Supporting Information

Cationic defects-enriched hydroxides as anodic catalysts for efficient seawater electrolysis

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Fig. S1 SEM images of (a-c) NiFe LDH and (d-f) CD-NiFe LDH-E.



Fig. S2 (a-d) TEM images of CD-NiFe LDH-E.



Fig. S3 Optical photograph of (a) residual solution with/without including EDTA-2Na after the solvothermal reaction, (b) a higher magnification toward the specified area for (a), and (c) different residually solution for UV/Vis measurement. The numbers 1-4 in (c) were correspond to tested various solutions for UV/Vis, including: (1) reagent blank, i.e. D-NiFe LDH(RS), (2) reagent blank added with 1 mg/mL of EDTA-2Na and 1 mg/mL of Ni(NO₃)₂·6H₂O, (3) reagent blank added with 1 mg/mL of EDTA-2Na 2Na and 1 mg/mL of Fe(NO₃)₃·9H₂O, and (4) CD-NiFe LDH-E(RS).



Fig. S4 Morphological and microstructural characterizations on D-NiFe LDH. (a, b) SEM images, (c) TEM images, and (d) HRTEM images. NOTE: the small amount of cation-clusters-defective structure were recorded toward a red dotted in Fig. S4d.



Fig. S5 (a) XRD patterns of different NiFe LDH samples. (b) ICP histogram for the concentration (ppm) for Ni ion, Fe ion, and total of Ni+Fe ions in the D-NiFe LDH(RS), and CD-NiFe LDH-E(RS).



Fig. S6 Performance of OER for the d-NiFe LDH-E in O₂-saturated 1 M KOH + 0.5 M NaCl solution. (a) OER polarization curves at 1600 rpm and corresponding (b) η_{50} and η_{200} .



Fig. S7 Voltammograms of the (a) CD-NiFe LDH-E, (b) D-NiFe LDH, and (c) NiFe LDH at various scan rates (20–100 mV s⁻¹) used to estimate the C_{dl} .



Fig. S8 Performance of OER for the cationic defects-enriched NiFe LDH with different Ni/Fe ratios obtained in O₂-saturated 1 M KOH + 0.5 M NaCl solution. (a) OER polarization curves at 1600 rpm and corresponding (b) η_{100} and η_{200} , and (c) Tafel.



Fig. S9 (a-d) SEM images of CD-NiFe LDH-E after stability test.



Fig. S10 (a-d) TEM images of NiFe LDH after stability test.

The TEM images of NiFe LDH after stability test displayed that it does not form any amorphous active species.



Fig. S11 (a) XPS survey of CD-NiFe LDH-E, D-NiFe LDH, and NiFe LDH. (b) High-resolution XPS spectra of Fe 2p toward CD-NiFe LDH-E and NiFe LDH.



Fig. S12 EDS spectra of (a) NiFe LDH, and (b) CD-NiFe LDH-E. Histogram of elementic percent toward (c) NiFe LDH, (d) CD-NiFe LDH-E.



Fig. S13 Spin states of octahedral sites of Ni^{3+} and Ni^{2+} .



Fig. S14 Spin states of octahedral sites of Ni^{3+} and Ni^{2+} .