Supporting information

## Sulfidation and NaOH etching on CoFeAl LDH evolved catalysts for efficient overall water splitting in alkaline solution

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**Fig. SI1.** (a) XRD patterns of the as-prepared samples: CoFeAl LDH, CoFeAl-NaOH and CoFeAl-T, and the magnified XRD pattern of (b) CoFeAl LDH and (c) CoFeAl-T.





**Fig. SI2.** SEM images and corresponding elemental mapping images of the asprepared samples: (a-c) Ni foam-T, (d-f) CoFeAl LDH, (g-i) CoFeAl-NaOH and (j-l) CoFeAl-T.



Fig. SI3. TEM image, HRTEM image and corresponding elemental mapping imageS of the as-prepared samples: (a-c) CoFeAl LDH, (d-f) CoFeAl-NaOH and (g-i) CoFeAl-T.

Samples	CoFeAl LDH	CoFeAl-T	CoFeAl-NaOH	CoFeAl-T-NaOH	CoFeAl-NaOH-T
Fe/Al	3.9	2.9	6.1	12.7	638.6

Table SI1 The content of Al in the different catalysts by molar ratio of Fe/Al



**Fig. SI4.** XPS analysis of the as-prepared samples: XPS survey spectra (a) High-resolution spectra of (b) Co 2p, Fe 2p (c) and Al 2p (d).



Fig. SI5. CV records with the bias range of 0.948-1.1 V (*vs.* RHE) in 1.0 M KOH by the scan rates of 40, 60, 80, 100, 120, 140, 160, 180, and 200 mV s<sup>-1</sup> for evaluating the ECSA.



**Fig. SI6.** XRD pattern, and SEM image and corresponding elemental mapping image, of CoFeAl-T-NaOH after long-term stability test.



Fig. SI7. XPS spectra of CoFeAl-T-NaOH before and after long-term stability test.



Fig. SI8. Chronopotentiometric curves of the as-prepared samples for HER measured at the current density of 150 mA cm<sup>-2</sup> for 20 hours.



**Fig. SI9.** (a,b) CV with a bias range of 0.652-1 V (*vs.* RHE) was recorded, and scan rates of 40, 60, 80, 100, 120, 140, 160, 180, and 200 mV s<sup>-1</sup> were used to evaluate ECSA. (c) Chronopotentiometric curves measured at the current density of 100 mA/cm<sup>-2</sup> for 20 hours.