

## **Ni-Co hydroxide nanosheets on plasma-reduced Co-based metal-organic nanocages for electrocatalytic water oxidation**

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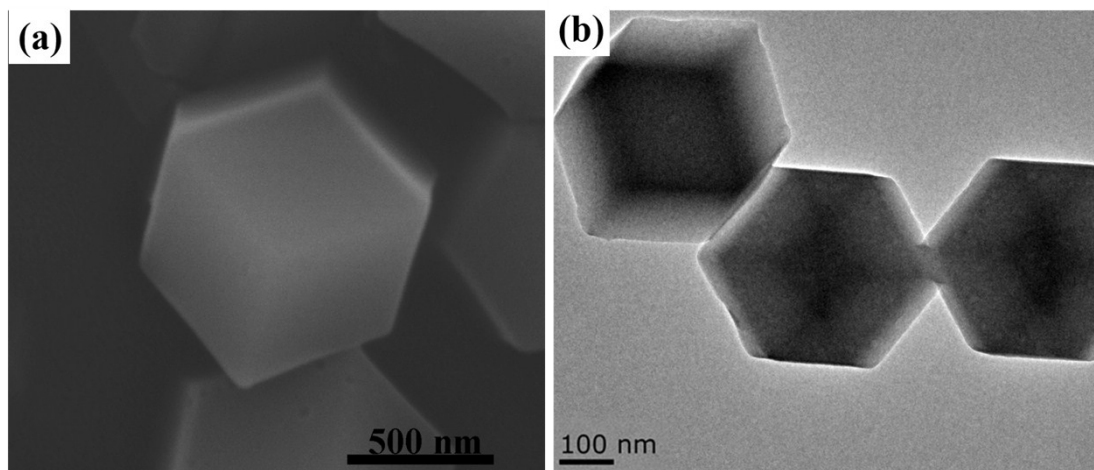
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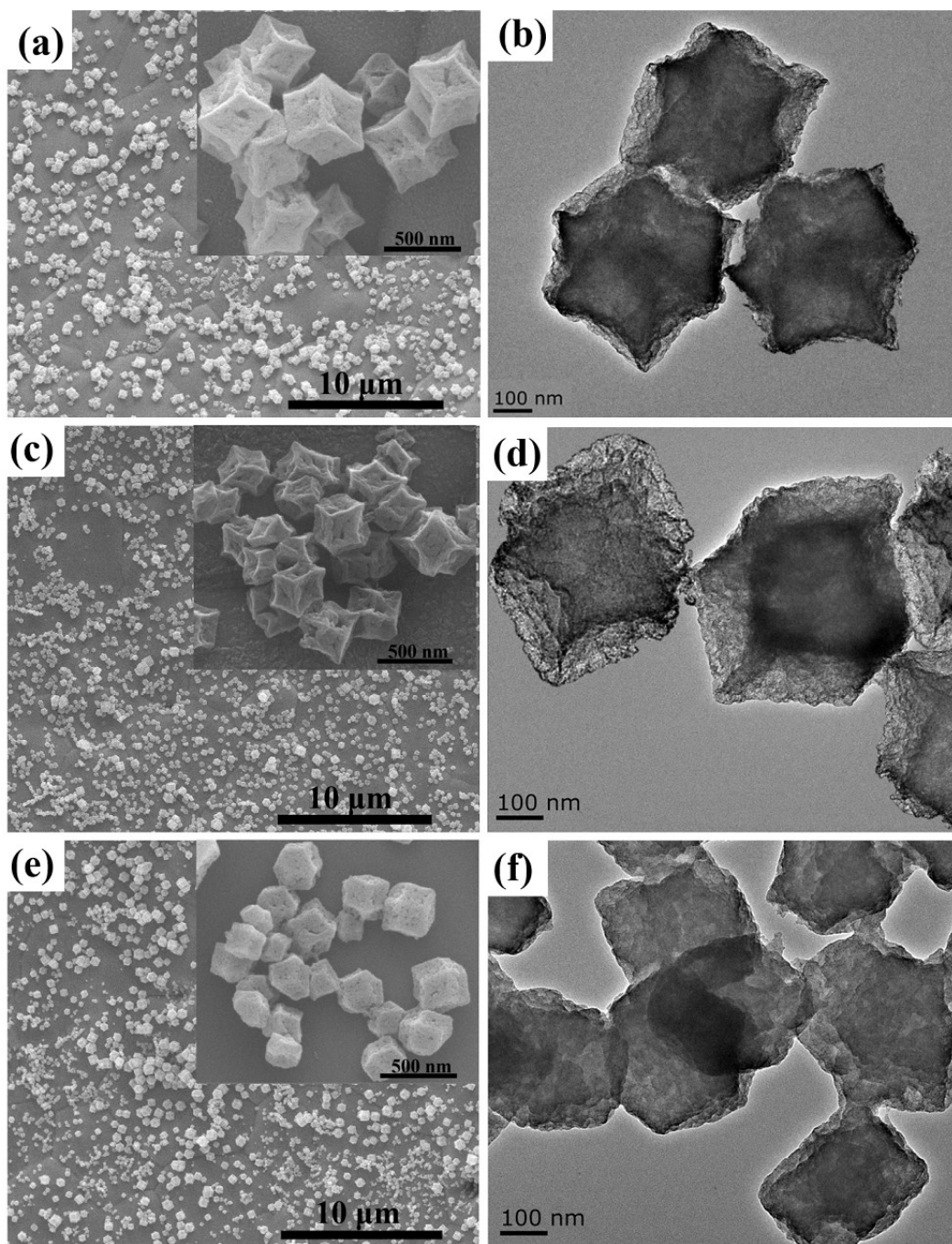
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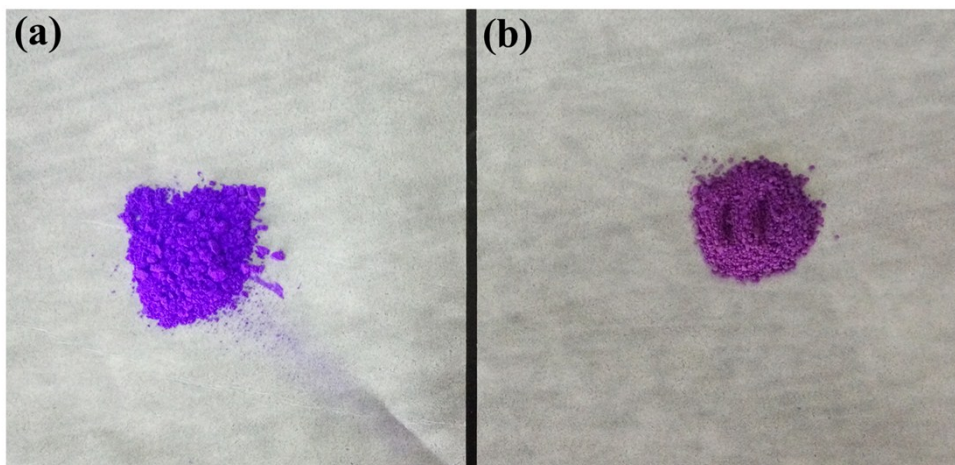
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**Fig. S1** (a) SEM and (b) TEM images of pure ZIF-67.



**Fig. S2** Preparation of ZIF-67-V<sub>0</sub>/NF: (a), (c) and (e) SEM (the inset shows the enlarged images and the scale bar is 500 nm) and (b), (d) and (f) TEM images of ZIF-67-V<sub>0</sub> treated by O<sub>2</sub>-Ar RF plasma in 120, 150 and 180W, respectively.



**Fig. S3** The optical images of (a) pure ZIF-67 and (b) ZIF-67-V<sub>o</sub> treated by O<sub>2</sub>-Ar RF plasma in 150W.

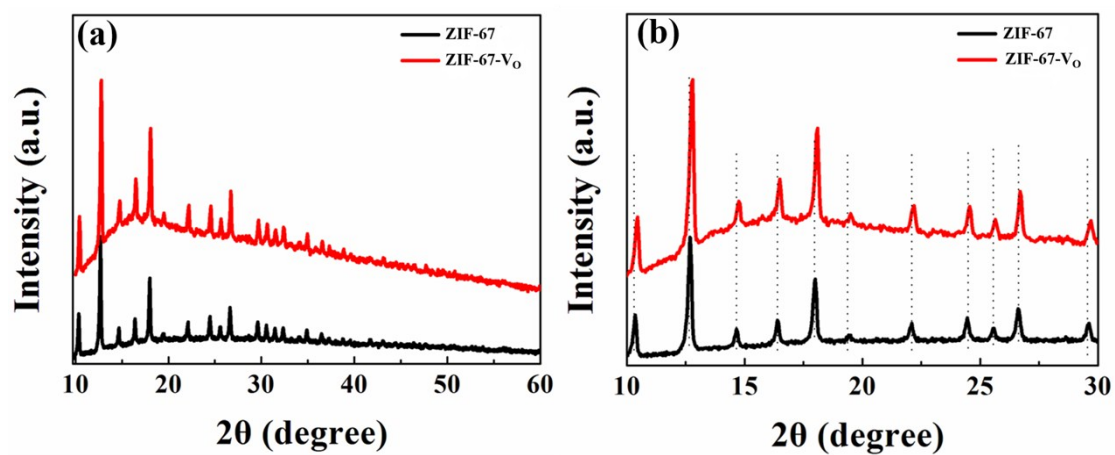
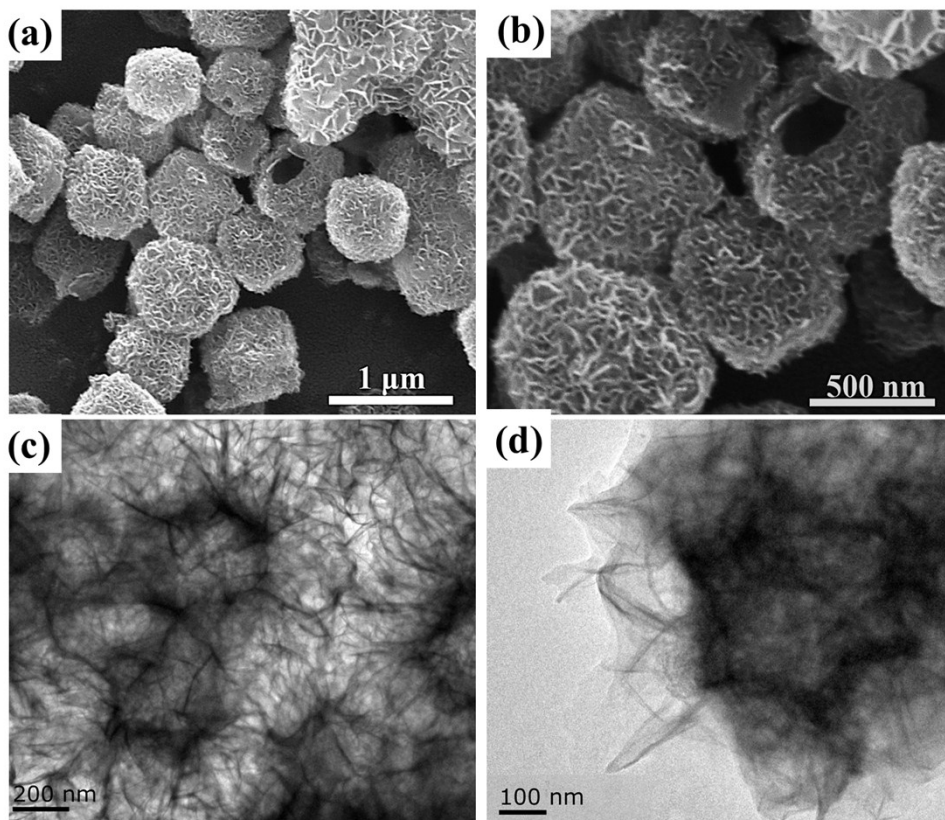
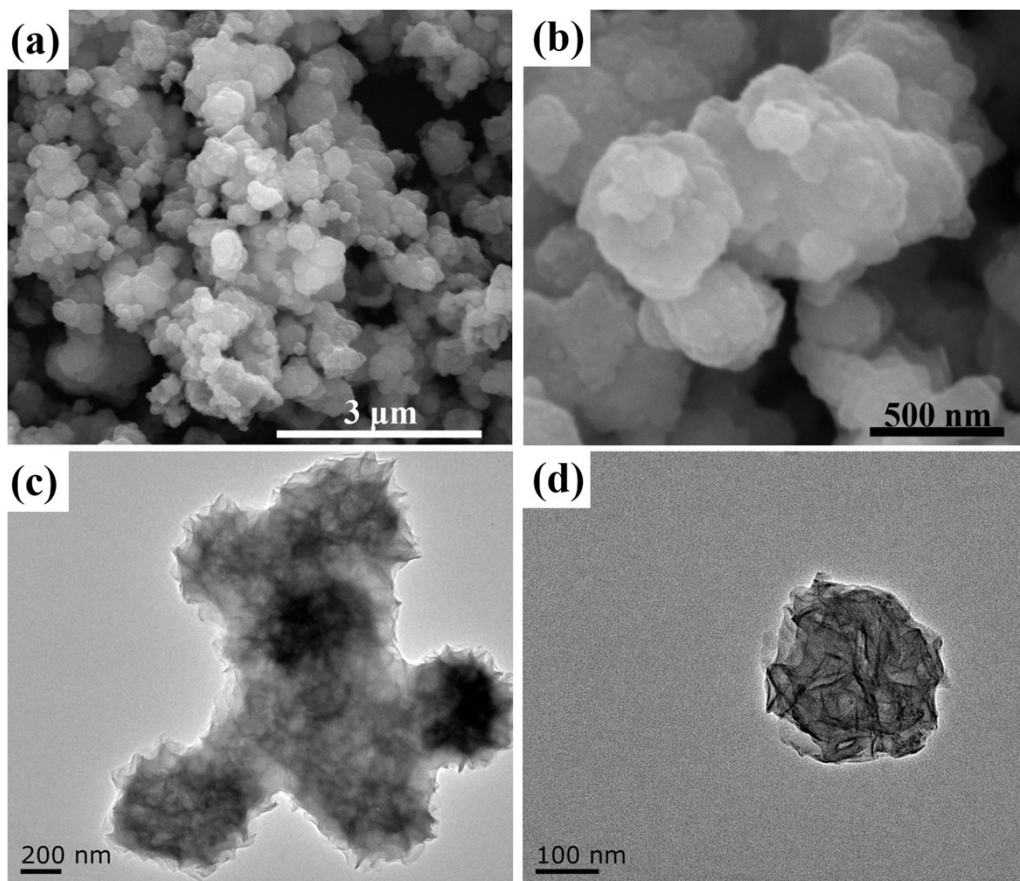


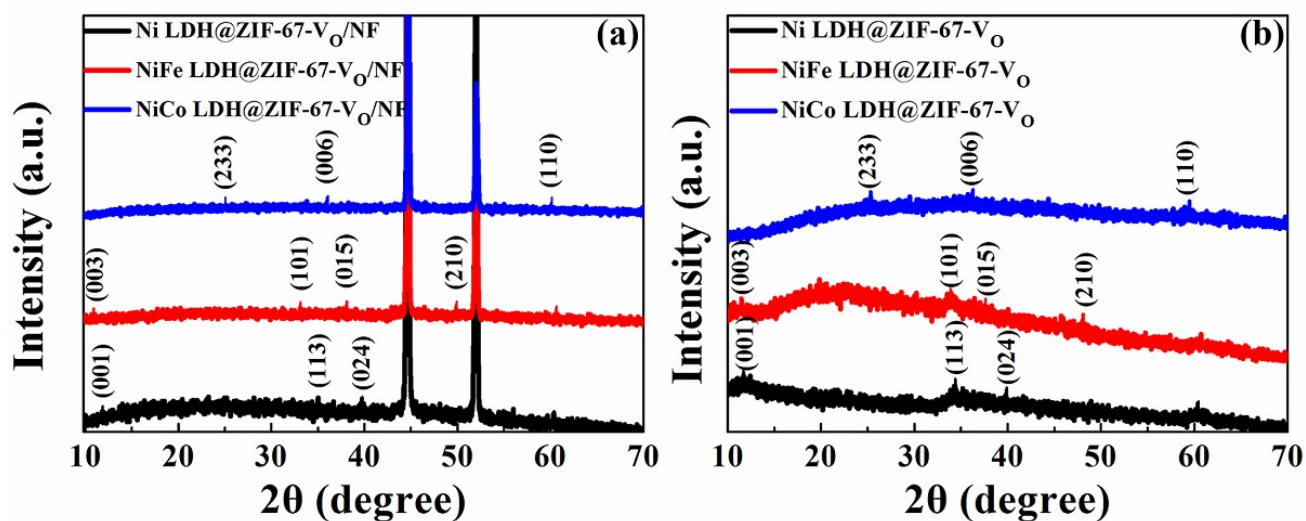
Fig. S4 (a) XRD patterns and (b) the enlarged XRD patterns images of the pure ZIF-67 and ZIF-67-V<sub>O</sub> powder.



**Fig. S5** (a) and (b) SEM, (c) and (d) TEM images of the Ni LDH@ZIF-67-V<sub>O</sub>/NF.

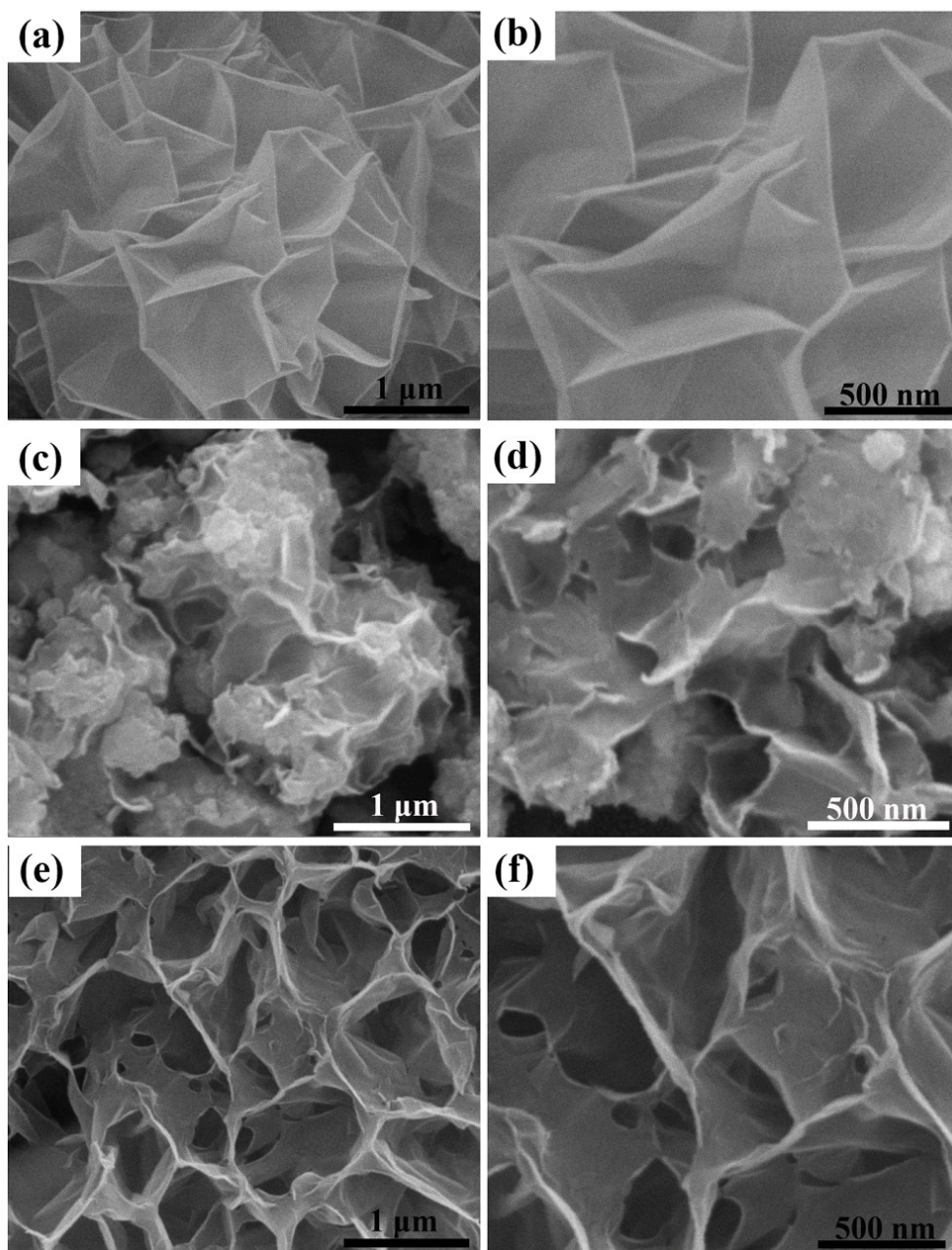


**Fig. S6** (a) and (b) SEM, (c) and (d) TEM images of the NiFe LDH@ZIF-67-V<sub>O</sub>/NF.

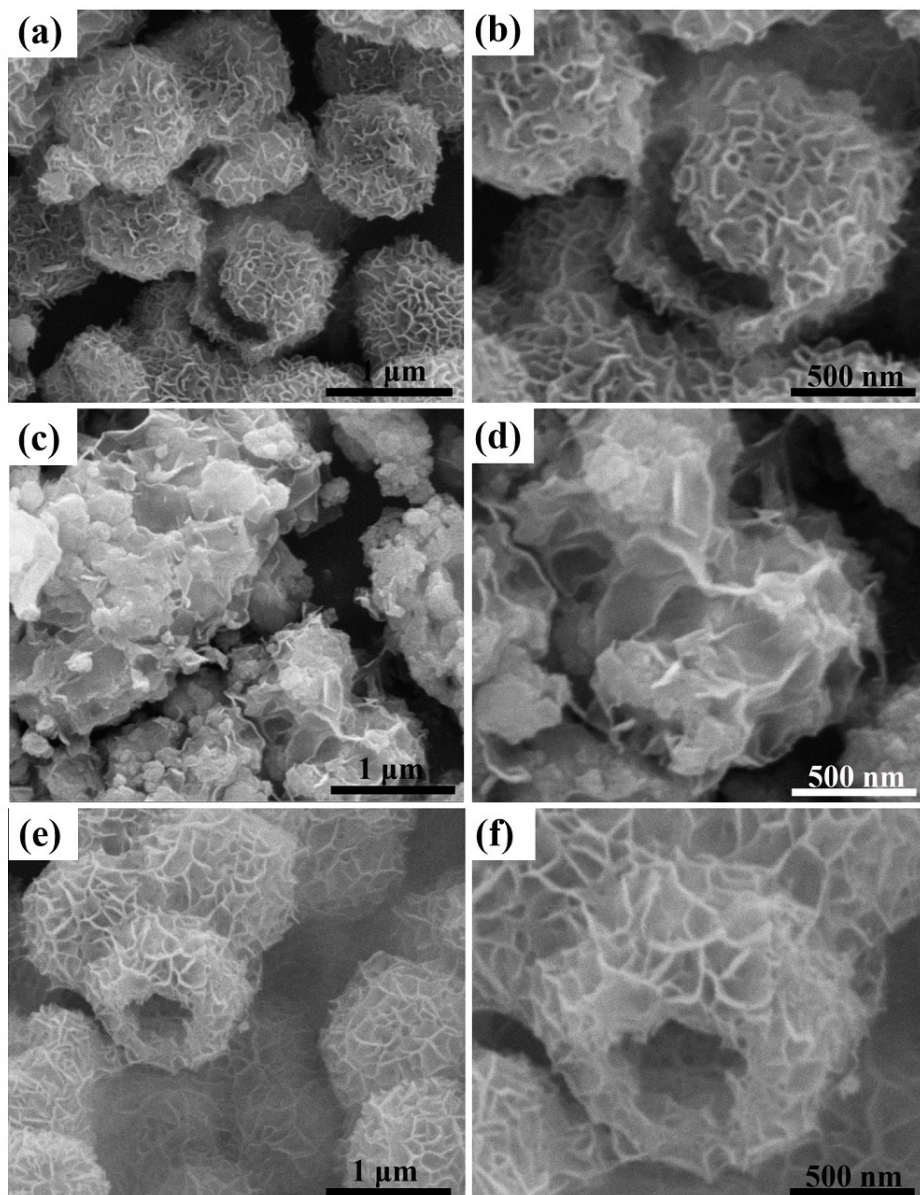


**Fig. S7** (a) The XRD patterns images of the Ni LDH@ZIF-67-V<sub>O</sub>/NF, NiFe LDH@ZIF-67-V<sub>O</sub>/NF, NiCo LDH@ZIF-67-V<sub>O</sub>/NF, and (b) the corresponding Ni LDH@ZIF-67-V<sub>O</sub>, NiFe LDH@ZIF-67-V<sub>O</sub>, NiCo LDH@ZIF-67-V<sub>O</sub> powder sample scraped from the substrate.

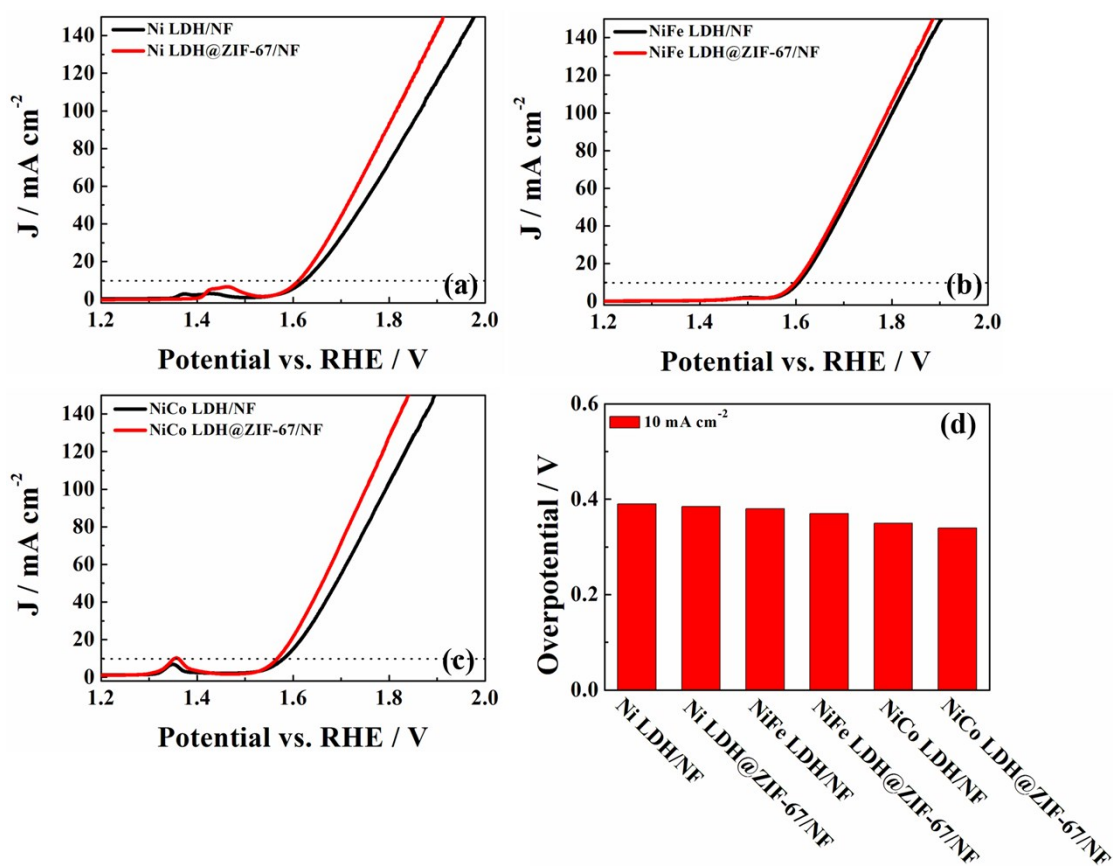




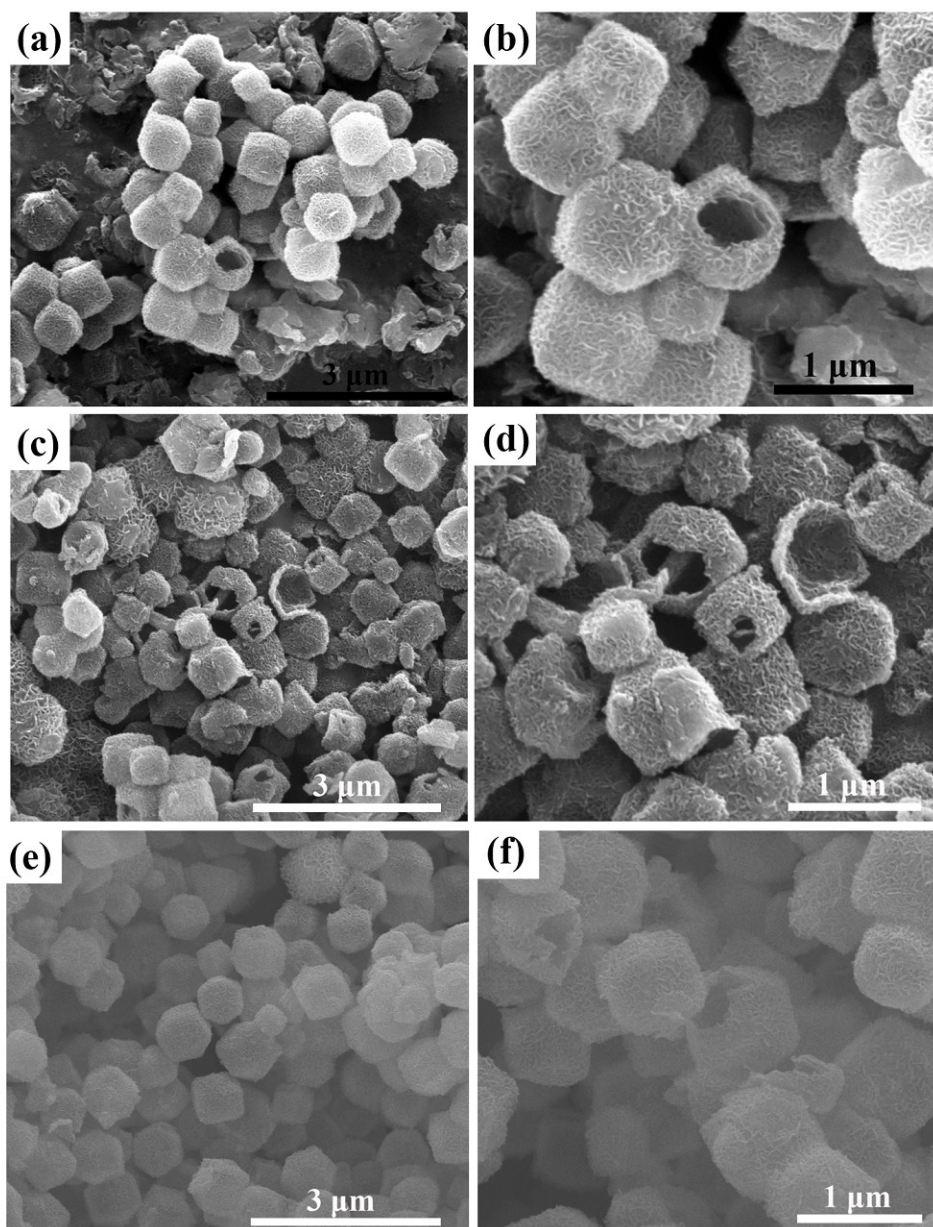
**Fig. S8** SEM images of (a) and (b) Ni LDH/NF, (c) and (d) NiFe LDH/NF, (e) and (f) NiCo LDH/NF.



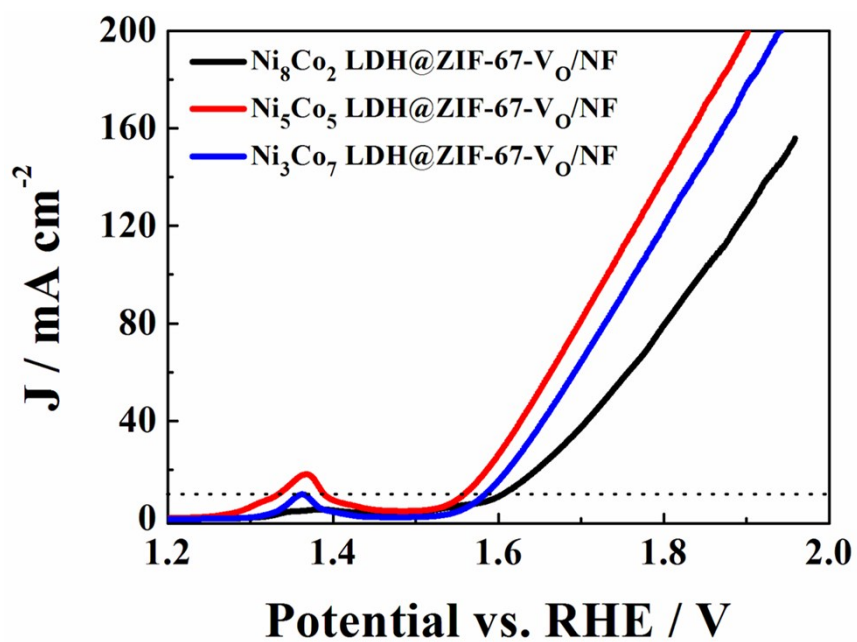
**Fig. S9** SEM images of (a) and (b) Ni LDH@ZIF-67/NF, (c) and (d) NiFe LDH@ZIF-67/NF, (e) and (f) NiCo LDH@ZIF-67/NF.



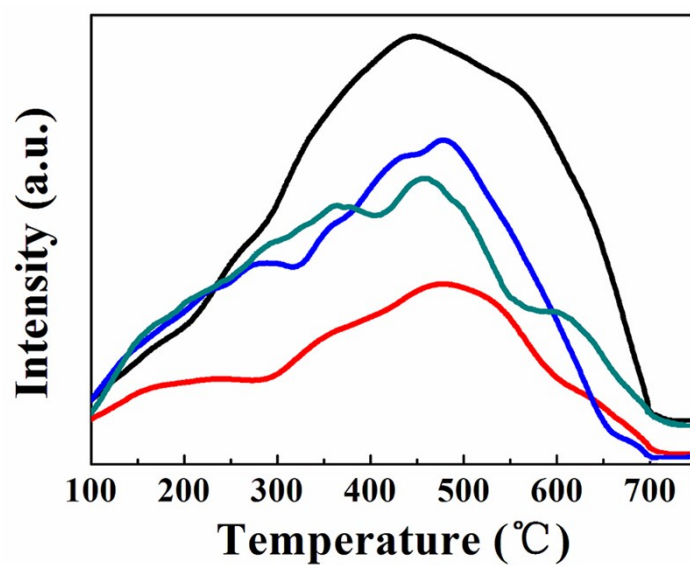
**Fig. S10** LSV curves of (a) Ni LDH/NF, Ni LDH@ZIF-67/NF, (b) NiFe LDH/NF, NiFe LDH@ZIF-67/NF, (c) NiCo LDH /NF and NiCo LDH@ZIF-67/NF for the OER, and (d) the corresponding overpotential at 10 mA cm<sup>-2</sup>.



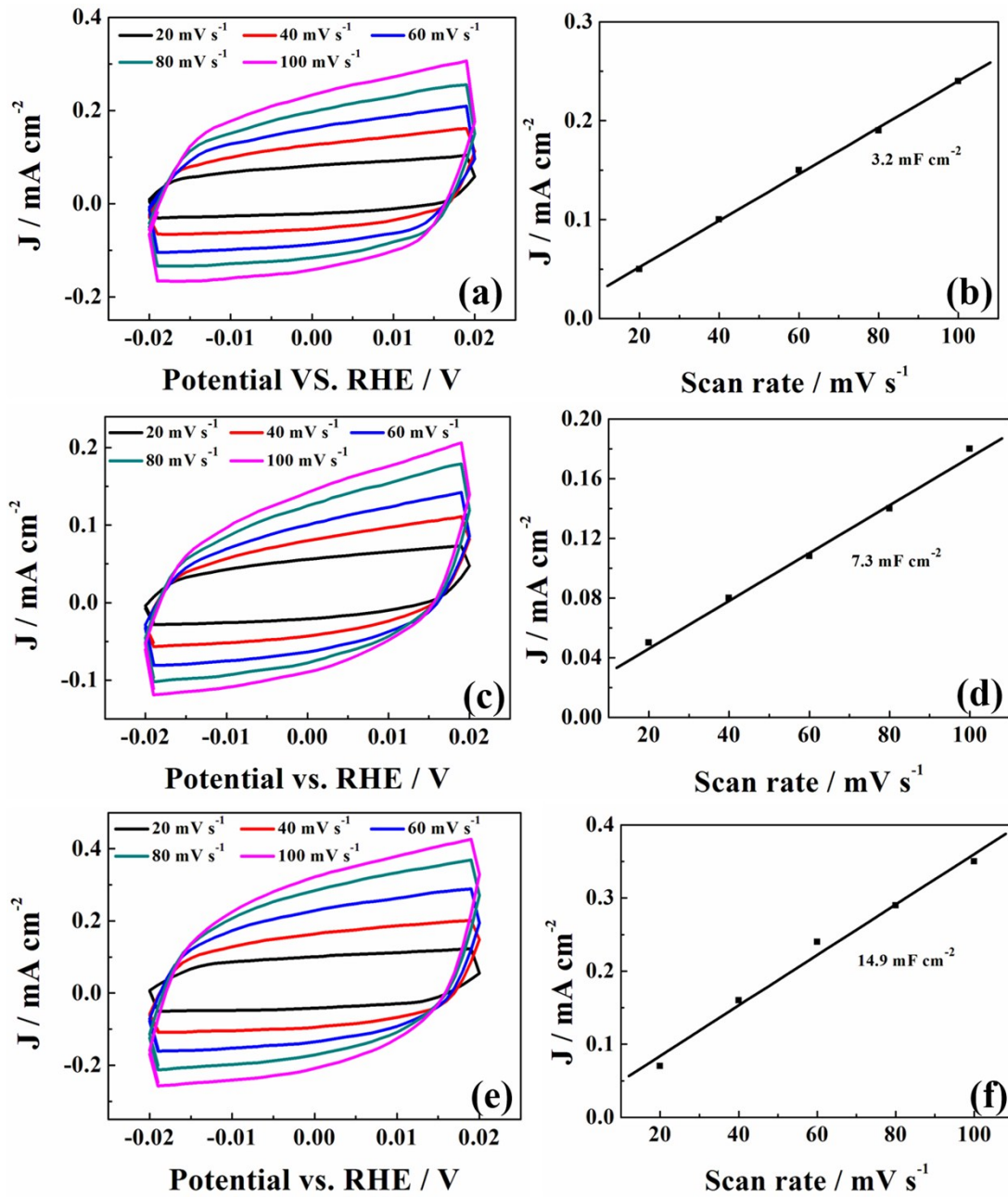
**Fig. S11** SEM images of the NiCo LDH@ZIF-67-V<sub>O</sub>/NF (Ni:Co=8:2), NiCo LDH@ZIF-67-V<sub>O</sub>/NF (Ni:Co=5:5) and Ni<sub>2</sub>Co LDH@ZIF-67-V<sub>O</sub>/NF (Ni:Co=3:7).



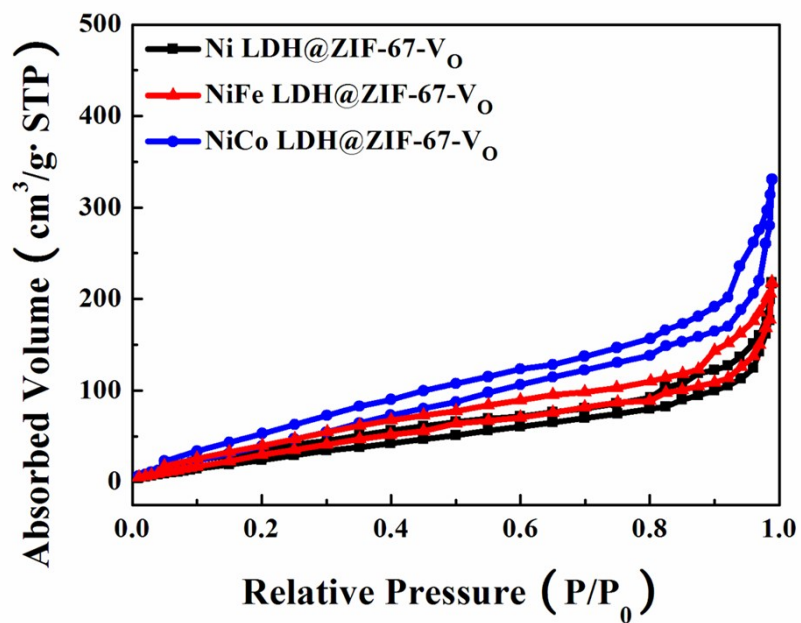
**Fig. S12** LSV curves of NiCo LDH@ZIF-67-V<sub>0</sub>/NF (Ni:Co=8:2), NiCo LDH@ZIF-67-V<sub>0</sub>/NF (Ni:Co=5:5) and NiCo LDH@ZIF-67-V<sub>0</sub>/NF (Ni:Co=3:7).



**Fig. S13** O<sub>2</sub>-TPD analysis of NiCo LDH@ZIF-67-V<sub>0</sub>/NF (black), NiFe LDH@ZIF-67-V<sub>0</sub>/NF (blue), NiCo LDH@ZIF-67/NF (green) and Ni LDH@ZIF-67-V<sub>0</sub>/NF (red), showing the O<sub>2</sub> desorption capabilities.

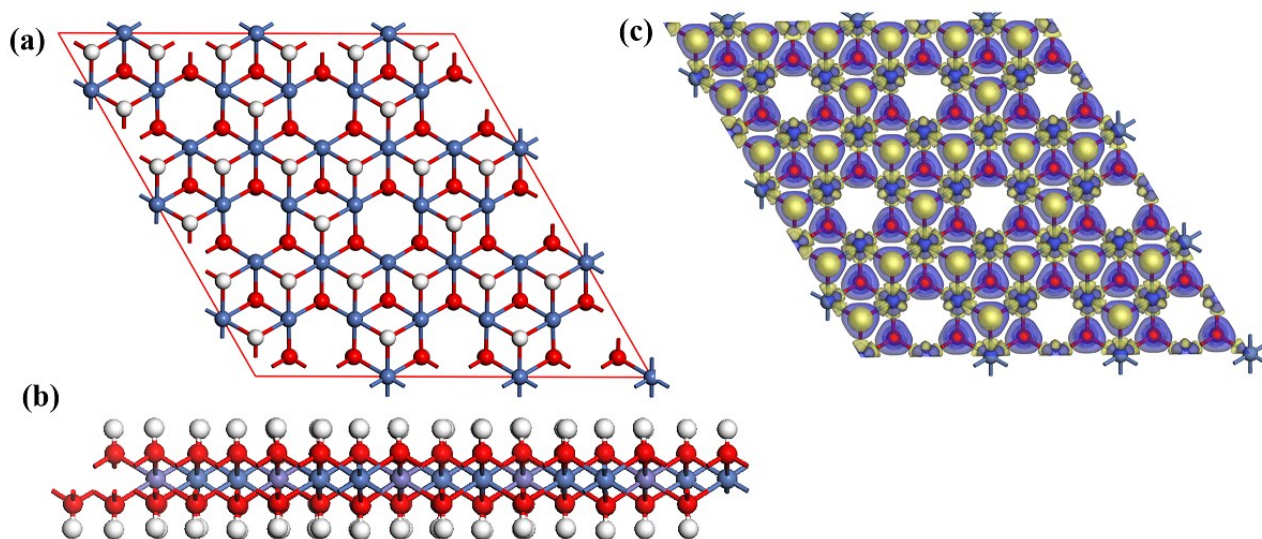


**Fig. S14** CV curves at various scan rates in the potential range -0.02~0.02 V vs. RHE for (a) Ni LDH@ZIF-67-V<sub>o</sub>/NF, (c) NiFe LDH@ZIF-67-V<sub>o</sub>/NF and (e) NiCo LDH@ZIF-67-V<sub>o</sub>/NF, and the double layer capacitances ( $C_{dl}$ ) of (b) Ni LDH@ZIF-67-V<sub>o</sub>/NF, (d) NiFe LDH@ZIF-67-V<sub>o</sub>/NF and (f) NiCo LDH@ZIF-67-V<sub>o</sub>/NF at the potential of 0 V vs. RHE, respectively.

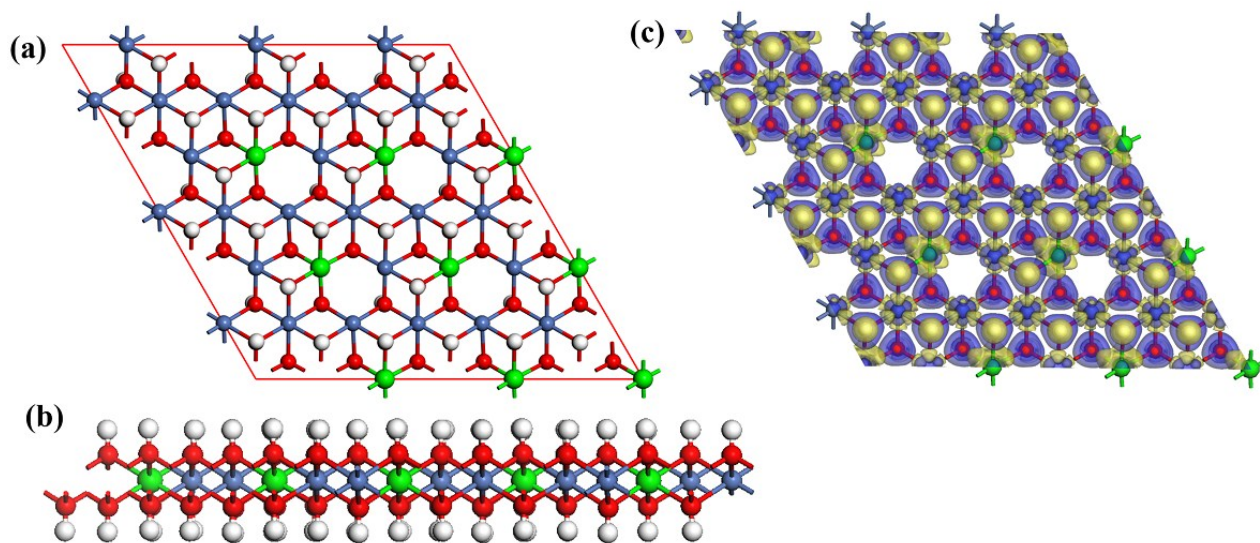


**Fig. S15** N<sub>2</sub> adsorption/desorption curve of Ni LDH@ZIF-67-V<sub>o</sub>, NiFe LDH@ZIF-67-V<sub>o</sub> and NiCo LDH@ZIF-67-V<sub>o</sub> powder sample scraped from the substrate at 77 K.

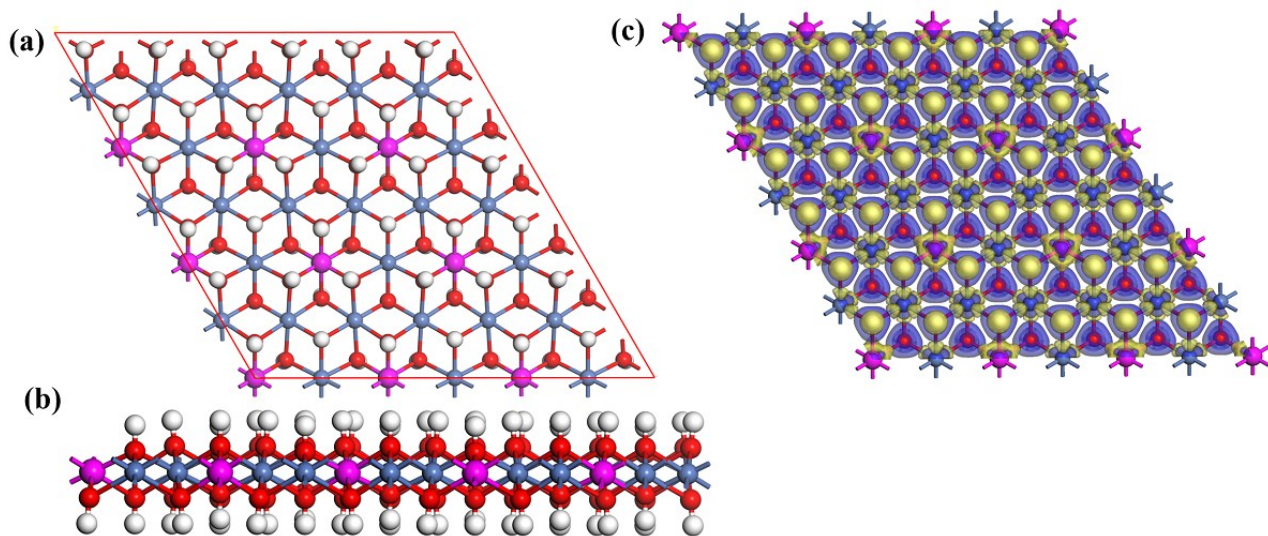




**Fig. S16** Atomic structure model and theoretical calculation analysis of the catalysts: (a) Top view and (b) side view of the model structure of Ni LDH-V<sub>O</sub>, (c) corresponding 3D charge distributions.



**Fig. S17** Atomic structure model and theoretical calculation analysis of the catalysts: (a) Top view and (b) side view of the model structure of NiFe LDH-V<sub>O</sub>, (c) corresponding 3D charge distributions.



**Fig. S18** Atomic structure model and theoretical calculation analysis of the catalysts: (a) Top view and (b) side view of the model structure of NiCo LDH, (c) corresponding 3D charge distributions.

**Table S1.** The comparison of OER performance with state-of-the-art electrocatalysts.

Materials	Supports	Electrolytes	$E_{J=10 \text{ mA cm}^{-2}}$ (V)	References
NiCo LDH@ZIF-67-V <sub>O</sub> /NF	Ni foam	1 M KOH	1.52	This work
NiCo LDH@ZIF-67/NF	Ni foam	1 M KOH	1.56	This work
Ni LDH@ZIF-67-V <sub>O</sub> /NF	Ni foam	1 M KOH	1.59	This work
Ni LDH@ZIF-67/NF	Ni foam	1 M KOH	1.615	This work
NiFe LDH@ZIF-67-V <sub>O</sub> /NF	Ni foam	1 M KOH	1.56	This work
NiFe LDH@ZIF-67/NF	Ni foam	1 M KOH	1.595	This work
CoO <sub>x</sub> -ZIF	-	1 M KOH	1.548	1
NiFe LDH/NiCo <sub>2</sub> O <sub>4</sub> /NF	Ni foam	1 M KOH	1.57	2
Ni(OH) <sub>2</sub> /NF	Ni Foam	1 M KOH	1.58	3
NiFe LDH	Ni Foam	1 M KOH	1.545	4
NiCo LDH	CFP	1 M KOH	1.597	5
NiFe/NiCo <sub>2</sub> O <sub>4</sub> /NF	Ni Foam	1M KOH	1.57	6

**Table S2.** BET surface areas, and BJH pore volume of the samples.

Sample	S <sub>BET</sub> (m <sup>2</sup> g <sup>-1</sup> )	Pore Volume (cm <sup>3</sup> g <sup>-1</sup> )
Ni LDH@ZIF-67-V <sub>O</sub>	127	0.390
NiFe LDH@ZIF-67-V <sub>O</sub>	131	0.413
NiCo LDH@ZIF-67-V <sub>O</sub>	138	0.427

**Table S3.** The atomic ratio of Ni<sub>8</sub>Co<sub>2</sub> LDH@ZIF-67-V<sub>O</sub>/NF, Ni<sub>5</sub>Co<sub>5</sub> LDH@ZIF-67-V<sub>O</sub>/NF and Ni<sub>3</sub>Co<sub>7</sub> LDH@ZIF-67-V<sub>O</sub>/NF samples.

Sample	Atomic concentration (%)				
	C	N	O	Ni	Co
Ni <sub>8</sub> Co <sub>2</sub> LDH@ZIF-67-V <sub>O</sub> /NF	38.71	3.92	47.59	7.86	1.92
Ni <sub>5</sub> Co <sub>5</sub> LDH@ZIF-67-V <sub>O</sub> /NF	37.98	4.01	48.20	4.89	4.92
Ni <sub>3</sub> Co <sub>7</sub> LDH@ZIF-67-V <sub>O</sub> /NF	39.52	3.95	46.53	2.98	7.02

## References

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