

## Supporting Information

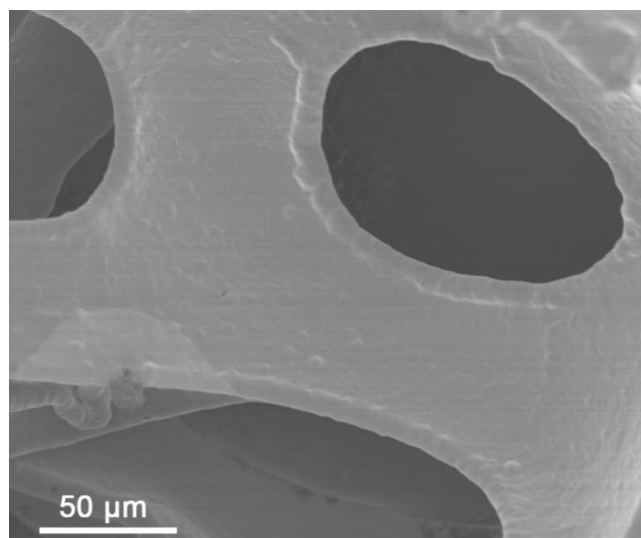
### **Sequentially Electrodeposited Fe<sub>6</sub>Ni/FeNi<sub>6</sub> NiFe Hydroxide/LDH-Like Double-Layer Electrode for Efficient Oxygen Evolution Reaction**

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Li<sup>a</sup>, Yiqing Jiao<sup>a</sup>, Zhihao Fang<sup>a,c\*</sup>, Yongwei Li<sup>a</sup> and Yan Zhou<sup>a</sup>*

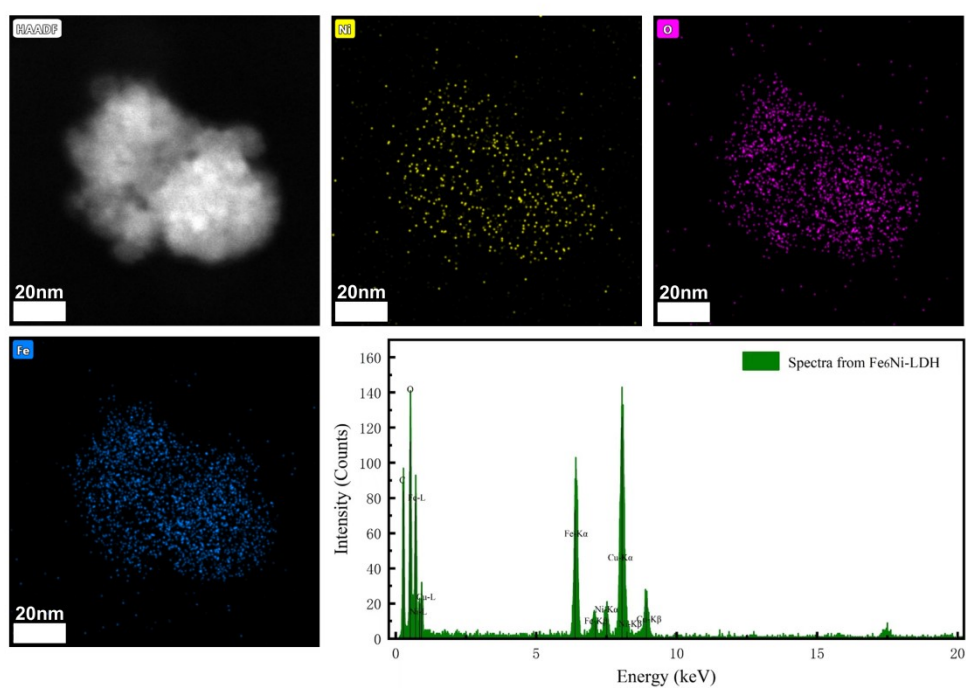
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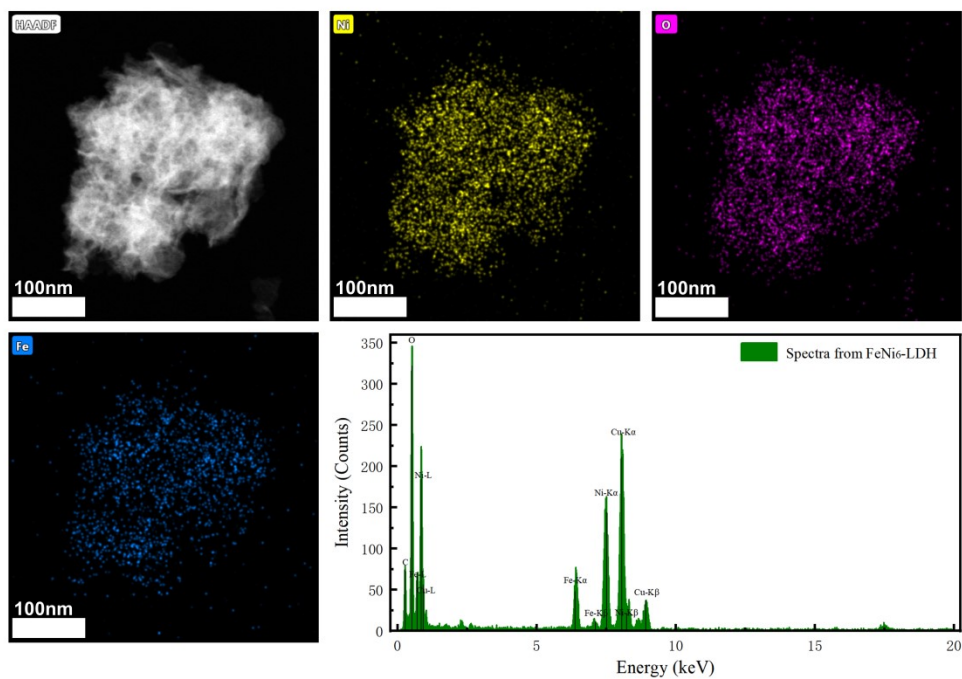
<sup>c</sup> Shandong Solid New Material Technology Co.,Ltd, Weifang 262700, China



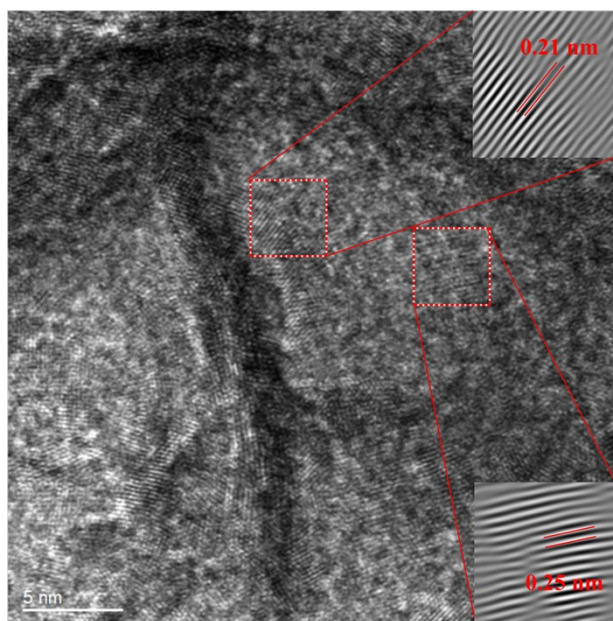
**Figure S1** SEM image of blank NF.



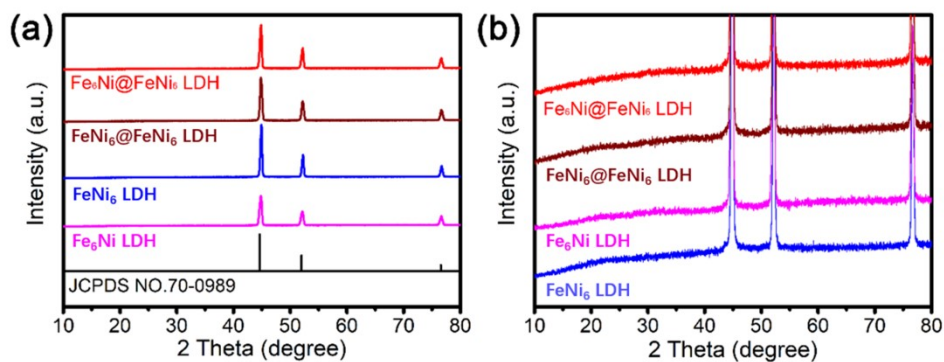
**Figure S2** Plan-view TEM image and corresponding EDX elemental mapping of the as-prepared Fe<sub>6</sub>Ni sample.



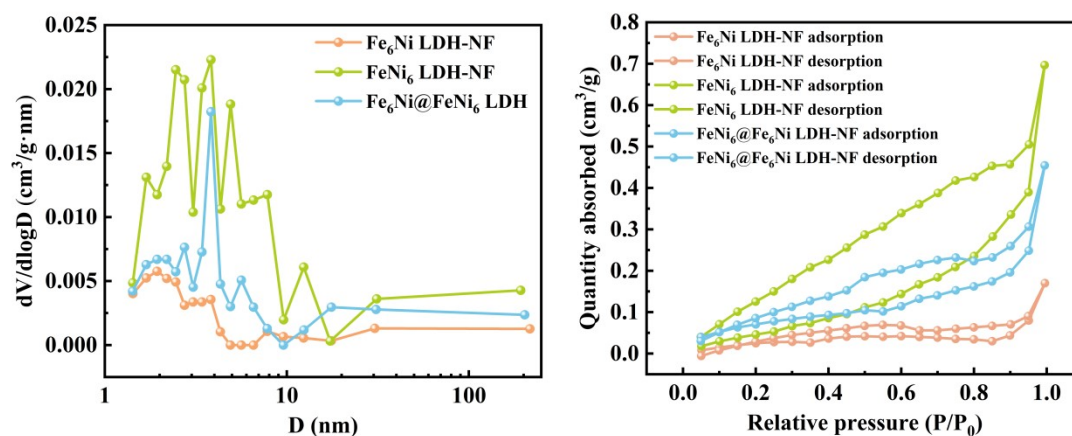
**Figure S3** A typical TEM EDX-mapping of as-prepared FeNi<sub>6</sub> LDH.



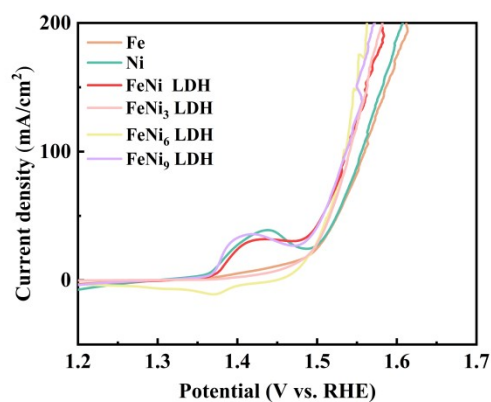
**Figure S4** HRTEM image of Fe<sub>6</sub>Ni@FeNi<sub>6</sub> LDH showing locally resolved lattice fringes with interplanar spacings of approximately 0.25 and 0.21 nm, consistent with the (012) and (015) planes of NiFe LDH.



**Figure S5** XRD patterns of the electrodeposited NiFe hydroxide/LDH-like samples on Ni foam.

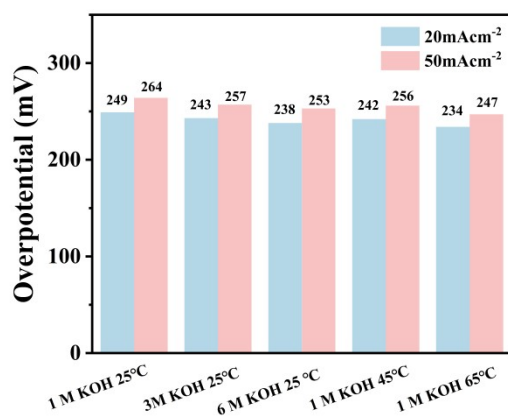


**Figure S6** Nitrogen adsorption-desorption isotherms and pore size distribution curves of NiFe LDH catalysts on Ni foam.

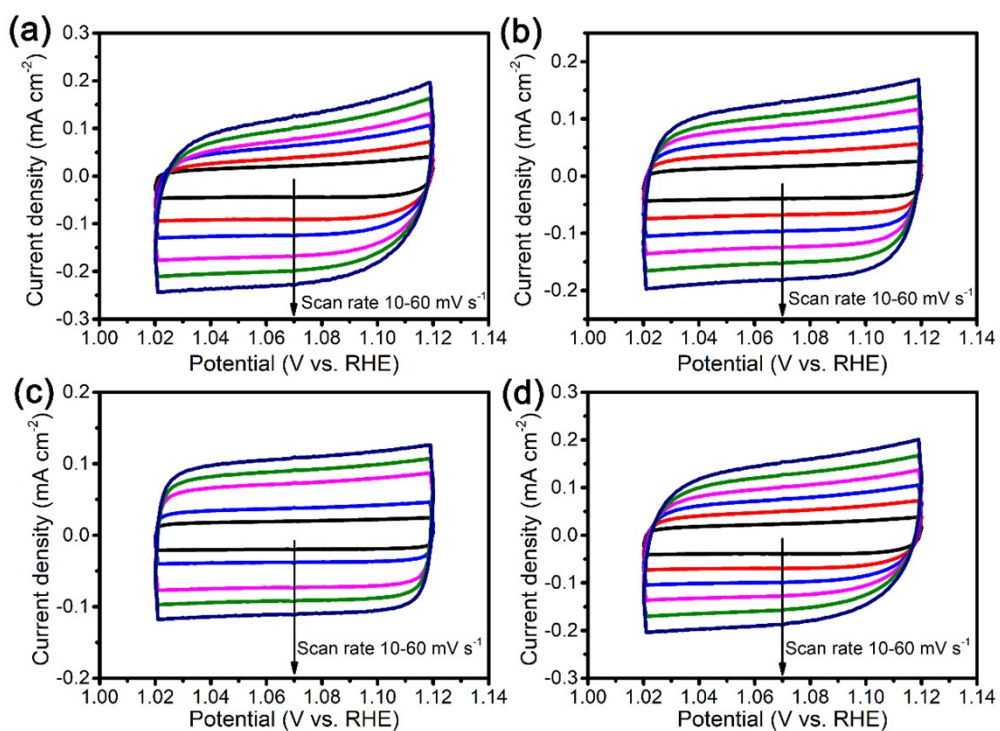


**Figure S7** OER performance in 1 M KOH: LSV curves comparing single-layer and double-layer

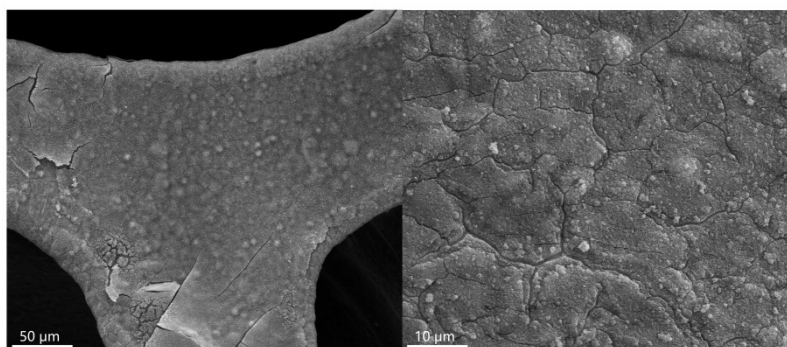
deposited FeNi LDH materials.



**Figure S8** Comparison of the overpotentials of Fe<sub>6</sub>Ni@FeNi<sub>6</sub> LDH required to reach current densities of 20 and 50 mA cm<sup>-2</sup> under different alkaline concentrations and temperatures.

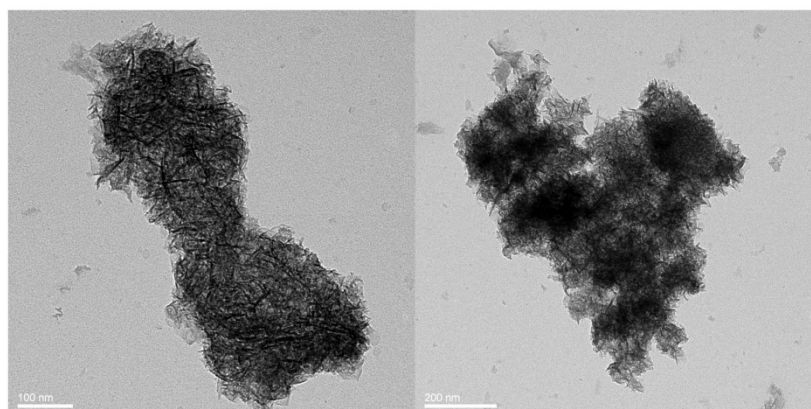


**Figure S9** CVs of FeNi<sub>6</sub> LDH, Fe<sub>6</sub>Ni LDH, FeNi<sub>6</sub>@FeNi<sub>6</sub> LDH and Fe<sub>6</sub>Ni@FeNi<sub>6</sub> LDH.



**Figure S10** SEM images of Fe<sub>6</sub>Ni@FeNi<sub>6</sub> LDH after 24 h chronopotentiometric OER testing at

20 mA cm<sup>-2</sup> in 1 M KOH.



**Figure S11** TEM images of Fe<sub>6</sub>Ni@FeNi<sub>6</sub> LDH after 24 h chronopotentiometric OER testing at 20 mA cm<sup>-2</sup> in 1 M KOH.

**Table S1** BET surface area and porosity parameters of NiFe LDH catalysts.

Sample	Specific surface area (m <sup>2</sup> /g)	Total pore volume (cm <sup>3</sup> /g)	Average pore diameter (nm)
Fe <sub>6</sub> Ni LDH-NF	15.335	0.017	2.454
FeNi <sub>6</sub> LDH-NF	14.314	0.004	1.690
Fe <sub>6</sub> Ni@FeNi <sub>6</sub> LDH	8.876	0.009	1.690

**Table S2** Comparison of OER performance of representative NiFe-based electrocatalysts in 1 M KOH under ambient conditions.

Materials	OER (mV)	Electrolyte	Current density (mAcm <sup>-2</sup> )	Ref.
FeNi	313	1 M KOH	20	[1]
Pd/NiFe-LDH-u	267	1 M KOH	20	[2]
NiFe-LDH/CNTs	269	1 M KOH	20	[3]
RuO <sub>2</sub> /NiFe-LDH/NF	319	1 M KOH	50	[4]
Sulfated NiFe LDH	288	1 M KOH	50	[5]
Ni/Fe <sub>3</sub> O <sub>4</sub> /NiFe LDH	275	1 M KOH	20	[6]
Co <sub>1.98</sub> -NiFe LDH	266	1 M KOH	50	[7]
Co <sub>3</sub> O <sub>4</sub> @NiFe LDH	270	1 M KOH	50	[8]
NiCo <sub>2</sub> O <sub>4</sub> @NiFe-LDH-2	270	1 M KOH	50	[9]
NiFe hydroxide	307	1 M KOH	50	[10]

Fe <sub>6</sub> Ni@FeNi <sub>6</sub> LDH	249	1 M KOH	20	This work
Fe <sub>6</sub> Ni@FeNi <sub>6</sub> LDH	264	1 M KOH	50	This work

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