

## Supporting information

### **The release of metal ions induced surface reconstruction of layered double hydroxide electrocatalyst**

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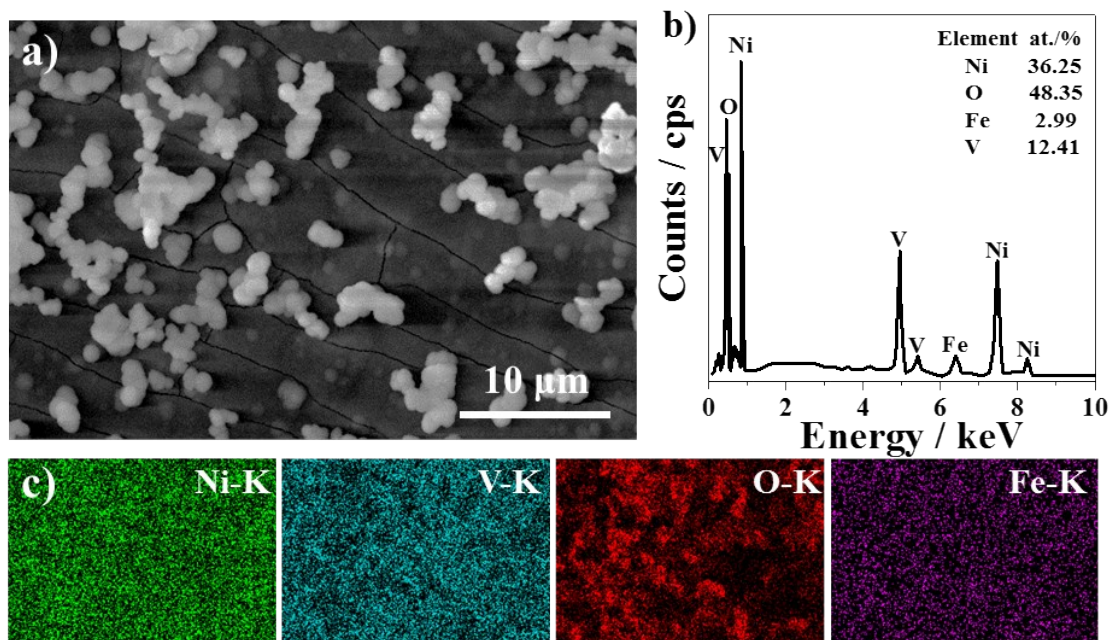
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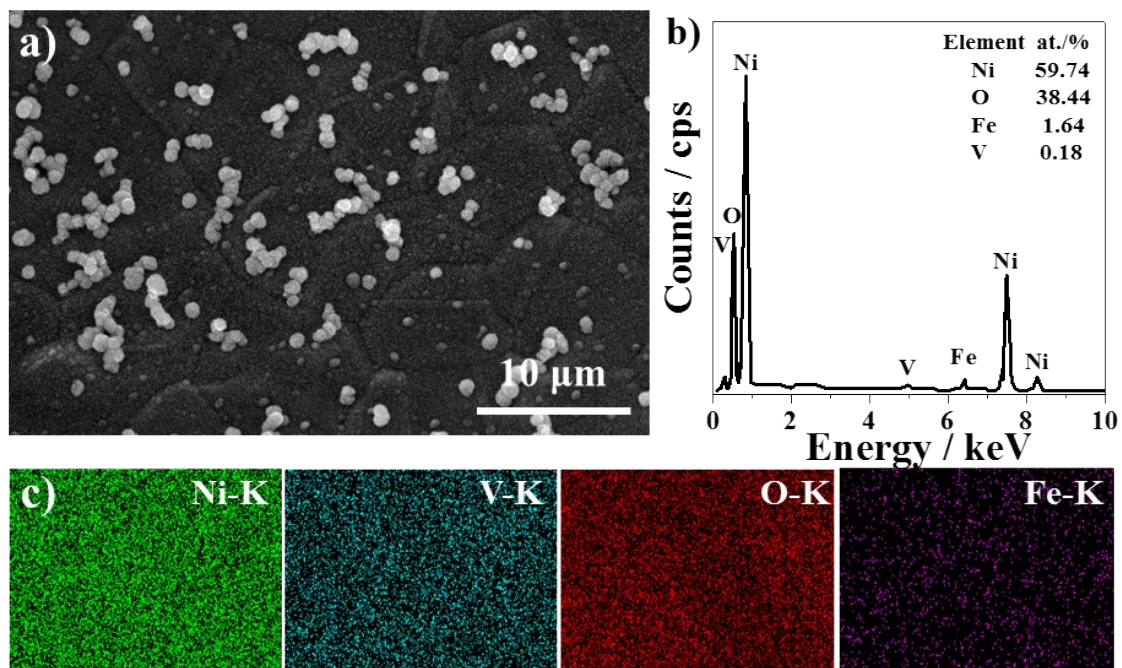
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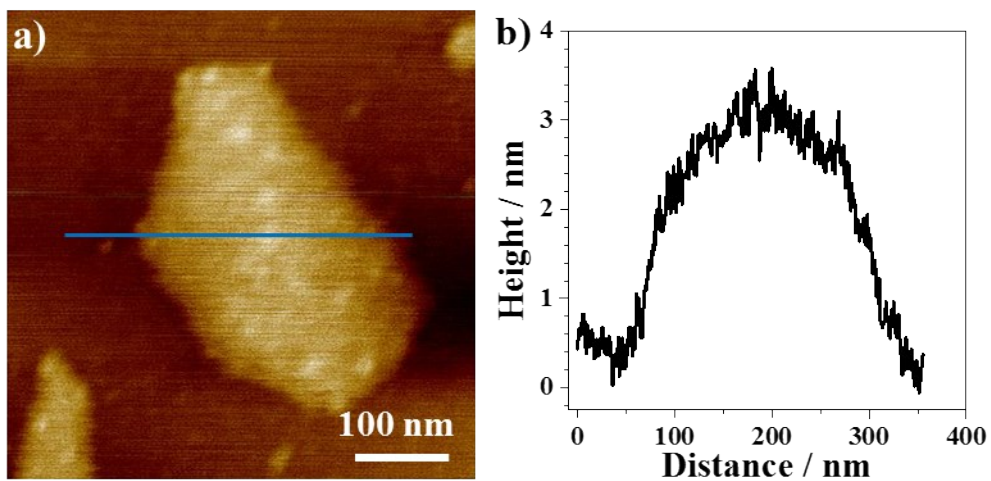
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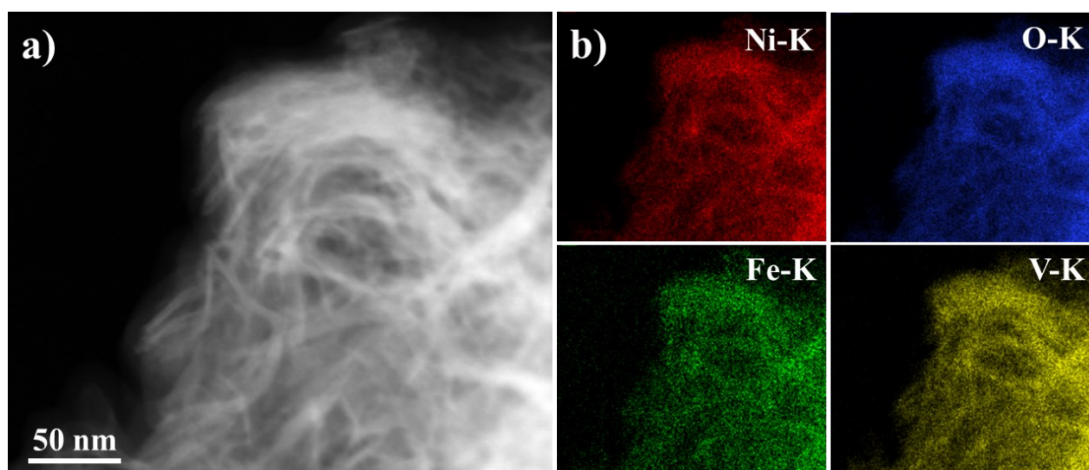
**Figure S1.** SEM image (a), EDS pattern (b), and corresponding elemental mapping images (c) of O-NiFeV-LDH.



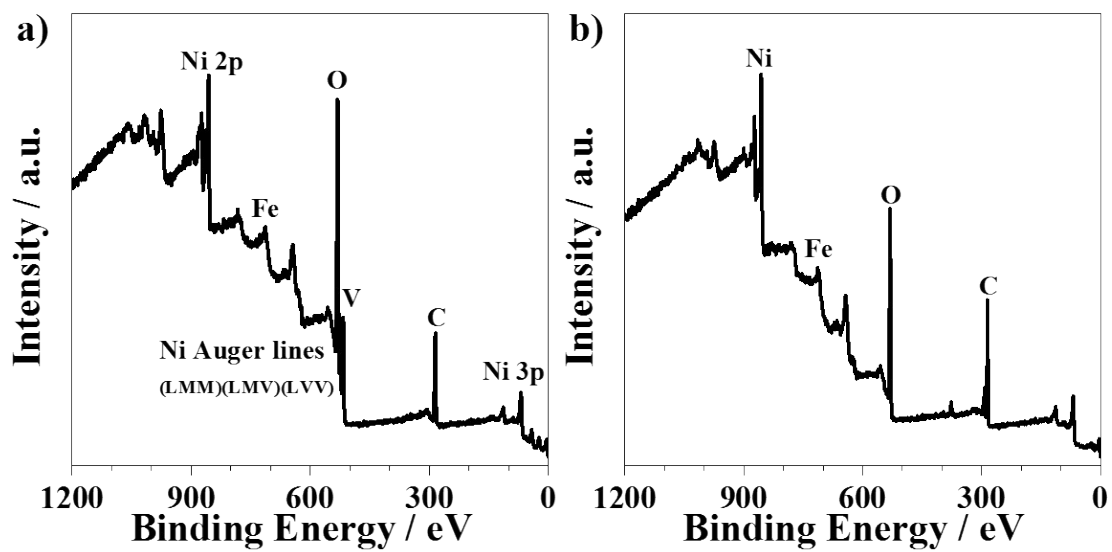
**Figure S2.** SEM image (a), EDS pattern (b), and corresponding elemental mapping images (c) of A-NiFeV-LDH.



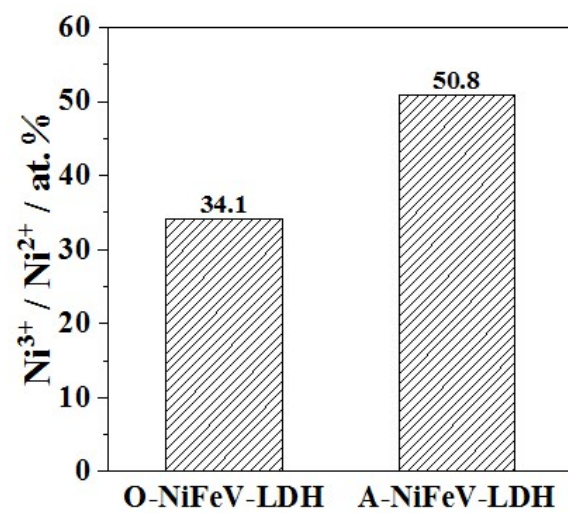
**Figure S3.** Representative AFM image (a) and corresponding line scan profile (b) of one O-NiFeV-LDH sheet deposited on a mica substrate.



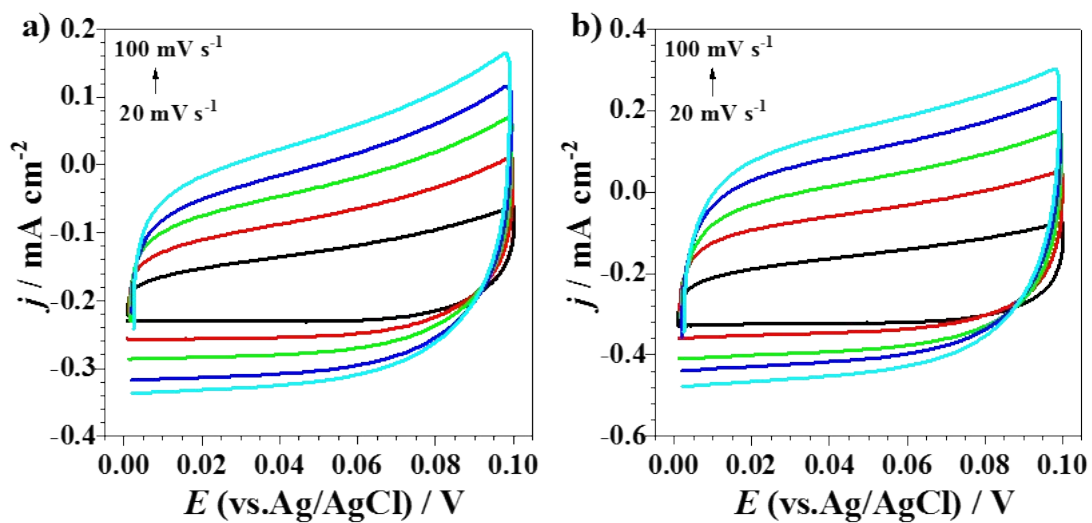
**Figure S4.** HAADF image (a) and corresponding STEM-EDS elemental mapping (Ni, O, Fe, and V) (b) of O-NiFeV-LDH.



**Figure S5.** Survey XPS spectra of O-NiFeV-LDH (a) and A-NiFeV-LDH (b).

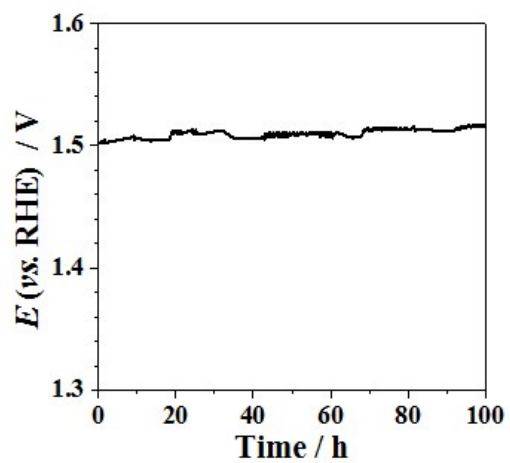


**Figure S6.** The valence state ratio of nickel ( $\text{Ni}^{3+} / \text{Ni}^{2+}$ ) of O-NiFeV-LDH and A-NiFeV-LDH.

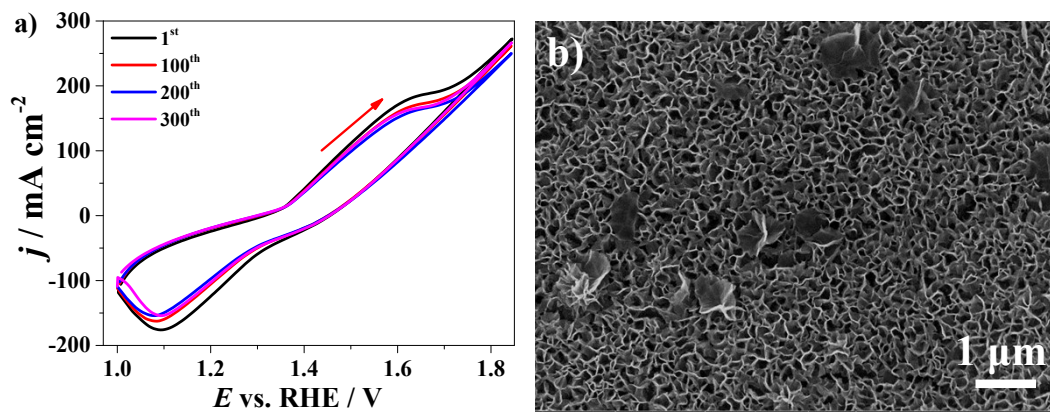


**Figure S7.** CV curves in the double layer region with various scan rates from 20 to 100  $\text{mV s}^{-1}$  for O-NiFeV-LDH (a) and A-NiFeV-LDH (b).

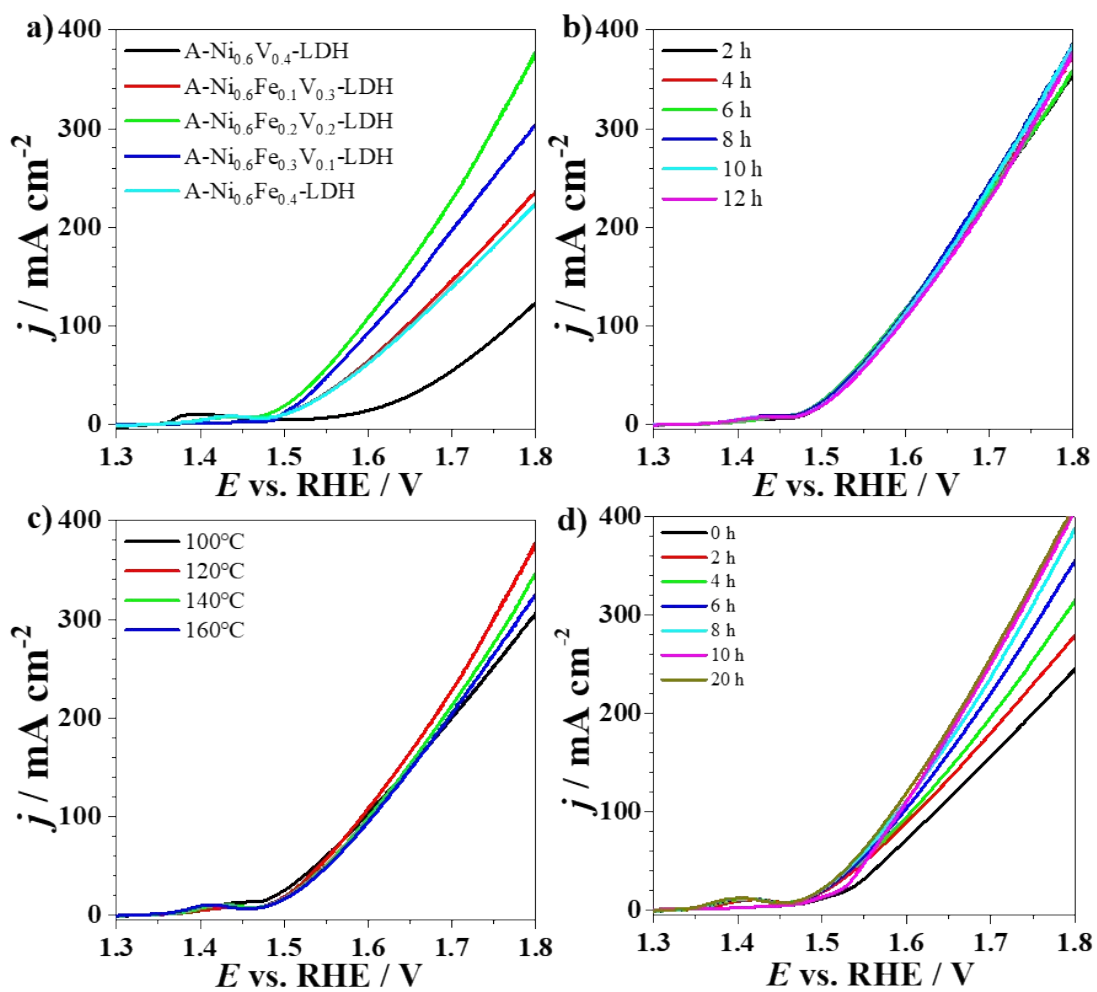




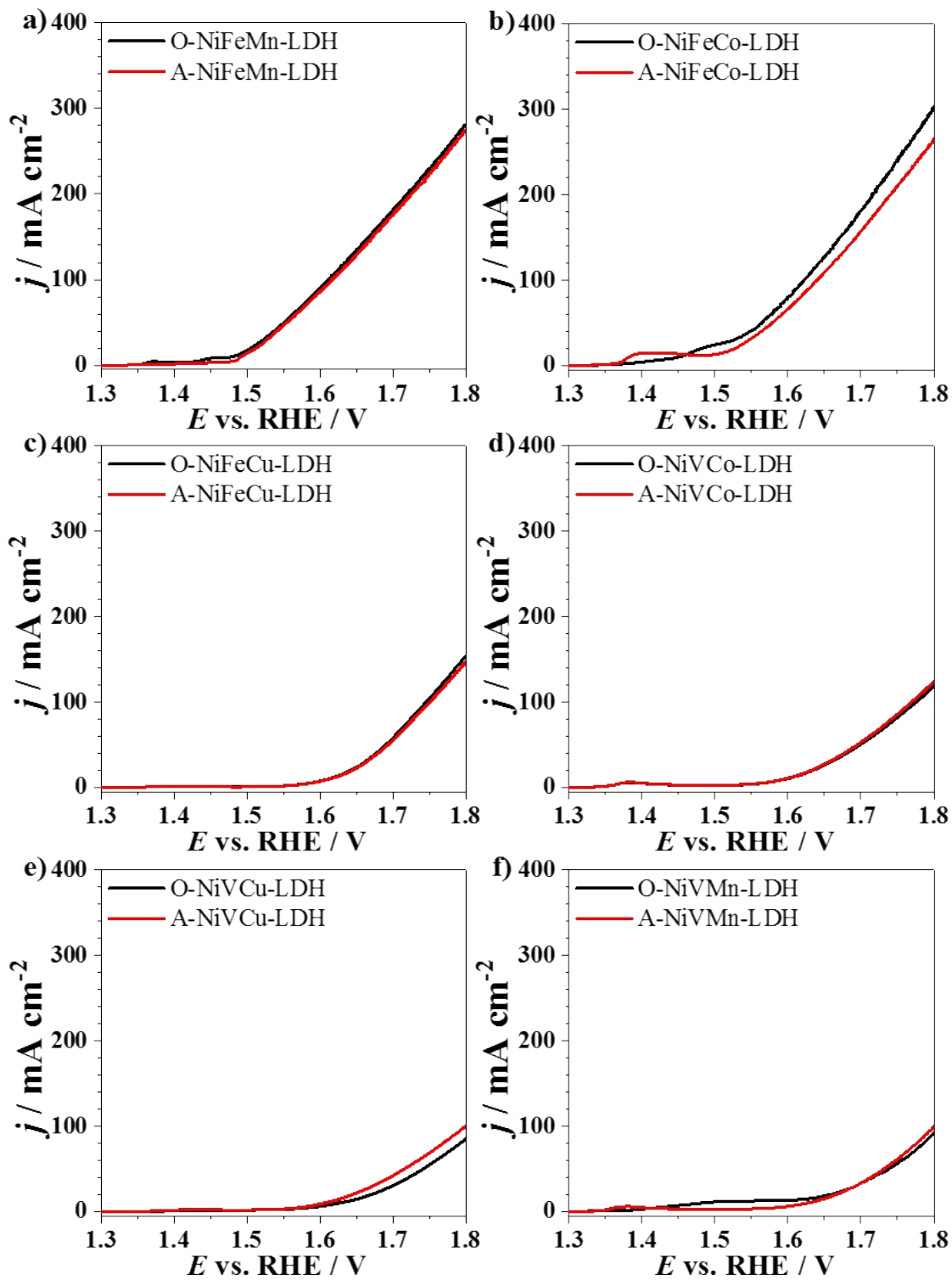
**Figure S8.** Chronopotentiometry curves of A-NiFeV-LDH during electrolysis at constant current densities of  $10 \text{ mA cm}^{-2}$  in  $1.0 \text{ M KOH}$ .



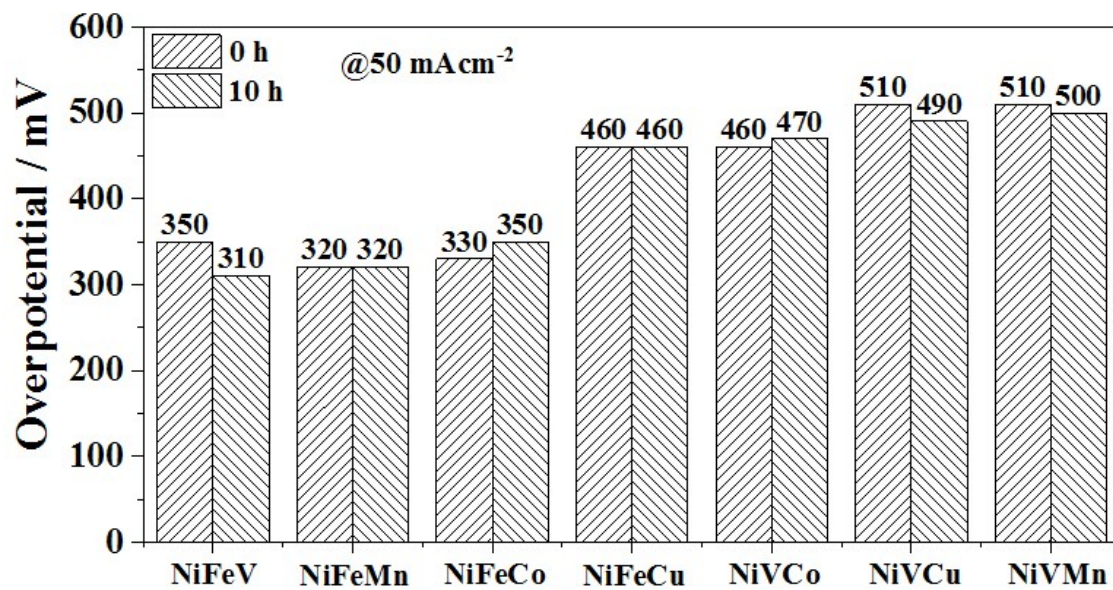
**Figure S9.** (a) 1<sup>st</sup> - 300<sup>th</sup> CV curves of A-NiFeV-LDH at a scan rate of 50  $\text{mV s}^{-1}$  in 1.0 M KOH. (b) SEM image of A-NiFeV-LDH after 300 consecutive CV scans.



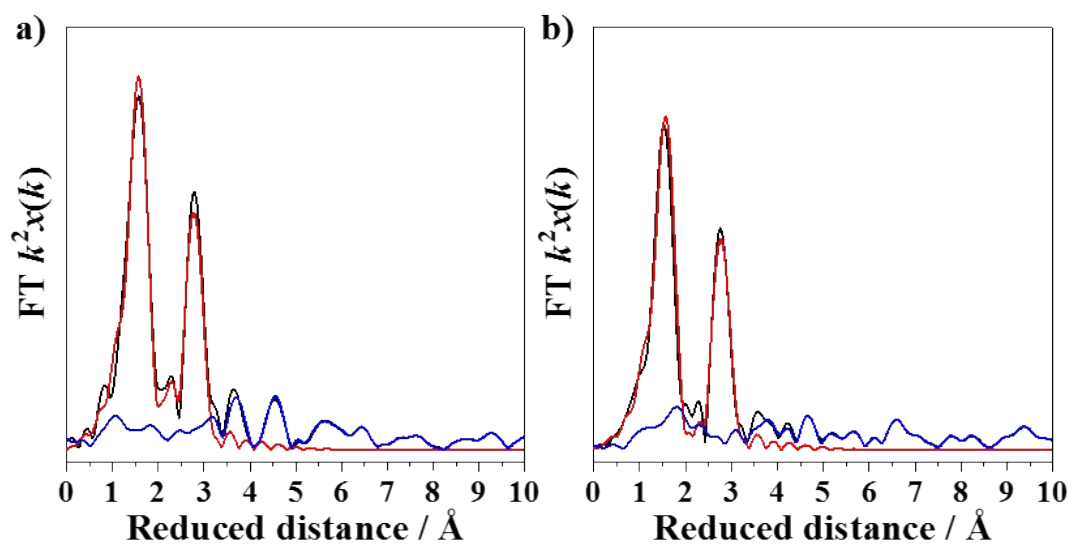
**Figure S10.** The influence of  $\text{Fe}^{3+}$  and  $\text{V}^{3+}$  concentrations (a), hydrothermal time (b), hydrothermal temperature (c), and activation time (d) on the LSV performance of A-NiFeV-LDH.



**Figure S11.** LSV curves of Ni-based LDHs before and after activation for 10 h: NiFeMn (a), NiFeCo (b), NiFeCu (c), NiVCo (d), NiVCu (e), and NiVMn (f).



**Figure S12.** The overpotential at a current density of  $50 \text{ mA cm}^{-2}$  for NiFeMn, NiFeCo, NiFeCu, NiVCo, NiVCu, and NiVMn before and after activation for 10 h.



**Figure S13.** FT-EXAFS spectra and fits of Fe K-edges from O-NiFeV-LDH (a), and A-NiFeV-LDH (b). The red and blue lines represent the fitting values. Co K edge XANES spectra.

**Table S1.** Summary of the fitting parameters of Fe K-edge EXAFS curves for the NiFeV-LDH and A-NiFeV-LDH.

Sample	Fe-O		Fe-Fe		D. W.	$\Delta E_0$ (eV)
	R (Å)	CN	R (Å)	CN		
O-NiFeV-LDH	$2.01 \pm 0.007$	$6.8 \pm 0.3$	$2.60 \pm 0.03$	$0.9 \pm 0.3$	0.006 (O)	$-0.7 \pm 0.7$
			$3.24 \pm 0.01$	$4.7 \pm 0.5$		
A-NiFeV-LDH	$2.002 \pm 0.007$	$6.1 \pm 0.2$	$2.58 \pm 0.04$	$0.5 \pm 0.2$	0.008 (Fe)	$4.0 \pm 1.3$
			$3.23 \pm 0.01$	$4.2 \pm 0.4$		

Fe K edge (7112 eV)

Amp:  $0.73 \pm 0.07$  (Fe foil)