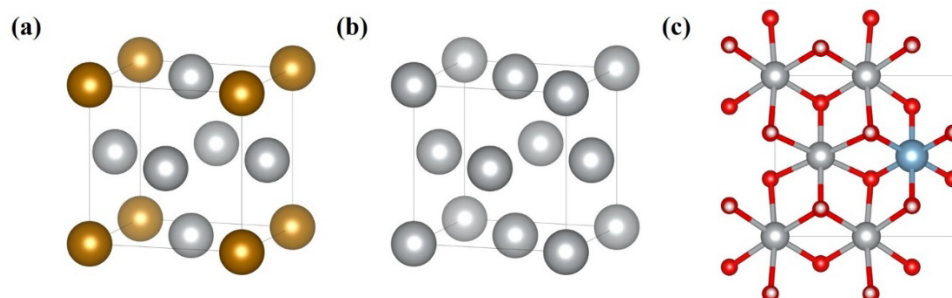
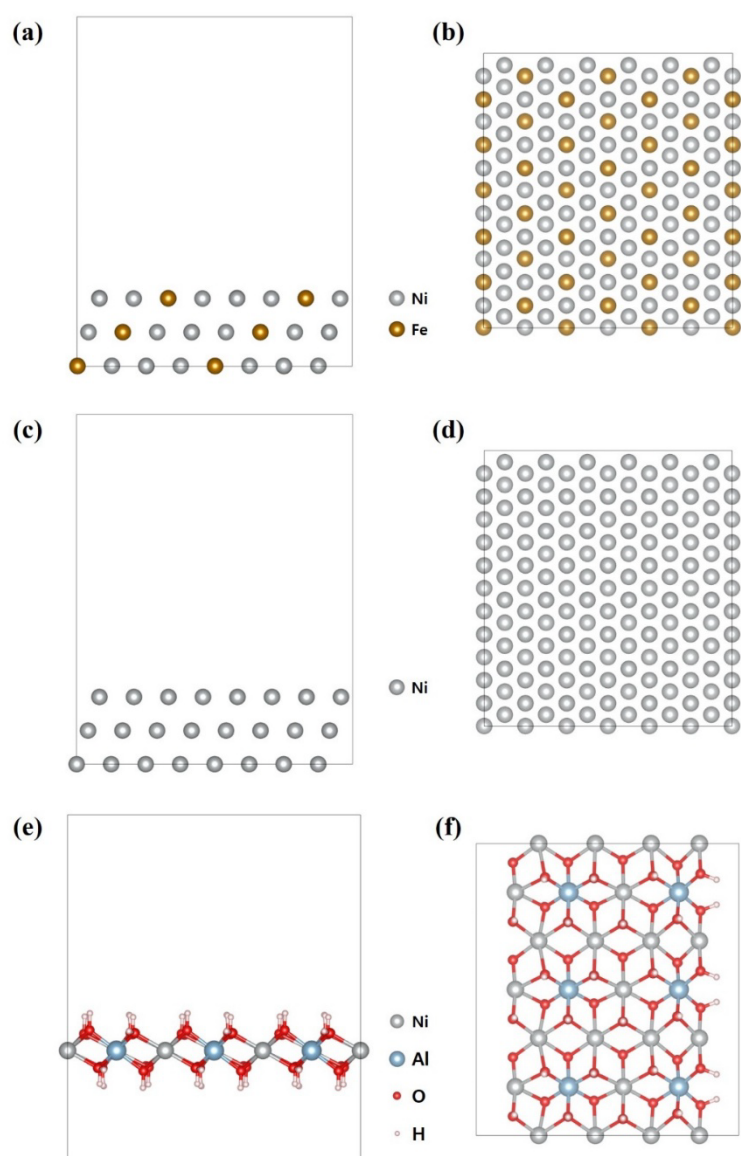


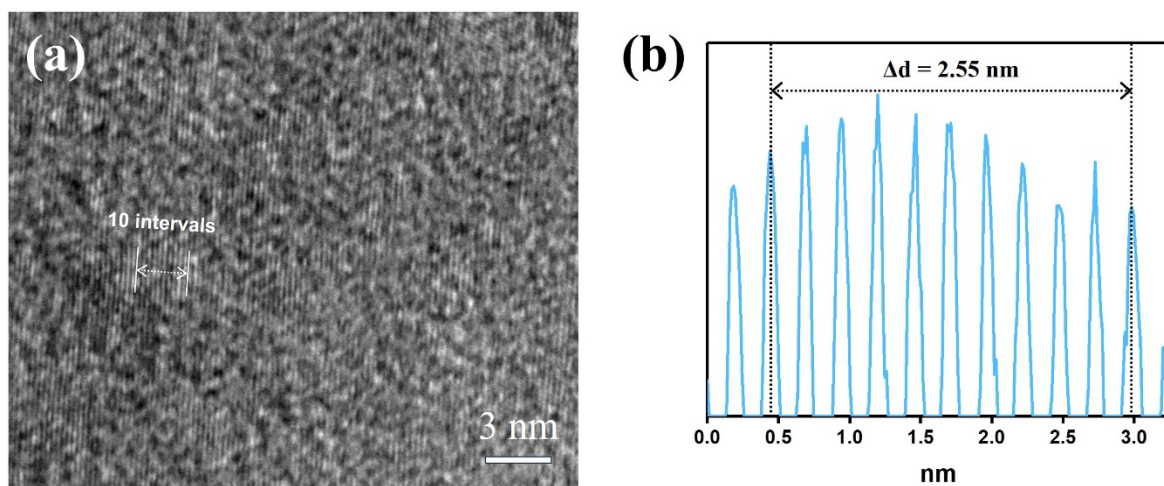
## Supplementary Materials



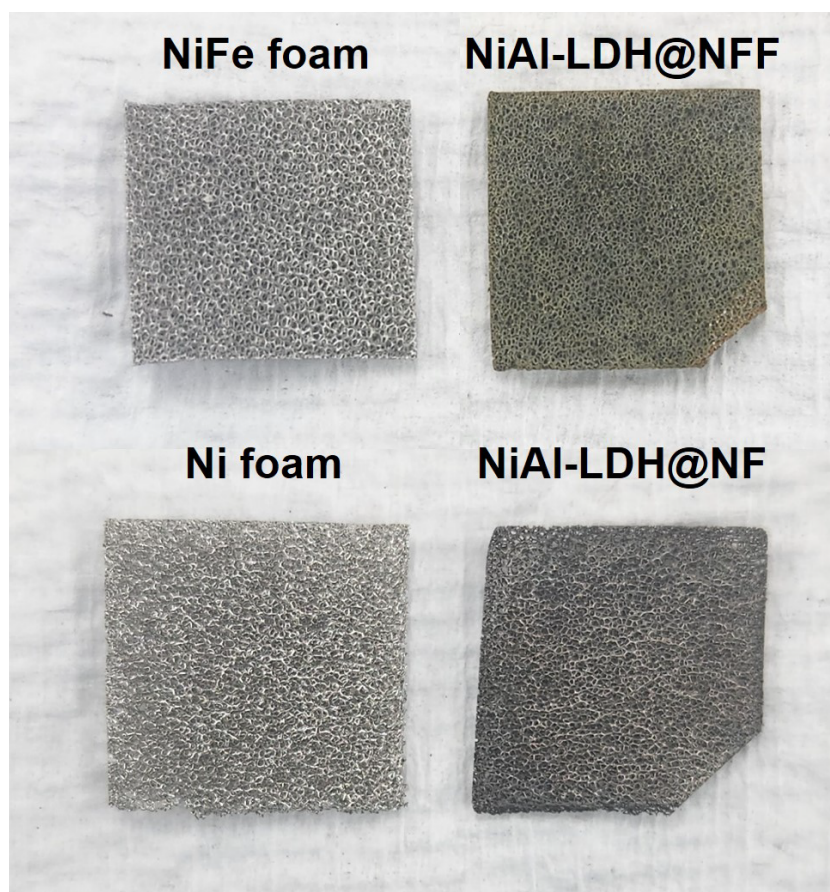
**Figure S1.** Bulk structures of (a)  $\text{FeNi}_3$ , (b)  $\text{Ni}$ , and (c)  $\text{NiAl-LDH}$ .



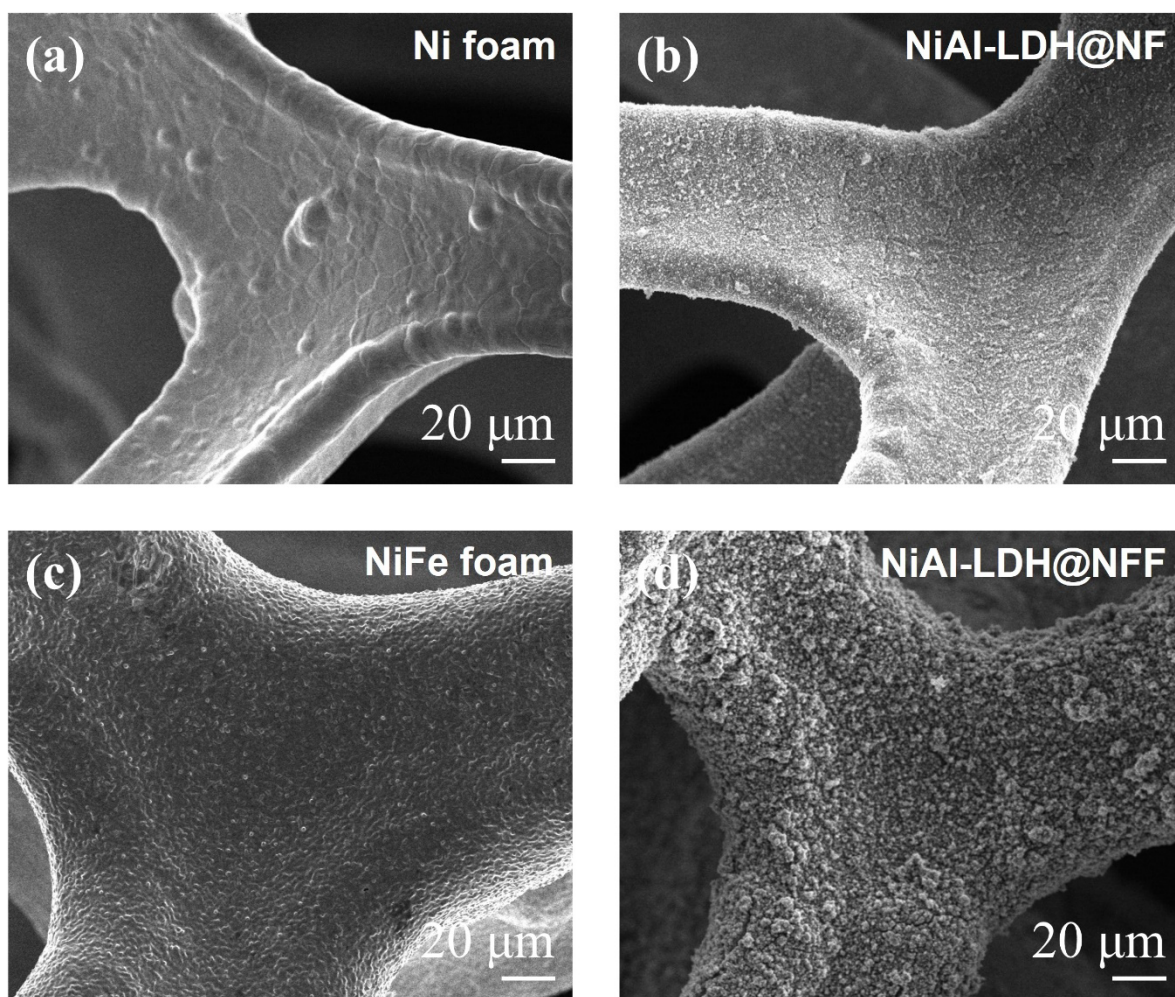
**Figure S2.** Optimized structures of  $\text{NiFe}(111)$ ,  $\text{Ni}(111)$ , and  $\text{NiAl-LDH}(001)$  slabs in (a, c, e) side views and (b, d, f) top views.



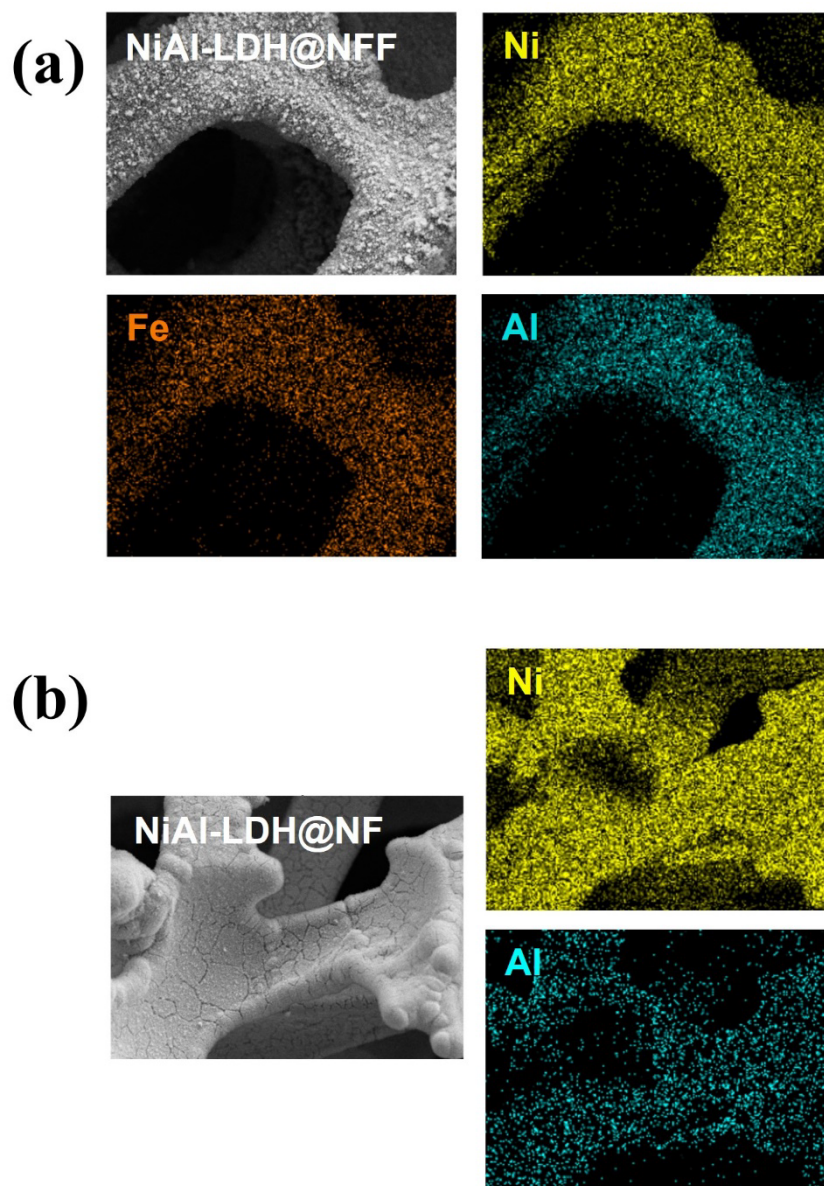
**Figure S3.** (a) High resolution TEM image and (b) line profile (measurement at 10 intervals) of NiAl-LDH@NFF.



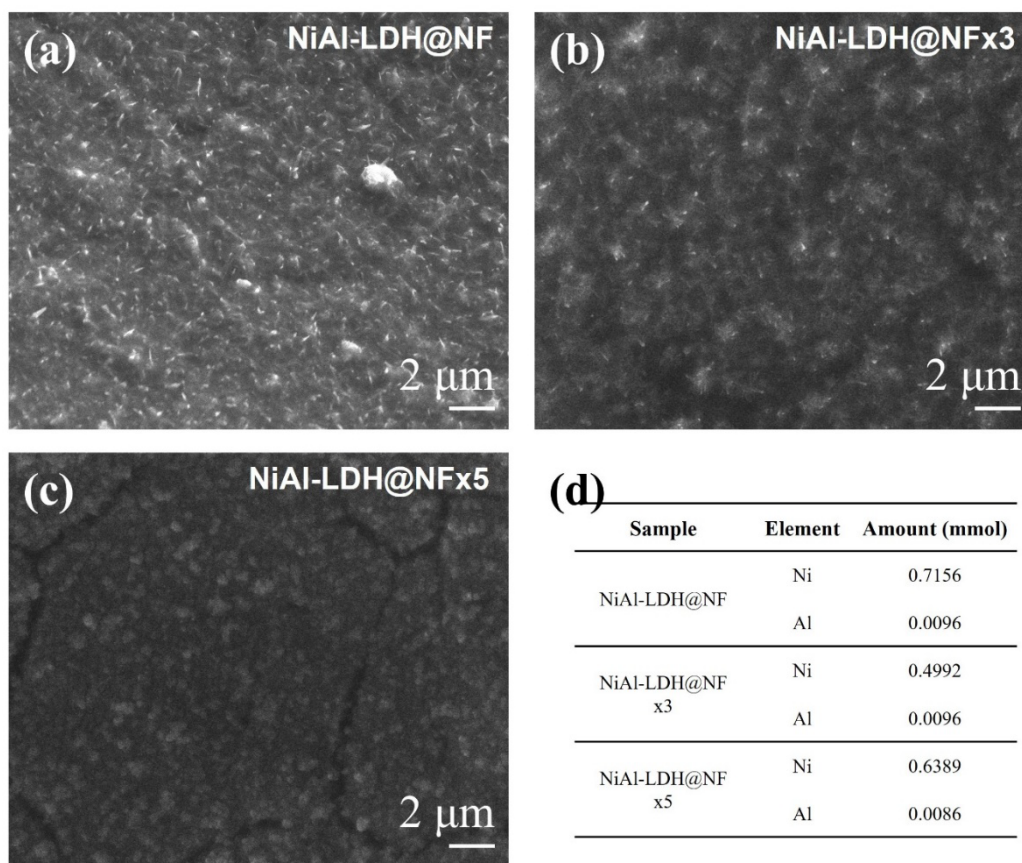
**Figure S4.** Photographs of NFF, NF, NiAl-LDH@NFF, and NiAl-LDH@NF.



**Figure S5.** SEM images (×1,000) of (a) NF, (b) NiAl-LDH@NF, (c) NFF, and (d) NiAl-LDH@NFF.



**Figure S6.** SEM images and EDS mappings (yellow: Ni, orange: Fe, cyan: Al) of (a) NiAl-LDH@NFF and (b) NiAl-LDH@NF.



**Figure S7.** SEM images ( $\times 10,000$ ) of (a) NiAl-LDH@NF, (b) NiAl-LDH@NFx3, and (c) NiAl-LDH@NFx5. (d) Table showing the Ni to Al molar ratio.

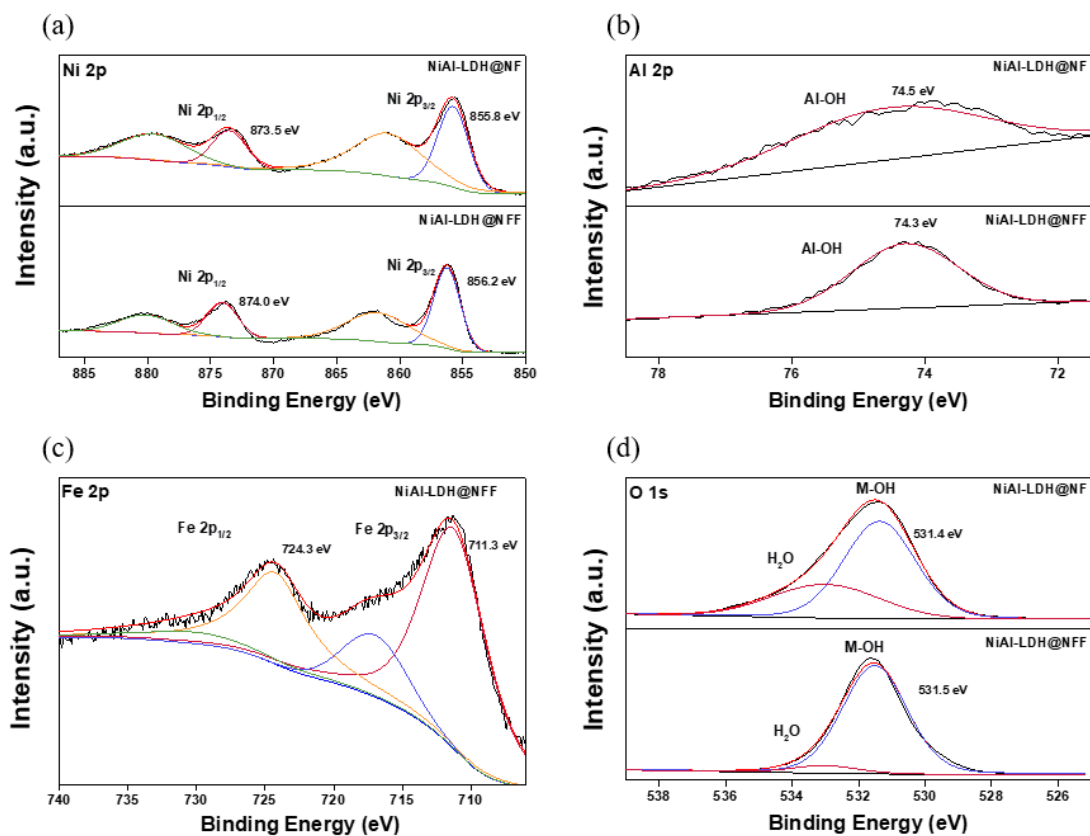


Figure S8. XPS spectra of (a) Ni 2p (b) Al 2p (c) Fe 2p (d) O 1s

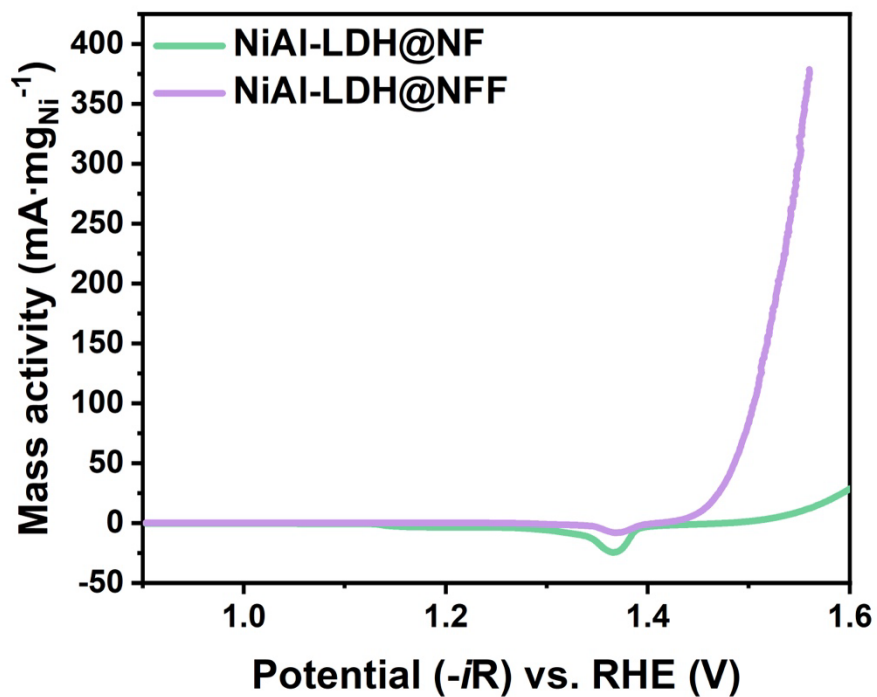
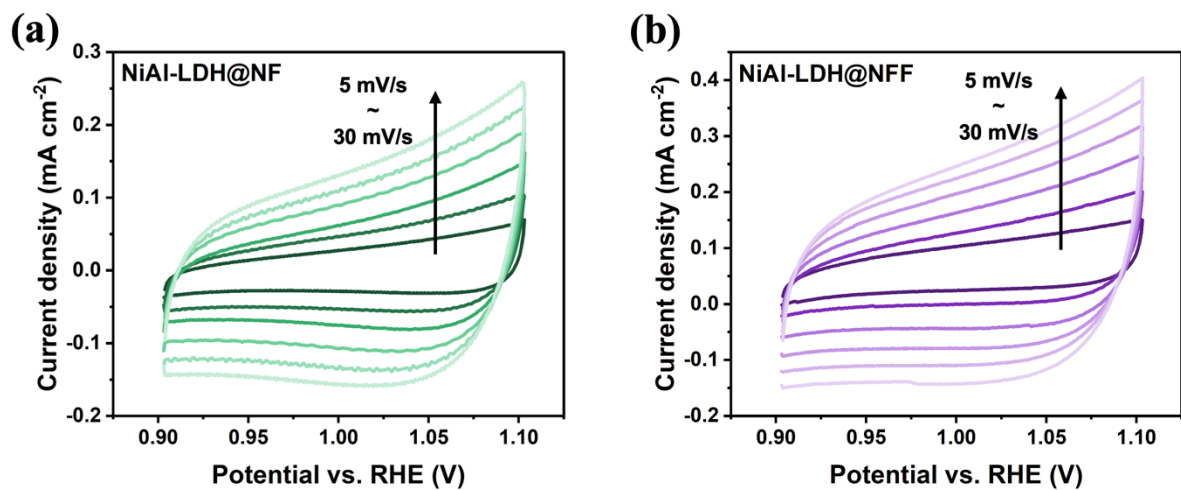
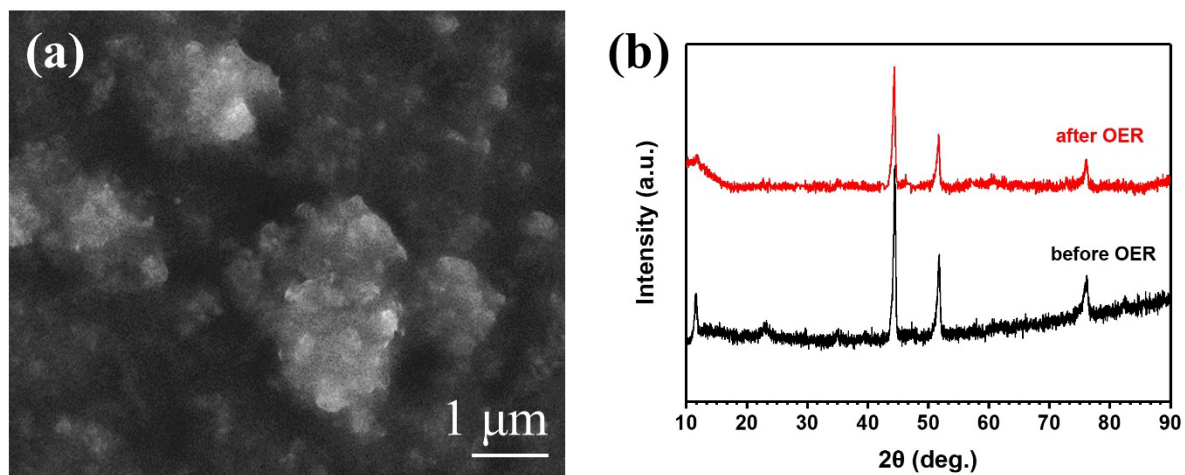


Figure S9. OER mass activity of NiAl-LDH@NF and NiAl-LDH@NFF.



**Figure S10.** CV curves of NiAl-LDH@NF and NiAl-LDH@NFF were measured with a 5 mV interval.



**Figure S11.** (a) SEM image of NiAl-LDH@NFF after OER test and (b) comparison of NiAl-LDH@NFF XRD patterns before and after OER.