

## **Supporting Information**

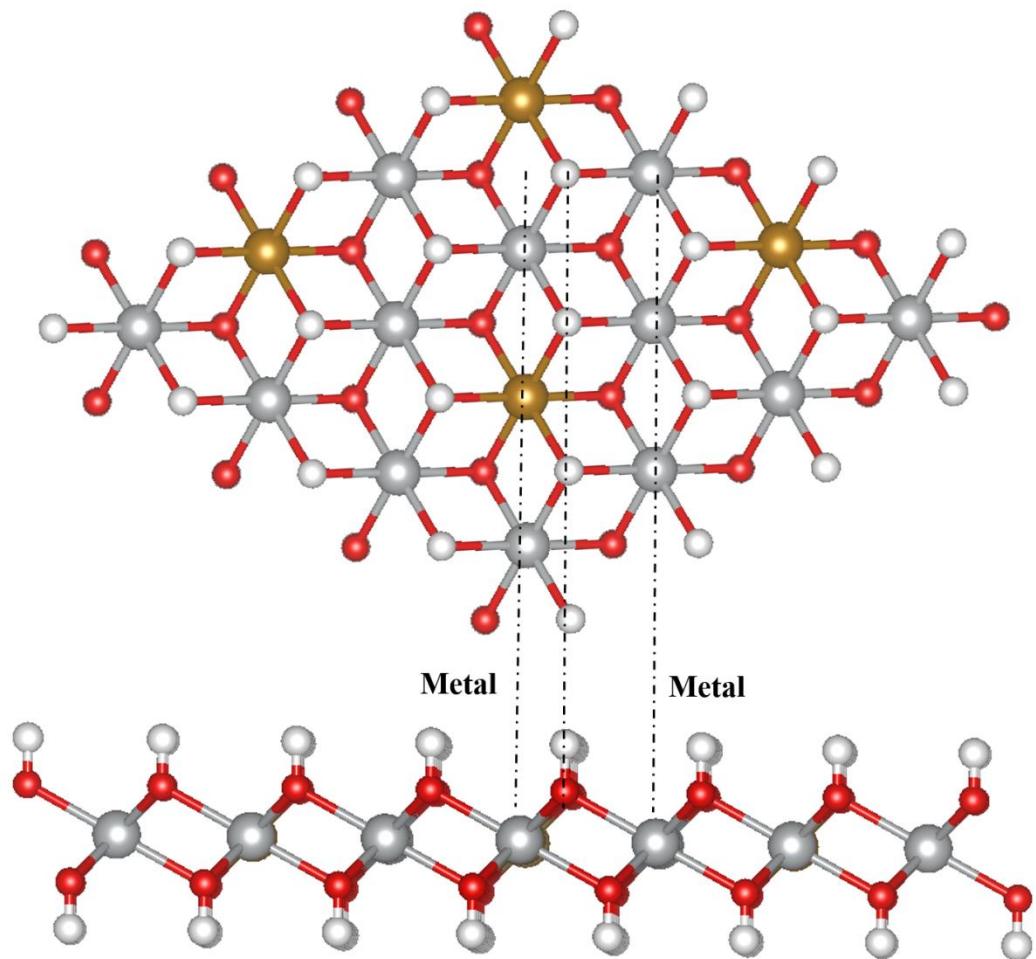
### **Hierarchical NiCo<sub>2</sub>S<sub>4</sub>@NiFe LDH Heterostructures Supported on Nickel Foam for Enhanced Overall-Water-Splitting Activity**

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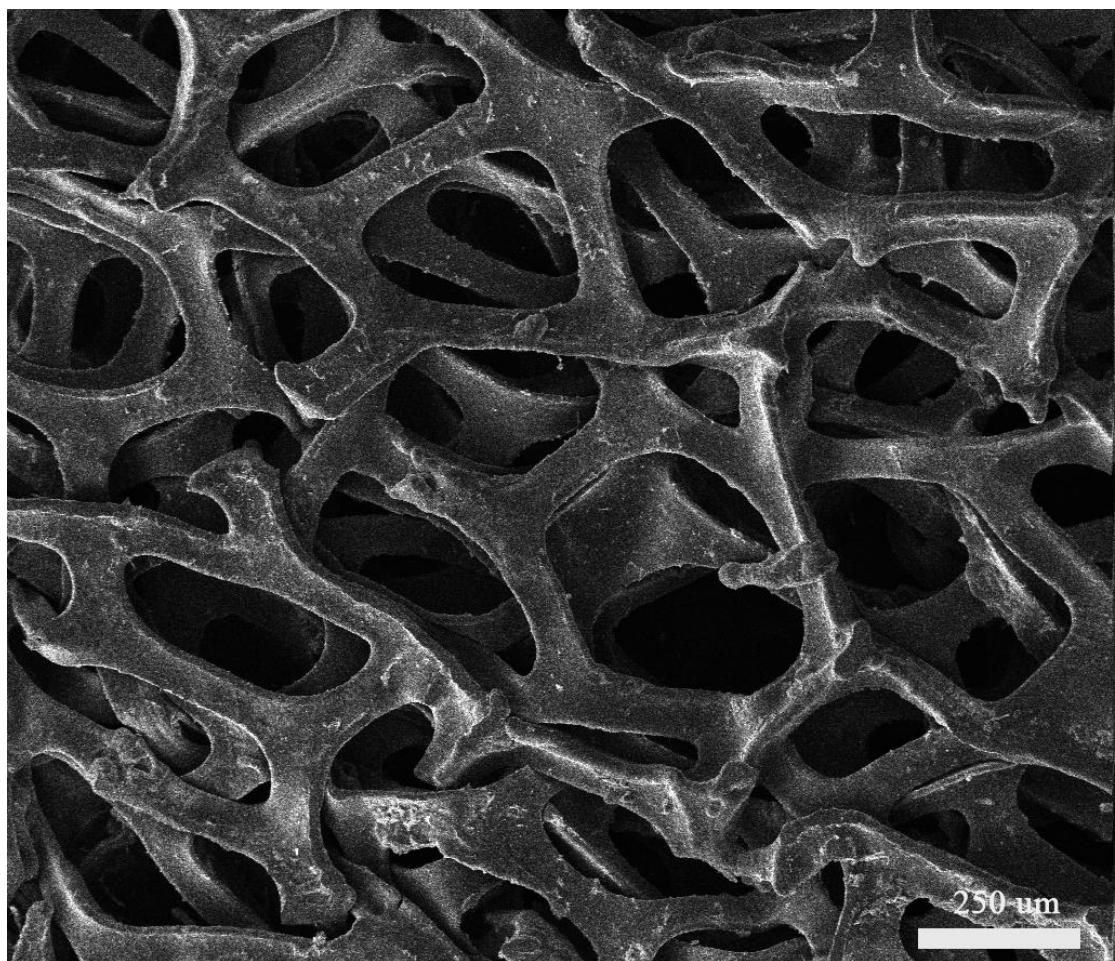
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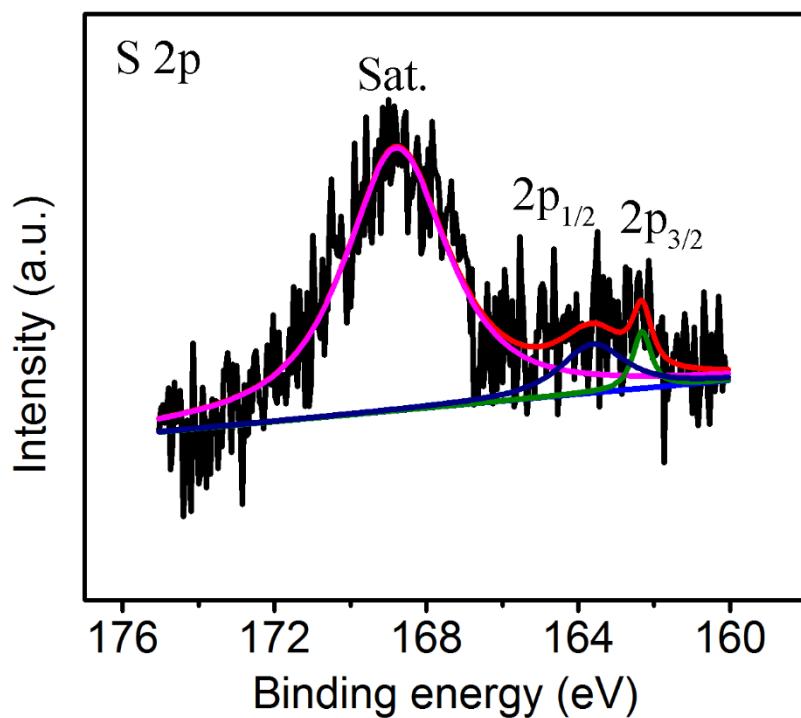
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**Figure S1** Schematic showing the structure of NiFe-LDH with the top view and side view. The red, white, grey and golden atoms represent the O, H, Ni and Fe atoms, respectively.



**Figure S2** SEM images of bare Ni Foam.



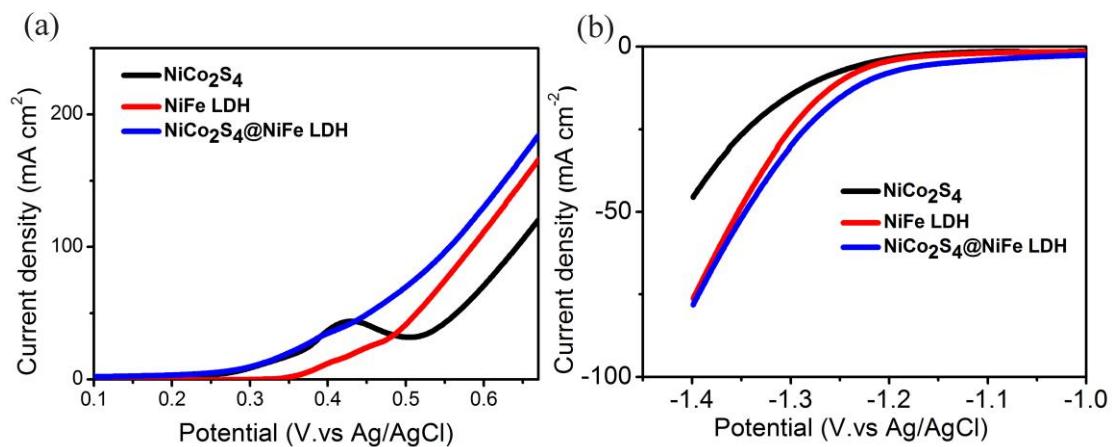
**Figure S3** XPS spectra of Co 2p in NiCo<sub>2</sub>S<sub>4</sub>@NiFe LDH/NF heterostructures.

**Table S1. Comparison of OER activity data with some representative non-noble OER catalysts.**

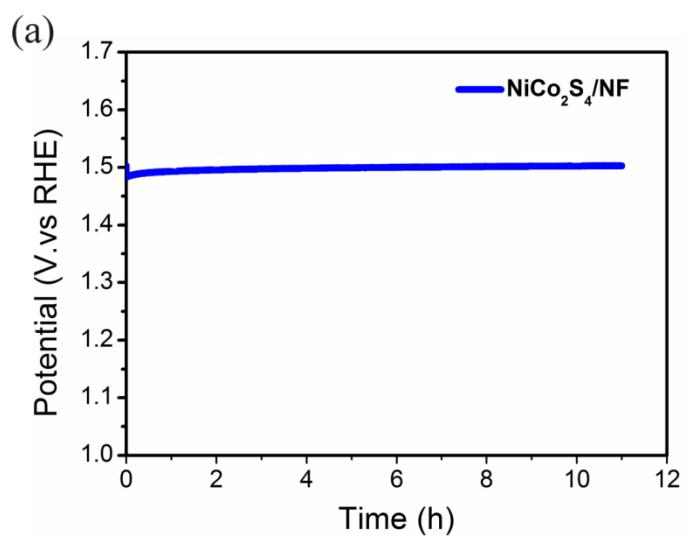
Catalyst	Electrolyte	Overpotential (mV)@j (mA cm <sup>-2</sup> )	Ref.
NiCo <sub>2</sub> S <sub>4</sub> @NiFe LDH/NF	1 M KOH	201 @60	This work
NiFe LDH/NF	1 M KOH	260 @60	This work
NiCo <sub>2</sub> S <sub>4</sub> /NF	1 M KOH	306 @60	This work
Co(OH) <sub>2</sub>	1 M KOH	360 @10	<sup>1</sup>
Co <sub>4</sub> O <sub>4</sub>	1 M KOH	408 @10	<sup>2</sup>
NiCoFe LTH/CC	1 M KOH	239 @10	<sup>3</sup>
NiCo <sub>2</sub> O <sub>4</sub> @NiFe LDH/NF	1 M KOH	290 @50	<sup>4</sup>
NiCo <sub>2</sub> S <sub>4</sub> /NF	1 M KOH	260 @10	<sup>5</sup>
NiSe <sub>2</sub> NCs/GC	1 M KOH	250 @10	<sup>6</sup>

**Table S2. Comparison of HER activity data with some representative non-noble HER catalysts.**

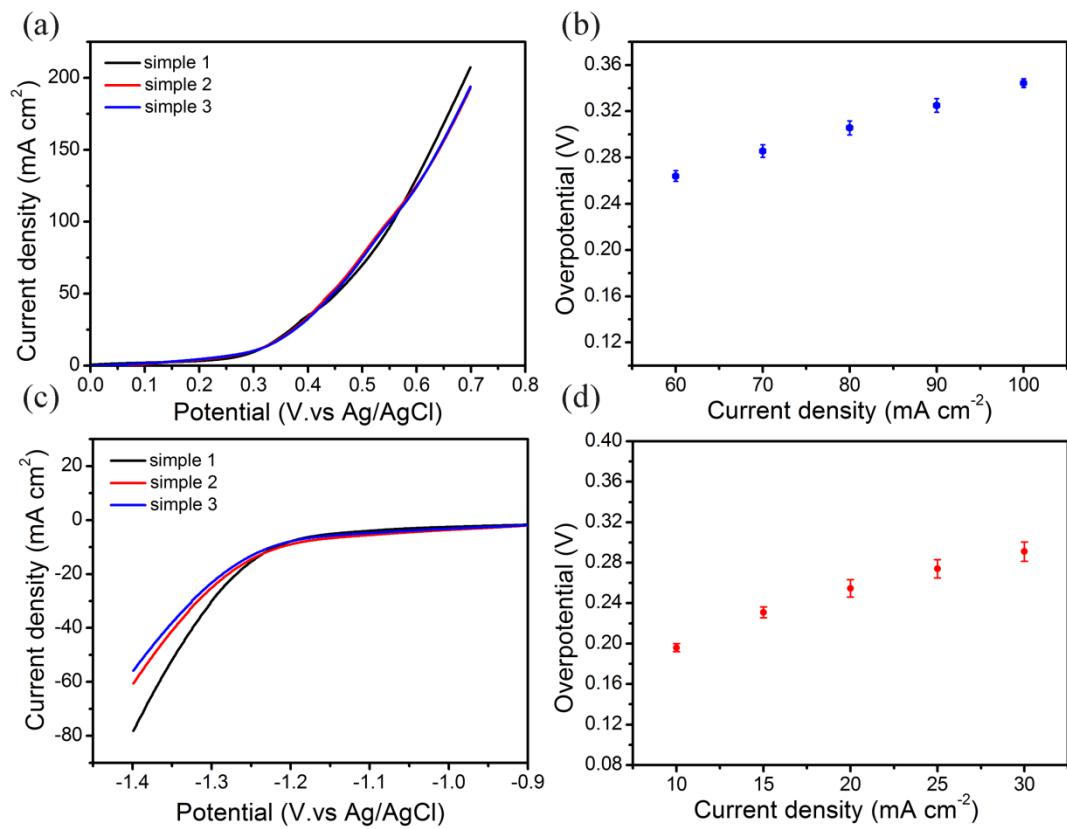
Catalyst	Electrolyte	Overpotential (mV)@j (mA cm <sup>-2</sup> )	Ref.
NiCo <sub>2</sub> S <sub>4</sub> @NiFe LDH/NF	1 M KOH	200 @10	This work
NiFe LDH/NF	1 M KOH	225 @10	This work
NiCo <sub>2</sub> S <sub>4</sub> /NF	1 M KOH	248 @10	This work
Ni nanowires	1 M KOH	350 @10	<sup>7</sup>
CoP/CC	1 M KOH	209 @10	<sup>8</sup>
NiCoFe LTH/CC	1 M KOH	200 @10	<sup>3</sup>
Porous NiSe <sub>2</sub> nanosheets	1 M KOH	184 @10	<sup>9</sup>
Ni <sub>9</sub> S <sub>8</sub> /NF	1 M KOH	230 @10	<sup>10</sup>
Ni <sub>2</sub> P/GC	1 M KOH	220@10	<sup>11</sup>
Ni <sub>3</sub> S <sub>2</sub> /NF	1 M KOH	200@10	<sup>12</sup>



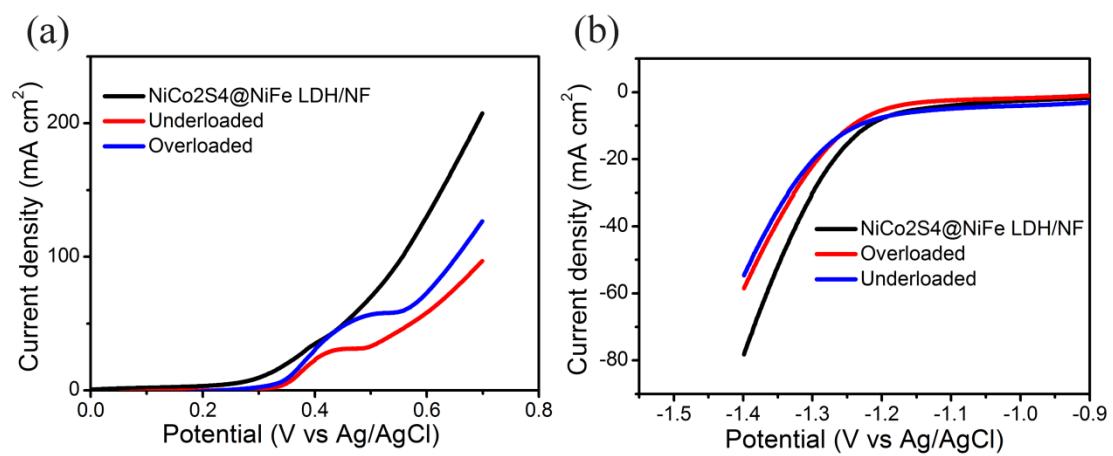
**Figure S4** (a) and (b) Polarization curves versus Ag/AgCl without iR-correction.



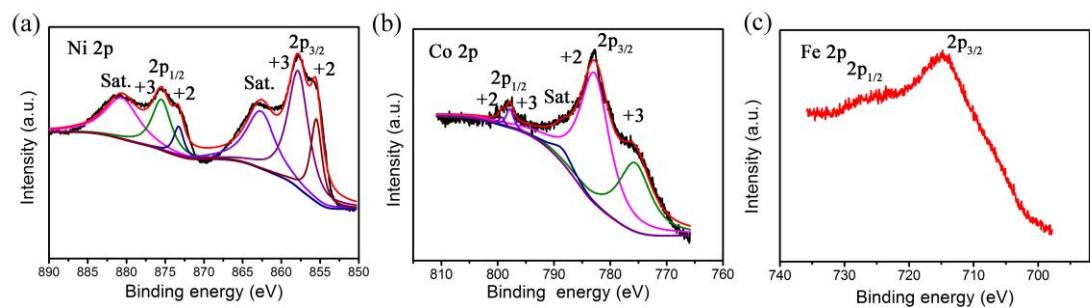
**Figure S5** (a) chronopotentiometry curves of  $\text{NiCo}_2\text{S}_4@\text{NiFe LDH/NF}$  at the current density of  $10 \text{ mA cm}^{-2}$



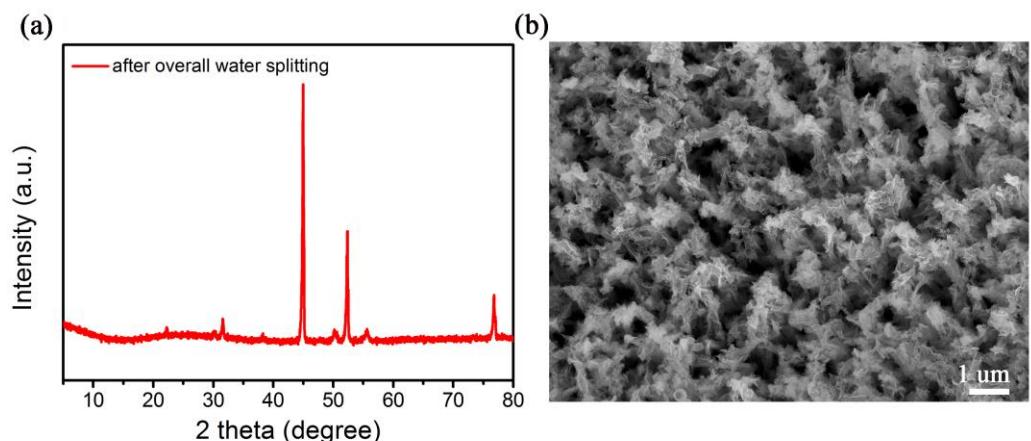
**Figure S6.** (a) and (c) polarization curves for OER and HER; (b) and (d) the corresponding error bars



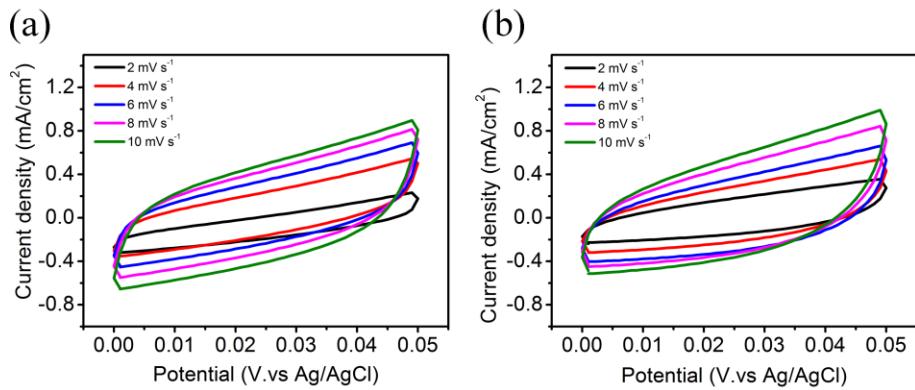
**Figure S7.** (a-b) Polarization curves for samples with underloaded, optimal and overloaded NiFe-LDH.



**Figure S8.** (a-c) The XPS spectra of Ni 2p, Co 2p, and Fe 2p in  $\text{NiCo}_2\text{S}_4@\text{NiFe}$  LDH/NF heterostructures after water splitting.



**Figure S9.** (a)The XRD, and (b) the morphology of NiCo<sub>2</sub>S<sub>4</sub>@NiFe LDH/NF heterostructures after water splitting.



**Figure S10.** Cycle voltammograms for (a) NiCo<sub>2</sub>S<sub>4</sub>/NF and (b) NiCo<sub>2</sub>S<sub>4</sub>@NiFe LDH/NF versus Ag/AgCl with various scan rates, 2, 4, 6, 8 mV s<sup>-1</sup>.

## Reference

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