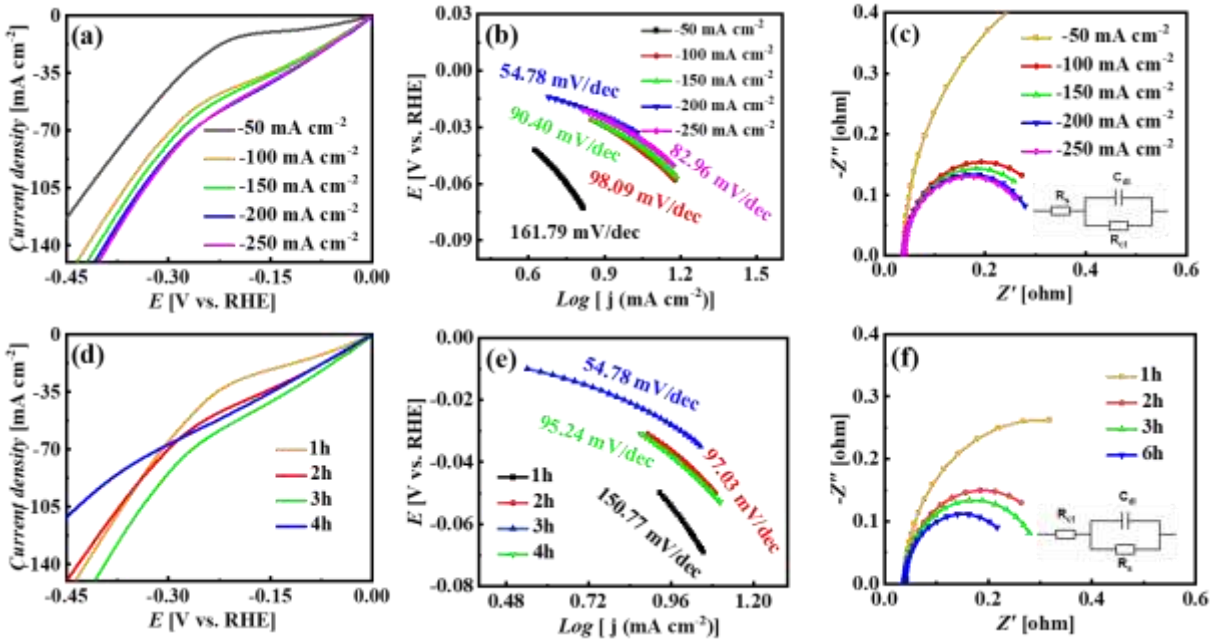


**Figure S4.** LSV, Tafel plots, and Nyquist plots of FeNi-(1:1) $O_x(OH)_y$ at different deposition currents and deposition time.

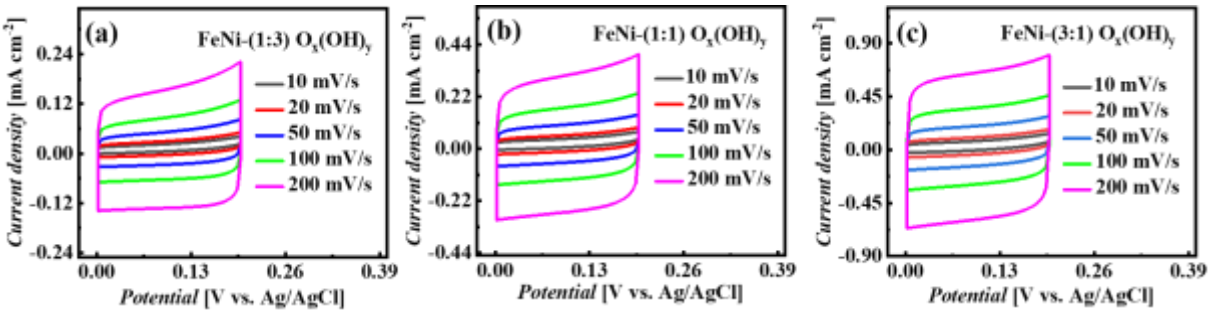


**Figure S5.** Charging currents measured in the non-Faradaic potential range of 0 to 0.2 V vs. Ag/AgCl at scan rates of 10, 20, 50, 100, 200 mV/s, FeNi-(1:3) $O_x(OH)_y$ (a), FeNi-(1:1) $O_x(OH)_y$ (b), FeNi-(3:1) $O_x(OH)_y$ (c).

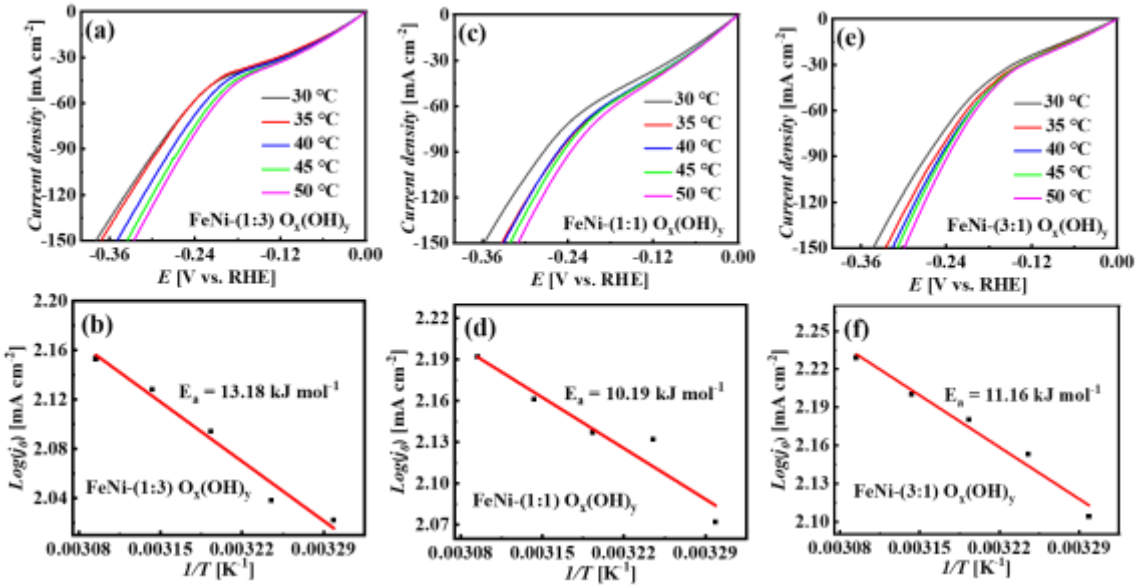


**Figure S6.** The figure of the polarization curves at different electrolyte temperatures correspond to Arrhenius plots of FeNi-(1:3) $O_x(OH)_y$, FeNi-(1:1) $O_x(OH)_y$, and FeNi-(3:1) $O_x(OH)_y$ for HER.

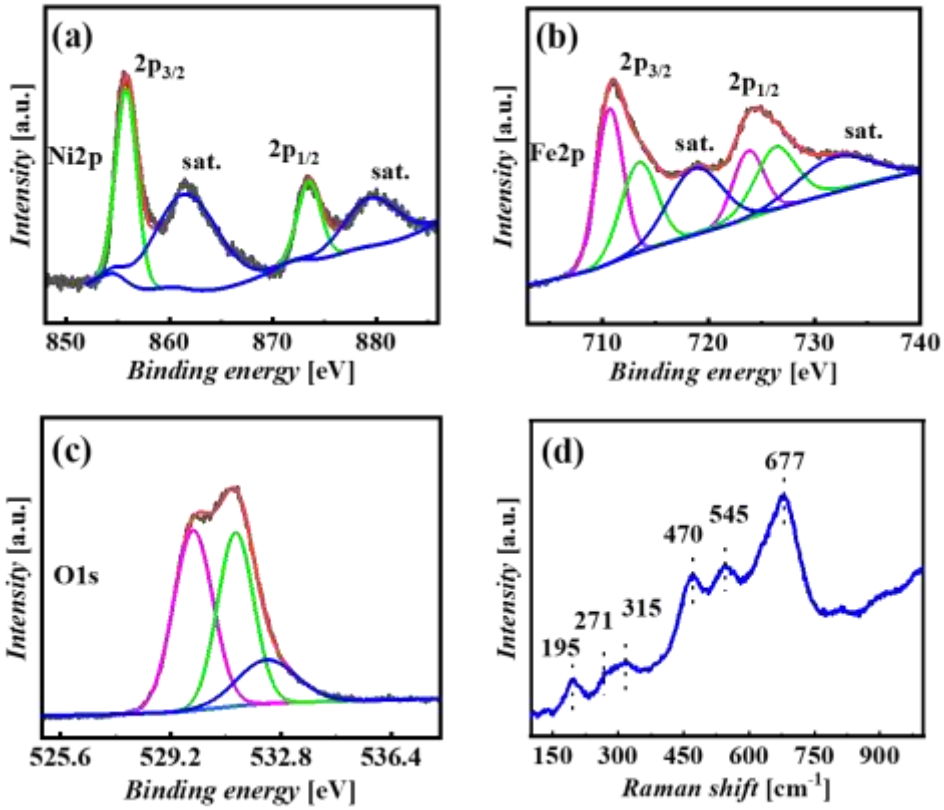


**Figure S7.** The XPS survey and Raman spectra of FeNi-(1:1) $O_x(OH)_y$ as cathode for HER after stability test.

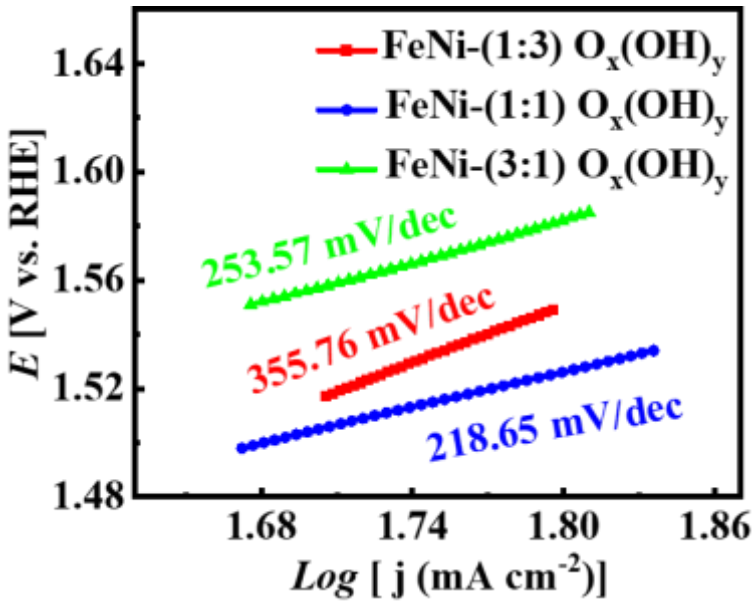


**Figure S8.** The Tafel slope was calculated at the current density of 100 mA $cm^{-2}$ for OER.

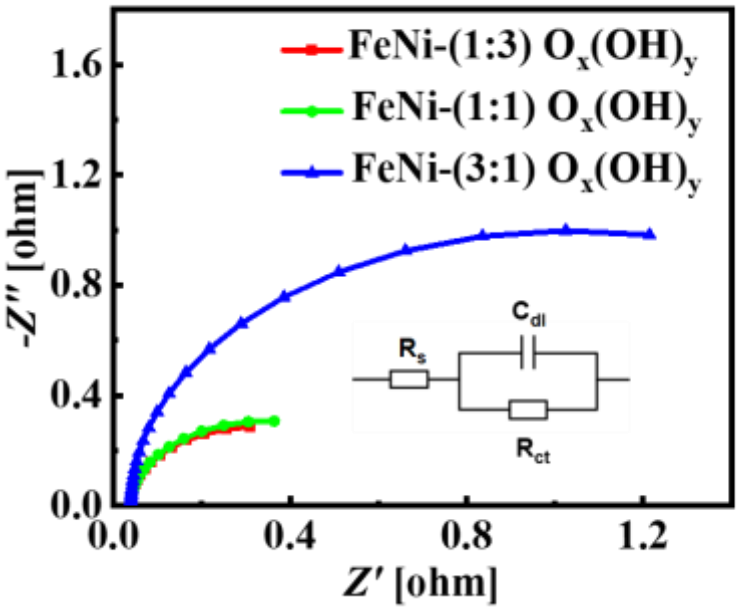


**Figure S9.** Nyquist plots for corresponding electrodes for OER.

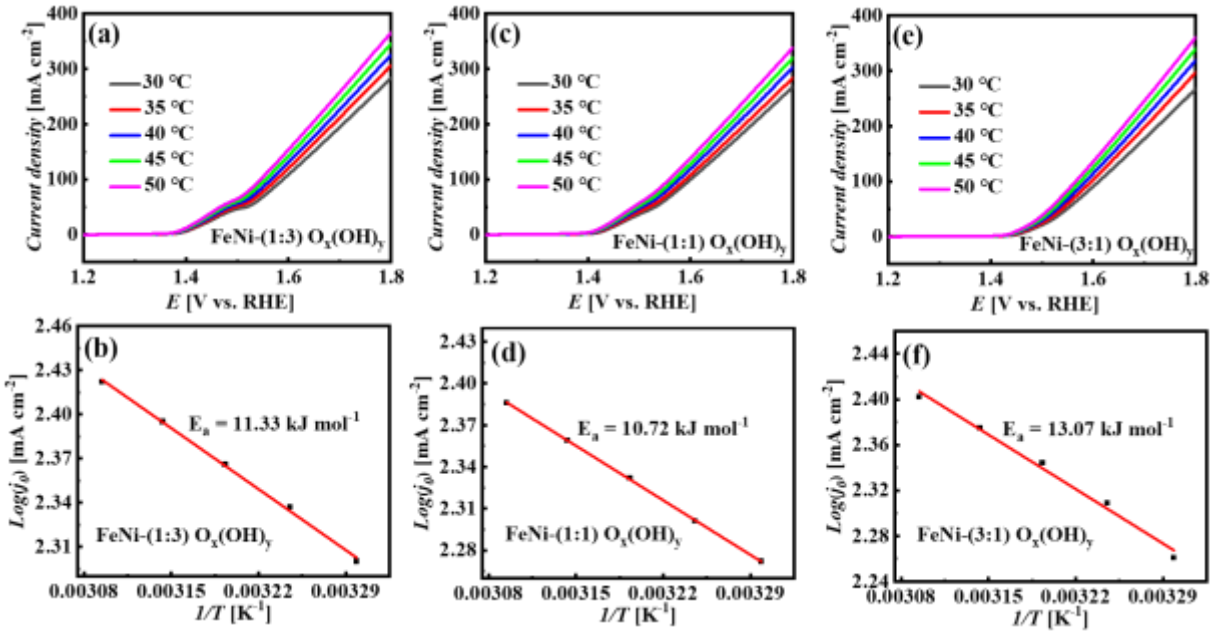


**Figure S10.** The figure of the polarization curves at different electrolyte temperatures correspond to Arrhenius plots of FeNi-(1:3) $O_x(OH)_y$, FeNi-(1:1) $O_x(OH)_y$, and FeNi-(3:1) $O_x(OH)_y$ for OER.

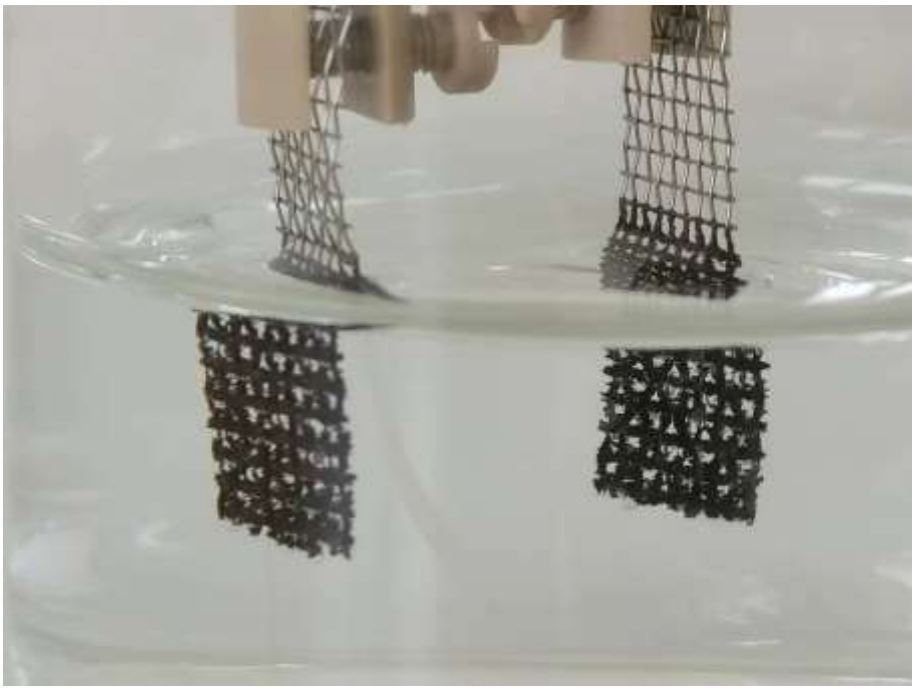

**Figure S11.** Overall water splitting for the device with FeNi-(1:1) $O_x(OH)_y$ || FeNi-(1:1) $O_x(OH)_y$ and 1.5 V battery. The video can be viewed in support materials.

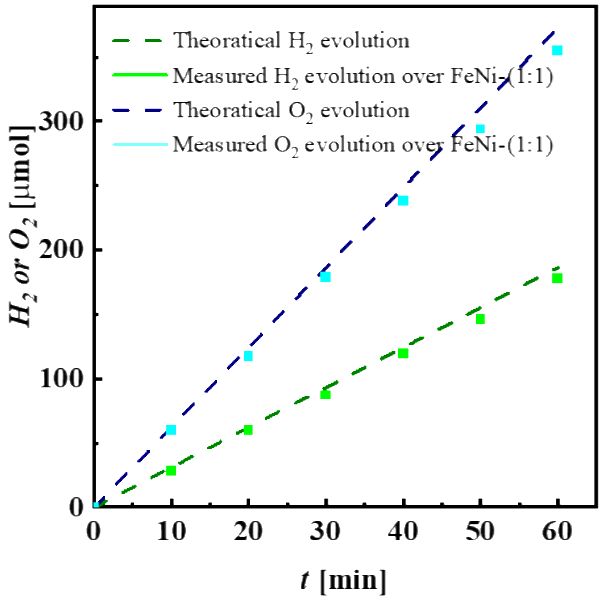


**Figure S12.** Experimental and theoretical amounts of $H_2$ and $O_2$ by the FeNi-(1:1) $O_x(OH)_y$ electrode at a fixed current density of 10 mA $cm^{-2}$.

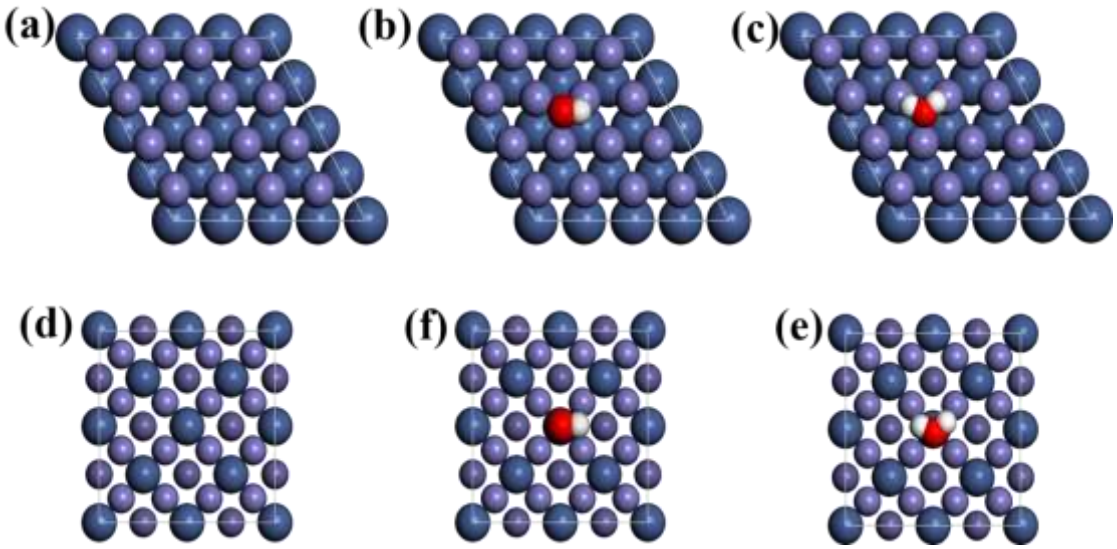


**Figure S13.** Models of kamacite and $Fe_{0.64}Ni_{0.36}$ were used to simulate the interface in FeNi-(1:3) $O_x(OH)_y$, FeNi-(1:1) $O_x(OH)_y$, FeNi-(3:1) $O_x(OH)_y$.

**Table S1**

Comparison of the obtained cell voltage in this study with those of the recently reported bifunctional electrocatalysts for overall water-splitting in alkaline solution.

| Materials | Electrolyte | Cell voltage (V) | J (mA $cm^{-2}$) | Ref. |
|---|---|---|---|---|
| **FeNi-(1:1) $O_x(OH)_y$** | **1.0 M KOH** | **1.897** | **100** | **This work** |
| W-NiFe | 1.0 M KOH | 1.59 | 10 | [1] |
| NiFe@$Co(OH)_2$ NSAs/NF | 1.0 M KOH | 1.58 | 10 | [2] |
| Ce-NiFe-LDH | 1.0 M KOH | 1.59 | 10 | [3] |
| $NiFe(OH)_x$ | 1.0 M KOH | 1.80 | 100 | [4] |
| NiFe-LDH@$Ni(OH)_2$ | 1.0 M KOH | 1.80 | 100 | [5] |
| Fe-CoNi-LDH | 1.0 M KOH | 1.81 | 100 | [6] |
| $Fe(OH)_3$/β-$Ni(OH)_2$ | 1.0 M KOH | 1.83 | 400 | [7] |
| NiFeMo alloy | 1.0 M KOH | 1.75 | 500 | [8] |
| NiFe oxyhydroxide@NiFe alloy | 1.0 M KOH | 1.76 | 1000 | [9] |

**Reference**


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