

Fig. S1 XRD patterns for CoFe-LDH-120 at different hydrothermal times.



Fig. S2 (a) XRD patterns for Fe(OH)₃ of different hydrothermal temperatures; SEM image of (b) Fe(OH)₃/NF-80-12, (c and d) Fe(OH)₃/NF-100-12.



(b) Co(OH)₂/NF-80-12, (c) Co(OH)₂/NF-100-12 and (d) Co(OH)₂/NF-120-12; (e) LSV curves and

(f) Tafel plots of Co(OH)₂/NF-x-12 (x=80, 100, 120); (g) the growth process of CCH nanowires.



Fig. S4 SEM image of (a) CoFe-LDH-120-8; (b) CoFe-LDH-120-10; (c) CoFe-LDH-120-12; (d) CoFe-LDH-120-14.



Fig. S5 SEM image of (a) CoFe-LDH-80-8; (b) CoFe-LDH-80-10; (c) CoFe-LDH-80-12; (d) CoFe-LDH-80-14.



Fig. S6 Typical energy-dispersive X-ray spectroscopy (EDS) for CoFe-LDH-120-12.



Fig. S7 (a) LSV curves of CoFe-LDH-120-8, CoFe-LDH-120-10, CoFe-LDH-120-12, CoFe-LDH-120-14, Ni foam (NF) and RuO₂ in 1 M KOH solution (scan rate $2mV s^{-1}$); (b) Tafel plots of the corresponding synthesized catalysts; (c) Overpotential of CoFe-LDH-120-8, CoFe-LDH-120-10, CoFe-LDH-120-12, CoFe-LDH-120-14 at 100 and 200 mA cm⁻²; (d) Overpotentials of CoFe-LDH-80-8, CoFe-LDH-80-10, CoFe-LDH-80-12, CoFe-LDH-80-14 at 100 and 200 mA cm⁻².



Fig. S8 SEM image of CoFe-LDH-120 after continuous 97h OER measurement.



Fig. S9 XPS full spectrum and XPS spectra of (b) Co 2p, (c) Fe 2p and (d) O 1s performed on CoFe-LDH-120-12 before and after long-term OER electrolysis.



Fig. S10 The CV curves of different rates of (a) CoFe-LDH-80-12, (c) CoFe-LDH-100-12, (e) CoFe-LDH-140-12; linear relationships of the oxidation peak currents vs. scan rates for (b) CoFe-LDH-80-12, (d) CoFe-LDH-100-12, (f) CoFe-LDH-140-12.



Fig. S11 (a) Linear relationships of the oxidation peak currents vs. scan rates for CoFe-LDH-120-y (y=8, 10, 12, 14); (b) Turnover frequencies (TOFs) of CoFe-LDH-120- y (y=8, 10, 12, 14).