

## Electronic Supplementary Information

$\pi\cdots\pi$  interaction directed 2D FeNi-LDHs nanosheets from 2D Hofmann-MOFs for oxygen evolution reaction

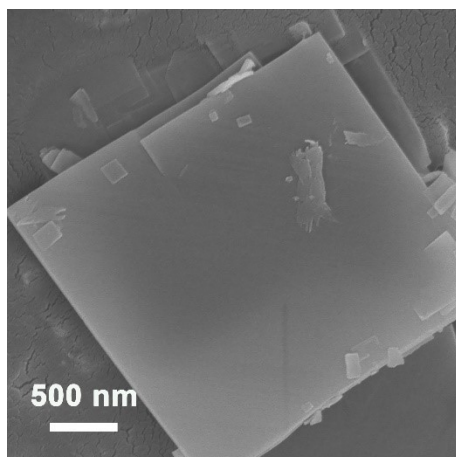
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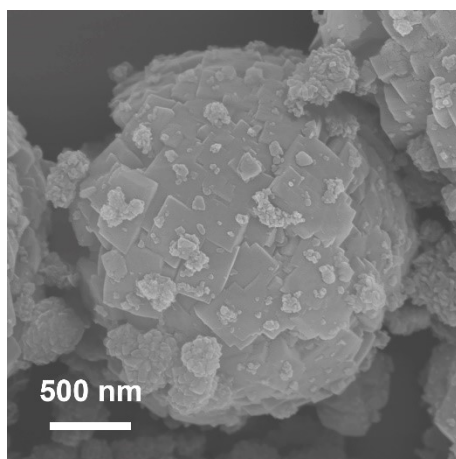
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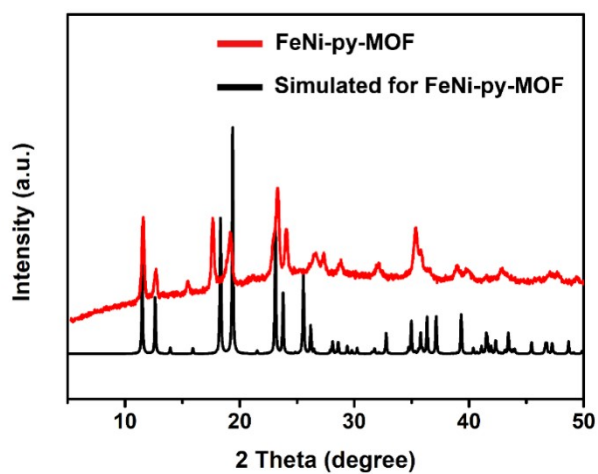
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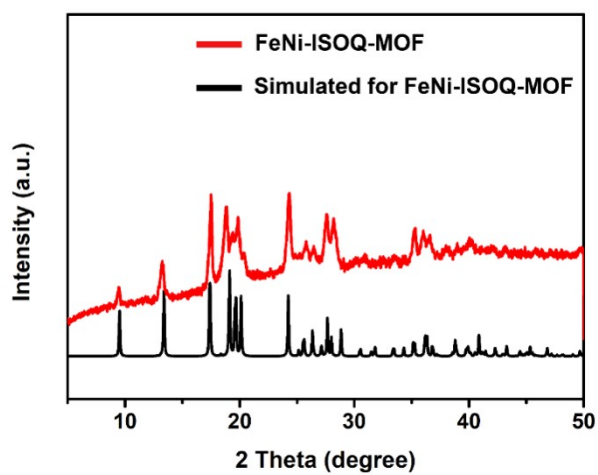
**Figure S1.** SEM image of FeNi-py-MOF precursor.



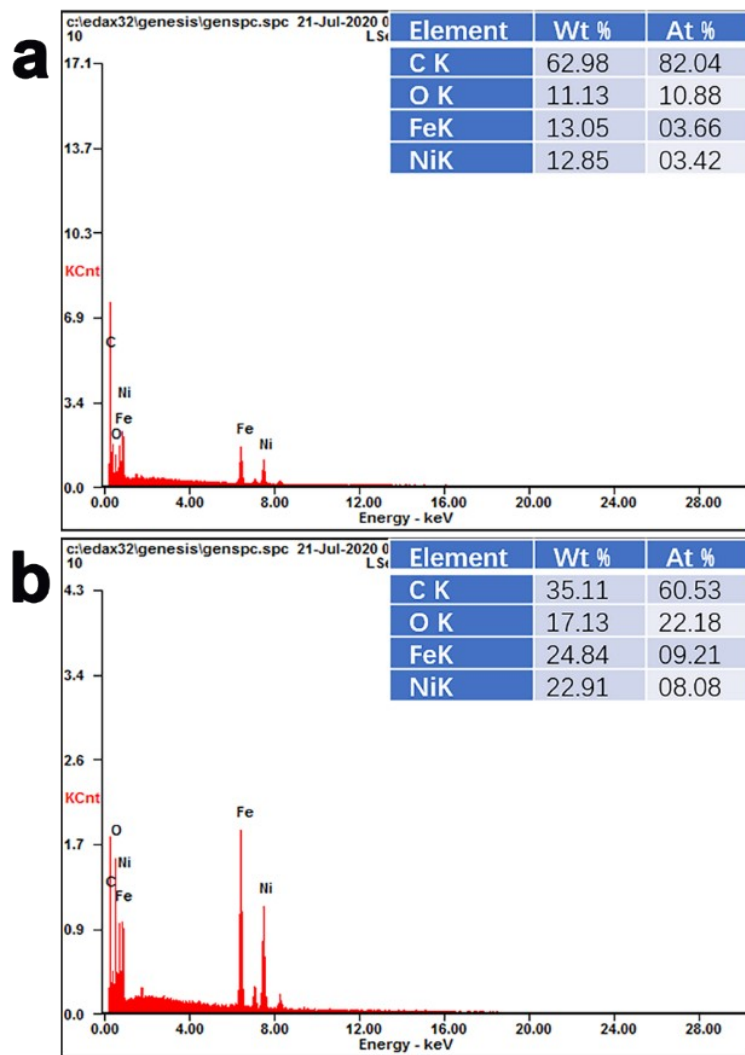
**Figure S2.** SEM image of FeNi-ISOQ-MOF precursor.



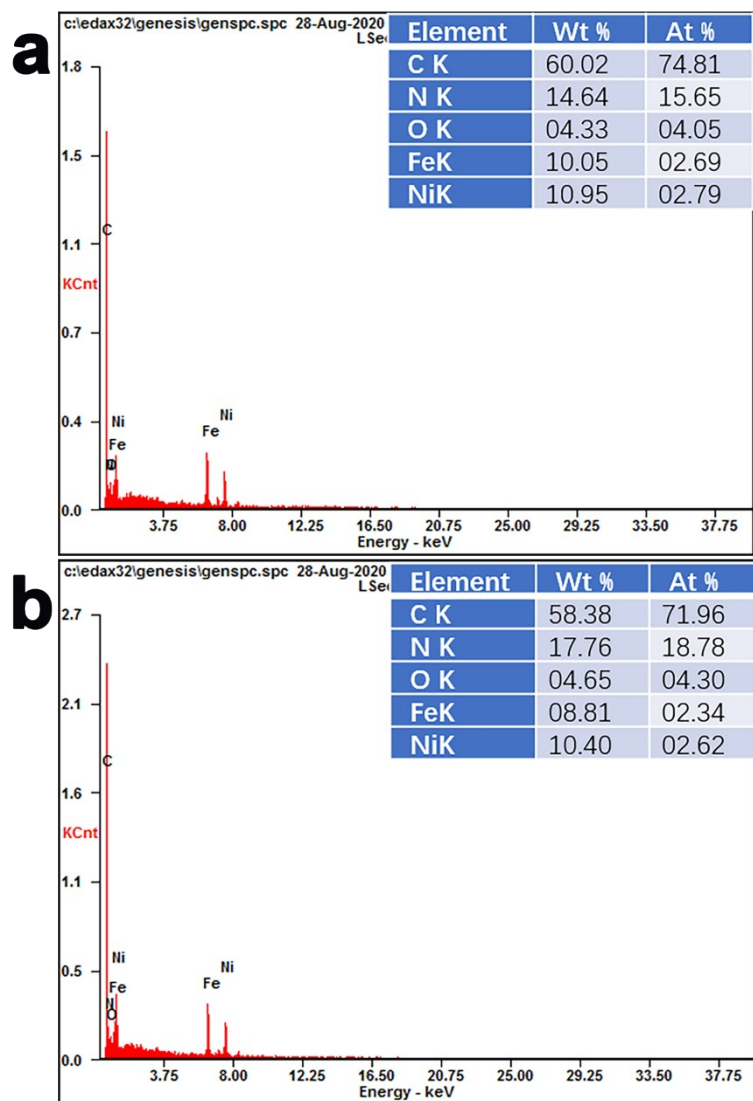
**Figure S3.** XRD pattern of FeNi-py-MOF precursor.



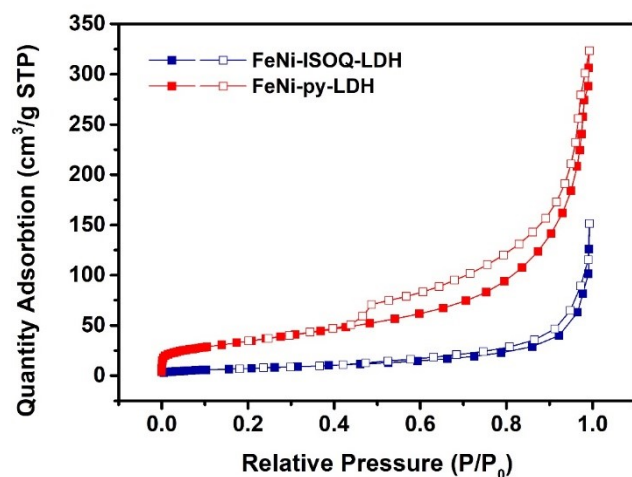
**Figure S4.** XRD pattern of FeNi-ISOQ-MOF precursor.



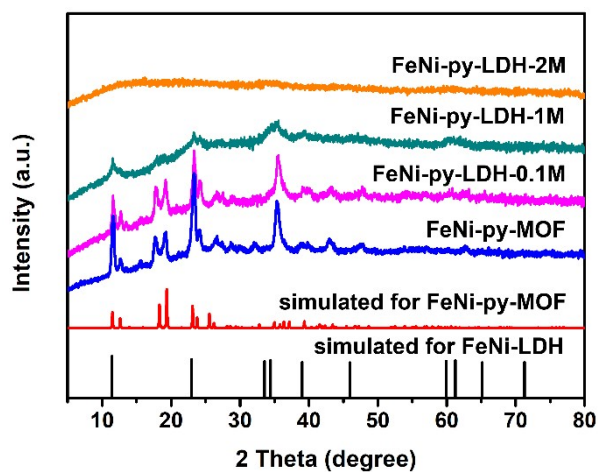
**Figure S5.** EDX patterns of (a) FeNi-py-MOF and (b) FeNi-ISOQ-MOF precursors.



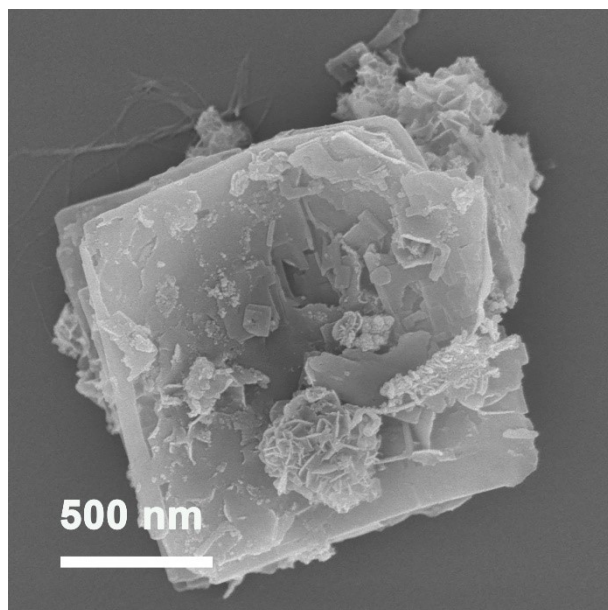
**Figure S6.** EDX patterns of (a) FeNi-py-LDH and (b) FeNi-ISOQ-LDH.



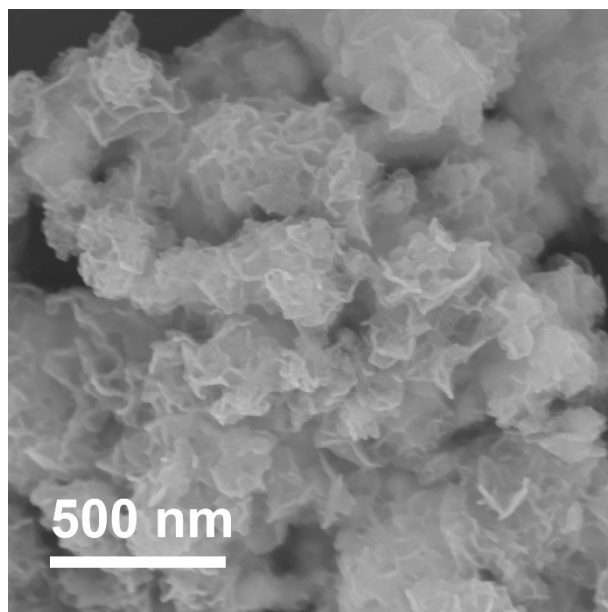
**Figure S7.** N<sub>2</sub> adsorption and desorption isotherms of FeNi-py-LDH and FeNi-ISOQ-LDH at 77 K.



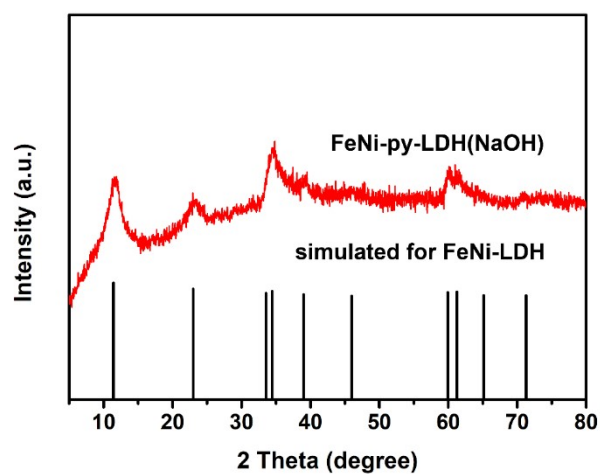
**Figure S8.** XRD patterns of FeNi-py-LDH-0.1M, FeNi-py-LDH-1M and FeNi-py-LDH-2M.



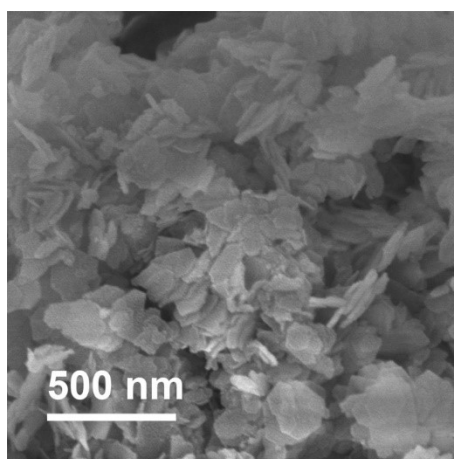
**Figure S9.** SEM image of FeNi-py-LDH-0.1M.



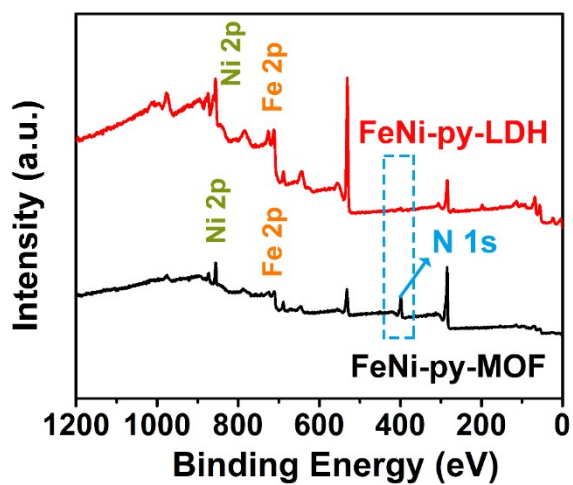
**Figure S10.** SEM image of FeNi-py-LDH-2M.



**Figure S11.** XRD patterns of FeNi-py-LDH(NaOH) and the simulated FeNi-LDH.

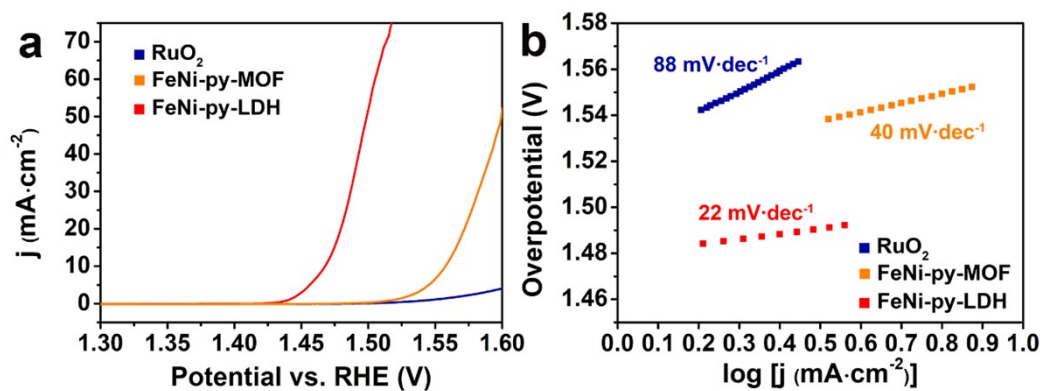


**Figure S12.** SEM image of FeNi-py-LDH(NaOH).

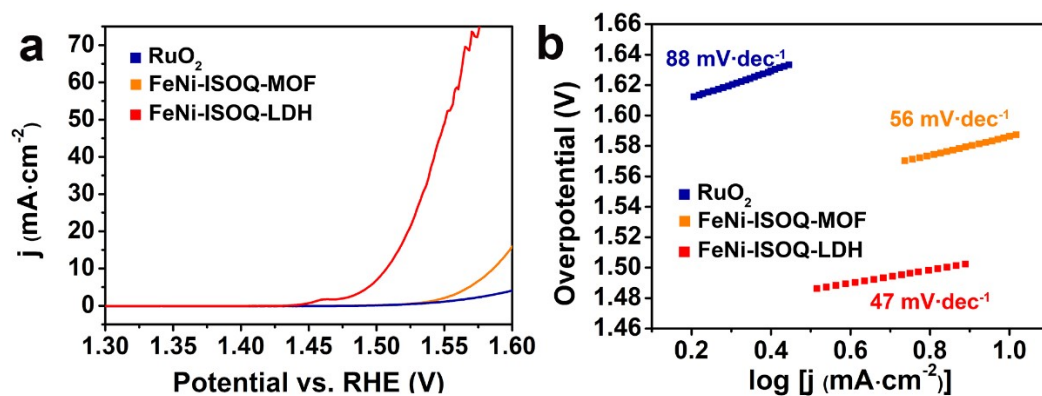


**Figure S13.** Survey XPS spectra of FeNi-py-MOF and FeNi-py-LDH.

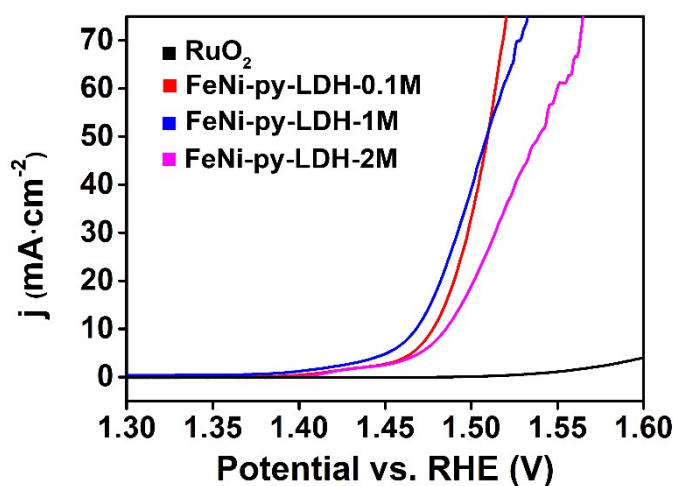




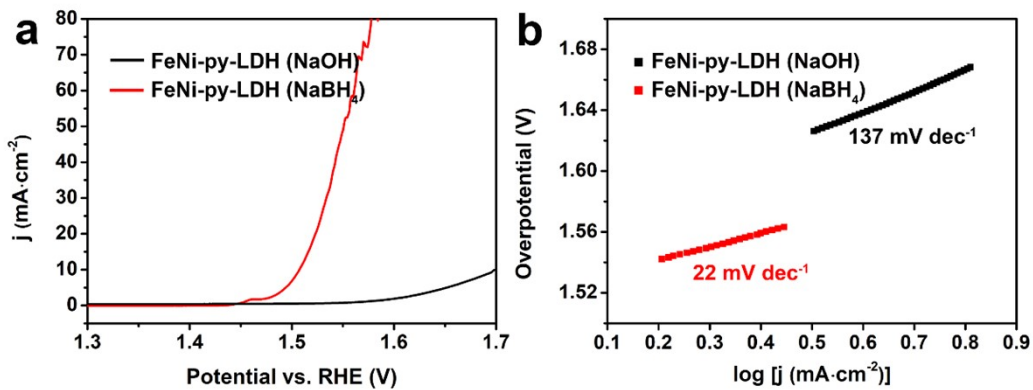
**Figure S14.** (a) The LSV curves of FeNi-py-LDH, FeNi-py-MOF and RuO<sub>2</sub>. (b) The Tafel slopes of FeNi-py-LDH, FeNi-py-MOF and RuO<sub>2</sub>.



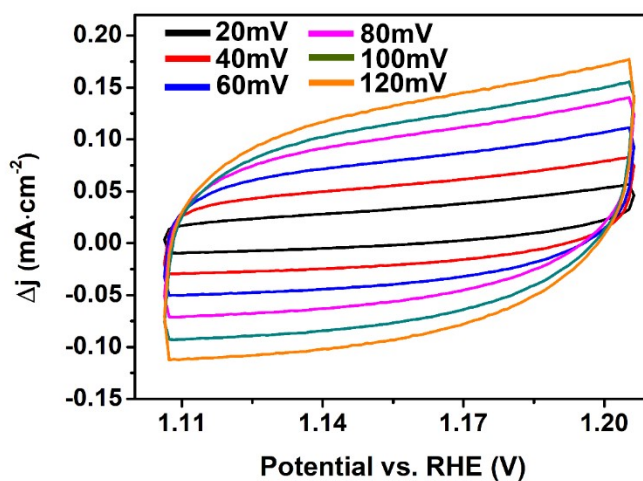
**Figure S15.** (a) The LSV curves of FeNi-ISOQ-LDH, FeNi-ISOQ-MOF and RuO<sub>2</sub>. (b) The Tafel slopes of FeNi-ISOQ-LDH, FeNi-ISOQ-MOF and RuO<sub>2</sub>.



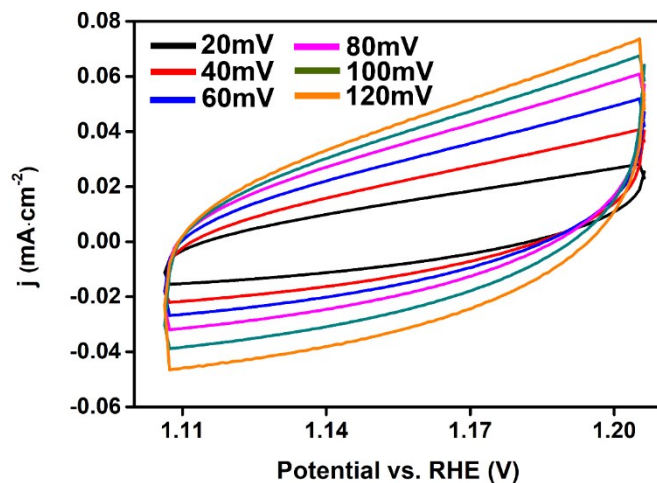
**Figure S16.** The LSV curves of FeNi-py-LDH-0.1M, FeNi-py-LDH-1M and FeNi-py-LDH-2M.



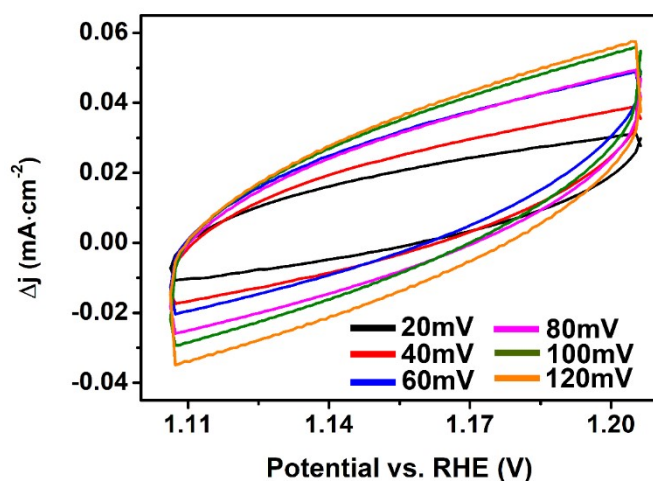
**Figure S17.** (a) The LSV curves of FeNi-py-LDH(NaOH) and FeNi-py-LDH(NaBH<sub>4</sub>). (b) The Tafel slopes of FeNi-py-LDH(NaOH) and FeNi-py-LDH(NaBH<sub>4</sub>).



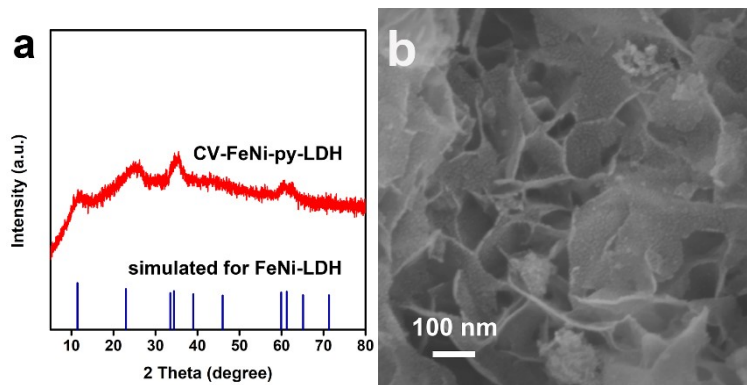
**Figure S18.** The CV curves of FeNi-py-LDH at different scan rate of 20, 40, 60, 80, 100 and 120 mV s<sup>-1</sup>.



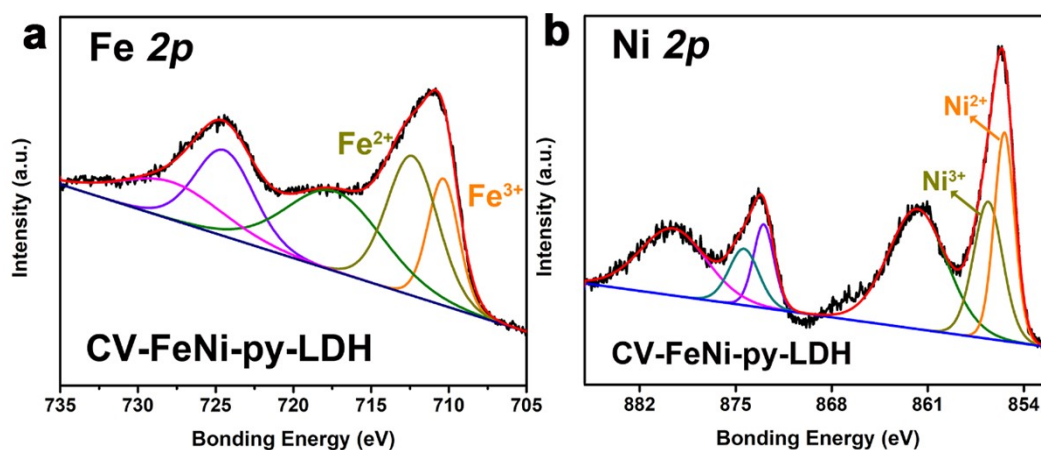
**Figure S19.** The CV curves of FeNi-ISOQ-LDH at different scan rate of 20, 40, 60, 80, 100 and 120 mV s<sup>-1</sup>.



**Figure S20.** The CV curves of RuO<sub>2</sub> at different scan rate of 20, 40, 60, 80, 100 and 120 mV s<sup>-1</sup>.



**Figure S21.** (a) XRD pattern and (b) SEM image of FeNi-py-LDH after 1000 cycles.



**Figure S22.** (a) Fe 2p (b) Ni 2p XPS spectra of FeNi-py-LDH after 1000 cycles.

**Table S1.** Comparison of electrocatalytic performance of OER catalysts in 1 M KOH.

Catalysts	MOFs	Overpotential at 10 mA·cm <sup>-2</sup> (mV)	Tafel slope (mV·dec <sup>-1</sup> )	C <sub>dl</sub> (mF cm <sup>-2</sup> )	Ref.
FeNi-py-LDH	FeNi-py-MOF	238	22	1.9	This work
FeNi-ISOQ-LDH	FeNi-ISOQ-MOF	278	42	1.1	This work
RuO <sub>2</sub> nanoparticles	-	389	88	0.2	This work
Co <sub>9</sub> S <sub>8</sub> @NiFe LDH	-	220	52	31.8	1
Ni-Fe LDH-V <sub>O</sub>	-	230	39.6	-	2
NiFeV-LDH	-	241	53.7	-	3
Ce-Ni-Fe LDH	MIL-88A	242	34	1.46	4
Ni-Fe DSNCs	MIL-88B	246(20 mA·cm <sup>-2</sup> )	71	1.46	5
NiFeCo-LDH/CF	ZIF-67	249	42	0.71	6
Fe-Ni@NC	-	257	54.6	5.58	7
Co <sub>8</sub> Fe <sub>1</sub> -LDH	-	262	42	0.59	8
Fe <sub>2</sub> O <sub>3</sub> @NiMOF-74	Ni-MOF-74	264	48	3.33	9
Co(OH) <sub>2</sub>	-	267	62	-	10
Ni-Fe-OH/Ni <sub>3</sub> S <sub>2</sub> /NF	-	268	54	30.6	11
Fe-Ni LDH	-	280	49.4	-	12
γ-FeOOH/NF-6M	-	286	51	-	13
NiFe-LDH/NEGF	-	290	68	7.9	14
NaBH <sub>4</sub> -FeNi-LDH	-	307	48	-	15
FeNiPc-CP	-	317	-	95	16
NCO-HNSs	-	340	51	-	17
FeOOH HNTAs	-	350	79	-	18

<b>RuO<sub>2</sub></b>	-	<b>370</b>	-	-	19
<b>nanoparticles</b>					
<b>RuO<sub>2</sub></b>		<b>389</b>	-	-	20
<b>nanoparticles</b>					
<b>RuO<sub>2</sub></b>		<b>390</b>	-	-	21
<b>nanoparticles</b>					
<b>RuO<sub>2</sub></b>		<b>397</b>	-	-	22
<b>nanoparticles</b>					
<b>Beta-FeOOH</b>	-	<b>400</b>	<b>186</b>	-	23
<b>RuO<sub>2</sub></b>	-	<b>400</b>	-	-	24
<b>nanoparticles</b>					
<b>NiOOH</b>	-	<b>453</b>	<b>189</b>	<b>3.4</b>	25

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