

Appendix A Supplementary data

Construction of a $\text{Co}_2(\text{OH})_2\text{CO}_3@$ CoFe-LDH heterostructure on Ni foam with tunable interfacial properties for electrochemical applications

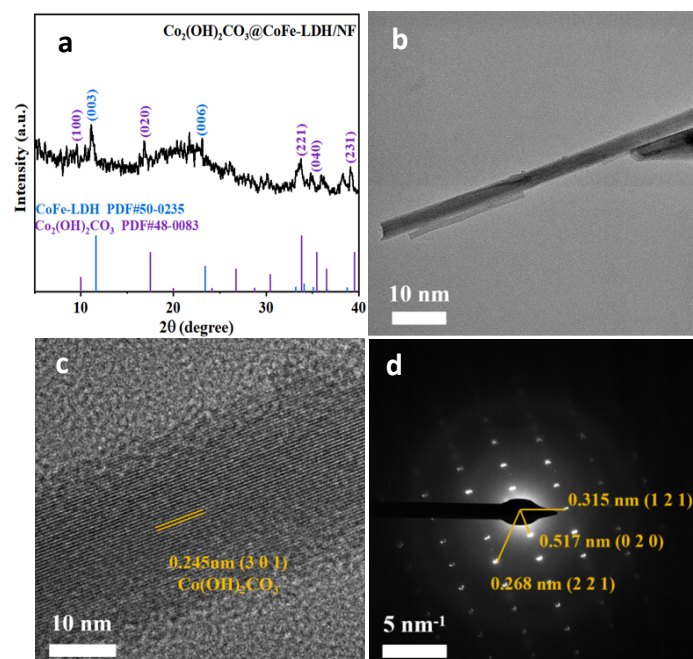
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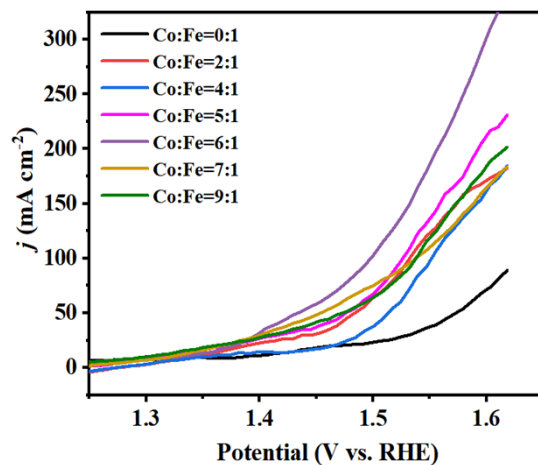
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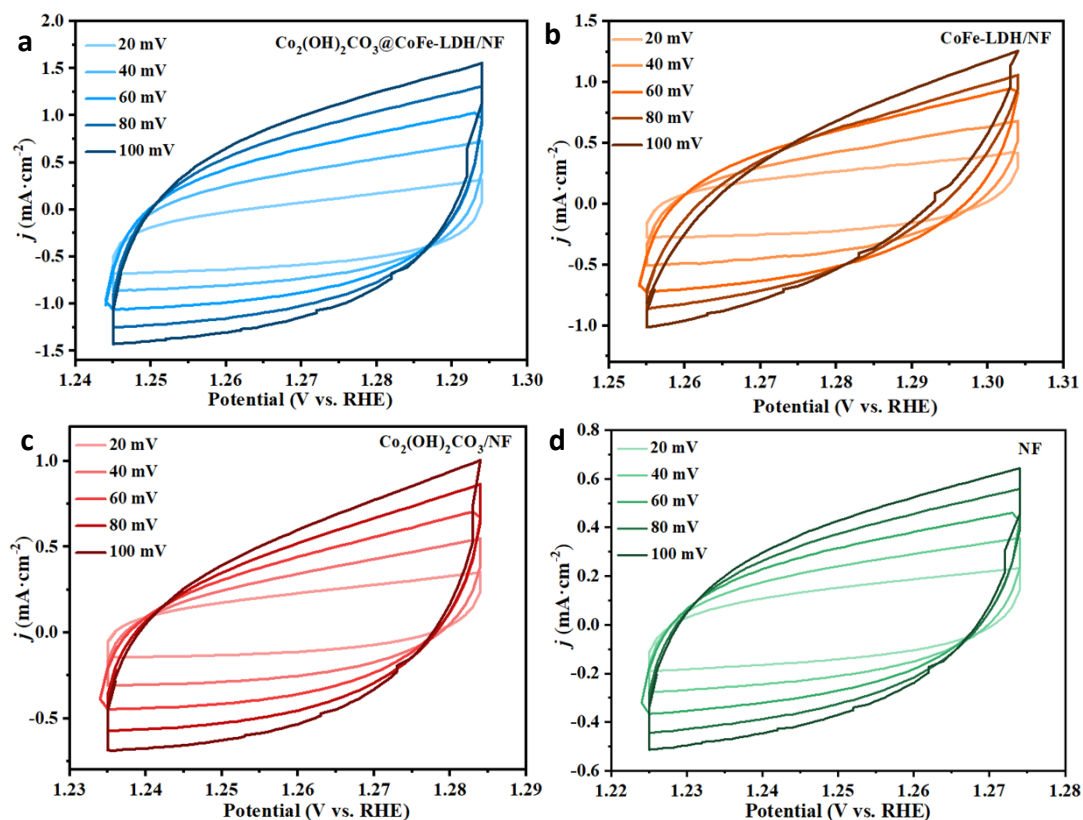
Supplementary Fig. 1. (a) XRD patterns of $\text{Co}_2(\text{OH})_2\text{CO}_3@\text{CoFe-LDH/NF}$; (b) TEM image; (c) HRTEM image and (d) SAED pattern of the nanoneedle-dominated region.



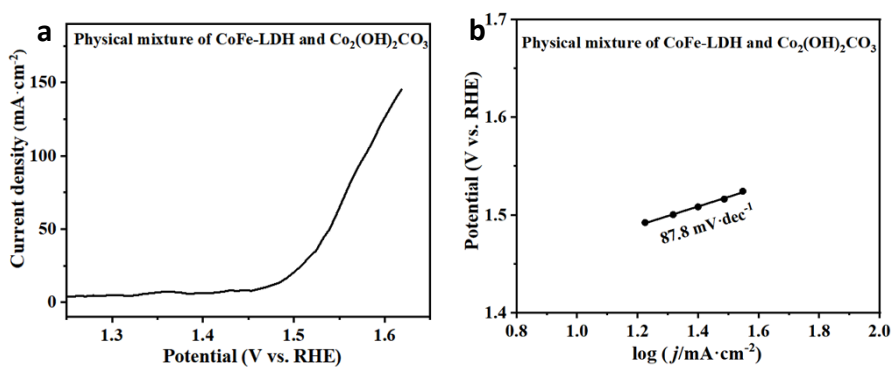
Supplementary Fig. 2 LSV curves of synthetic catalysts with different metal ion ratios.

Supplementary Table 1. The overpotentials required to achieve a current density of $50 \text{ mA}\cdot\text{cm}^{-2}$ for samples with different metal ion ratios

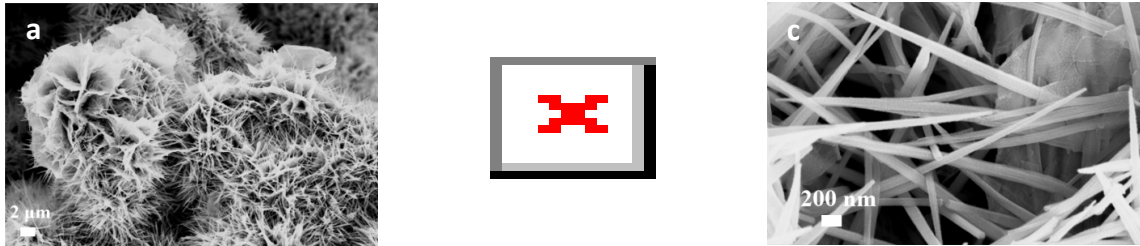
| $\text{Co}^{2+}:\text{Fe}^{3+}$ metal ion ratios | 0:1 | 2:1 | 4:1 | 5:1 | 6:1 | 7:1 | 9:1 |
|---|-----|-------|-------|-------|-------|-------|-------|
| Overpotential (mV) | 343 | 254.1 | 283.7 | 244.6 | 203.5 | 224.5 | 241.1 |



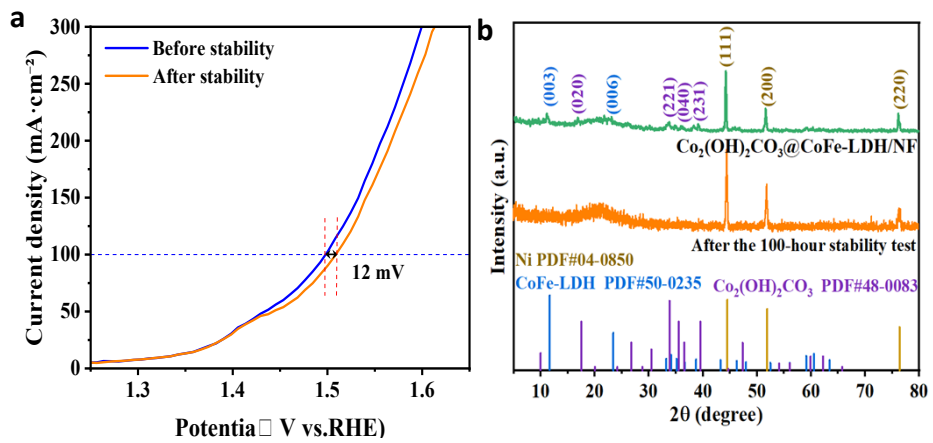
Supplementary Fig. 3 Cyclic voltammograms at different scan rates for (a) $\text{Co}_2(\text{OH})_2\text{CO}_3@\text{CoFe-LDH}/\text{NF}$, (b) $\text{CoFe-LDH}/\text{NF}$, (c) $\text{Co}_2(\text{OH})_2\text{CO}_3/\text{NF}$, and (d) NF .



Supplementary Fig. 4 (a) LSV curves and (b) the corresponding Tafel slope of the physical mixture of CoFe-LDH and $\text{Co}_2(\text{OH})_2\text{CO}_3$.



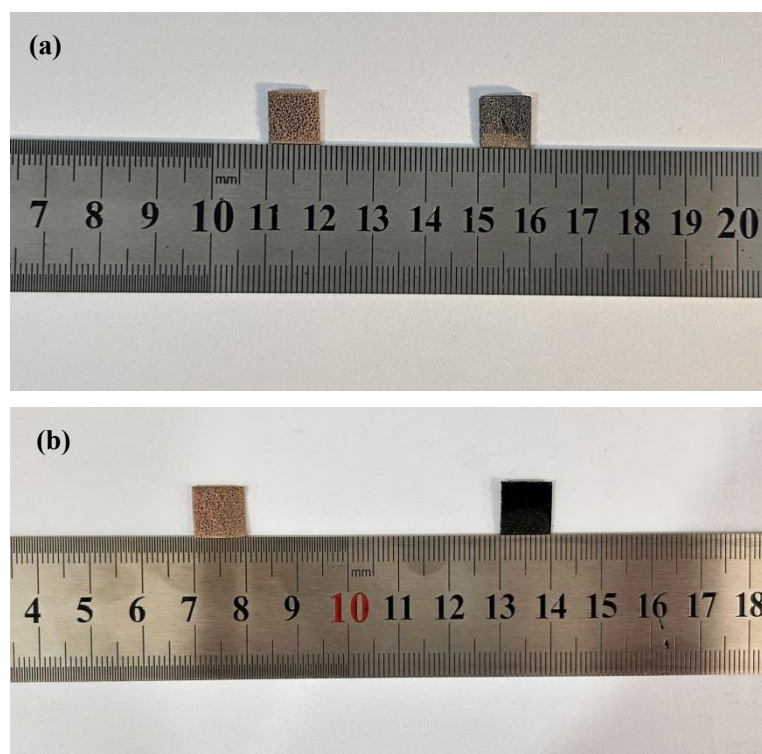
Supplementary Fig. 5 (a-c) SEM images of the catalyst at different magnifications after the durability test



Supplementary Fig. 6 comparison of (a) the LSV curves and (b) corresponding XRD patterns before and after the durability test.

Supplementary Table 2. Comparison of the OER performance of $\text{Co}_2(\text{OH})_2\text{CO}_3$ @CoFe-LDH with previously reported CoFe-LDH based electrocatalysts

| Catalysts | η_{10} (mV) | η_{50} (mV) | η_{100} (mV) | Tafel slope (mV \cdot dec $^{-1}$) | Ref |
|--|------------------|------------------|-------------------|---------------------------------------|-----------|
| Ru, Ir@CoFe-LDH/IF | - | - | 256 | 49.3 | [10] |
| $\text{Ni}_{0.85}\text{Se}$ @CoFe LDH/NF nanosheet | 223 | - | 280 | - | [12] |
| CoFe-LDH@NiSe | 127 | 285 | - | 37 | [13] |
| Mo-NiSe $_x$ /CoFe LDH | - | - | 200 | 77.3 | [14] |
| MoS $_2$ /CoFe-LDH/NF nanosheet | 224 | 255 | 278 | 57.91 | [15] |
| CoFe LDH/MoS $_2$ /Ni $_3$ S $_2$ /NF | 228 | 277 | - | 54.66 | [23] |
| Ni(OH)S@CoFe-LDH/NF | - | - | 238 | 48.2 | [24] |
| ZIF@CoFe-LDH NCs nanocage | - | 278 | - | 50 | [17] |
| CoFe-LDH@MnCo-CH nanosheet | 250 | 276 | 290 | 50.0 | [36] |
| CoFe LDH/Co $_2$ Fe $_2$ O $_4$ nanosheet | 162 | - | 400 | 85.6 | [39] |
| Co-LDH@ZIF-67 nanosheet | 187 | - | 290 | 59 | [31] |
| CoFe-LDH/GF nanowire | 252 | - | 285 | 61 | [51] |
| CoFe-LDH@MnCo-CH nanowire | 250 | - | - | 50.0 | [55] |
| CoFe-LDH | - | - | 360 | 38.6 | [34] |
| Ce $_{0.2}$ CoFe-LDH | 227 | - | 310 | 48.30 | [40] |
| CoFe/a-NiFe LDH | - | - | 265 | 38.8 | [8] |
| $\text{Co}_2(\text{OH})_2\text{CO}_3$ @CoFe-LDH/NF | 104 | 203.5 | 267.2 | 57.6 | This work |



Supplementary Fig. 7 (a) Digital photograph of $\text{Co}_2(\text{OH})_2\text{CO}_3@\text{CoFe-LDH/NF}$ unreacted samples and LSV reaction tests, (b) Digital photograph of $\text{Co}_2(\text{OH})_2\text{CO}_3@\text{CoFe-LDH/NF}$ unreacted samples and stability tests.