

## Electronic Supplementary Information

### Ultrathin sulfate-intercalated NiFe-layered double hydroxides nanosheets for efficient electrocatalytic oxygen evolution

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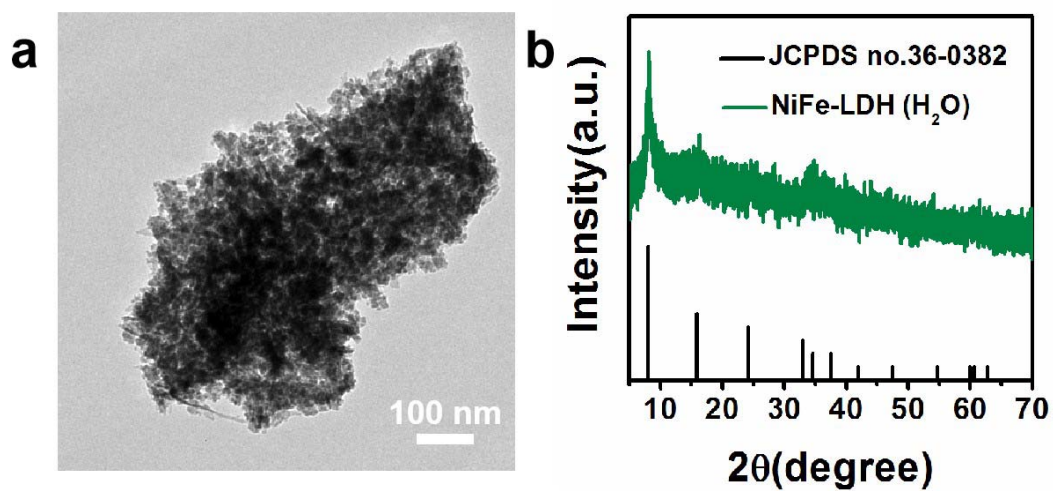
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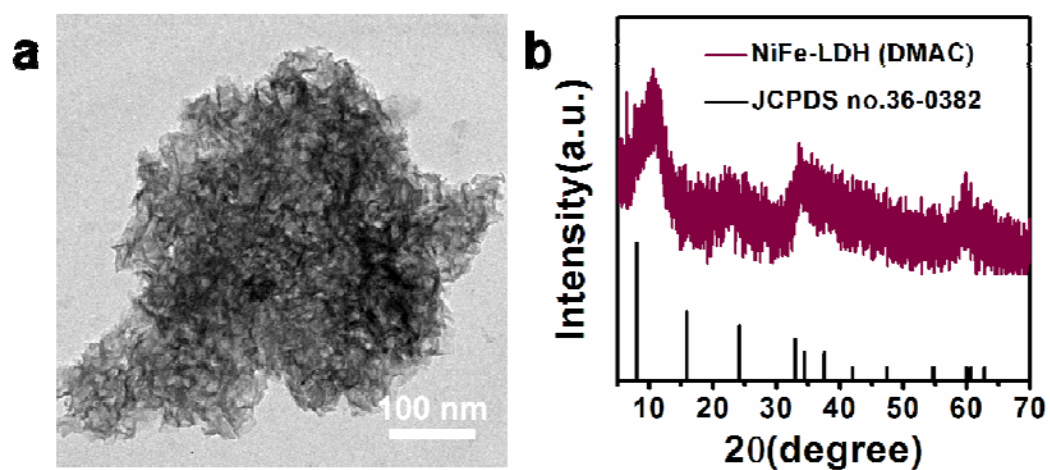
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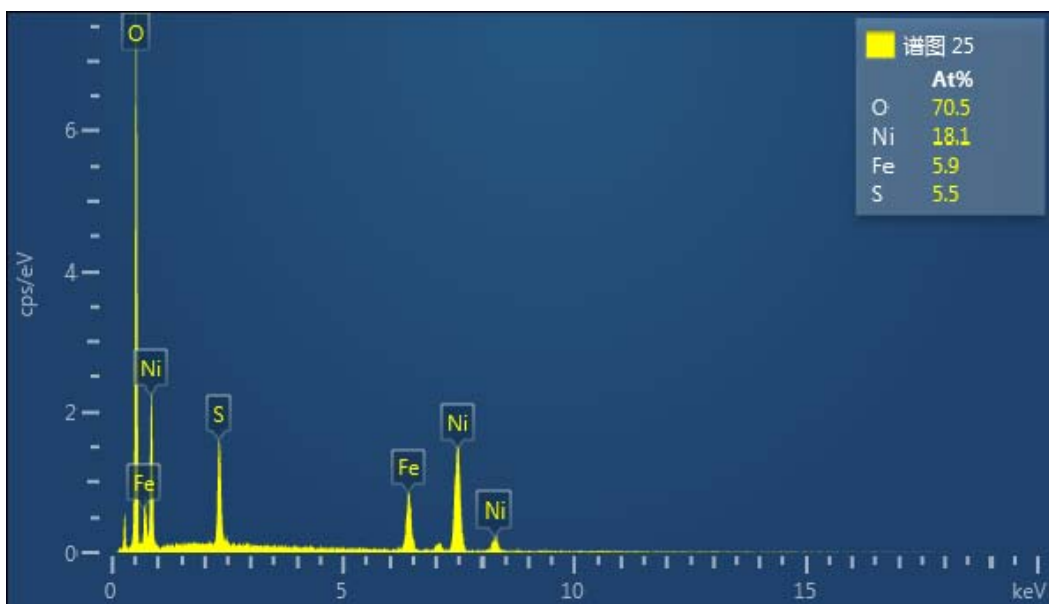
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**Fig. S1** (a) TEM image and (b) PXRD patterns of the products obtained under similar conditions of U-LDH( $\text{SO}_4^{2-}$ ) except that only water was used as the reaction solvent.

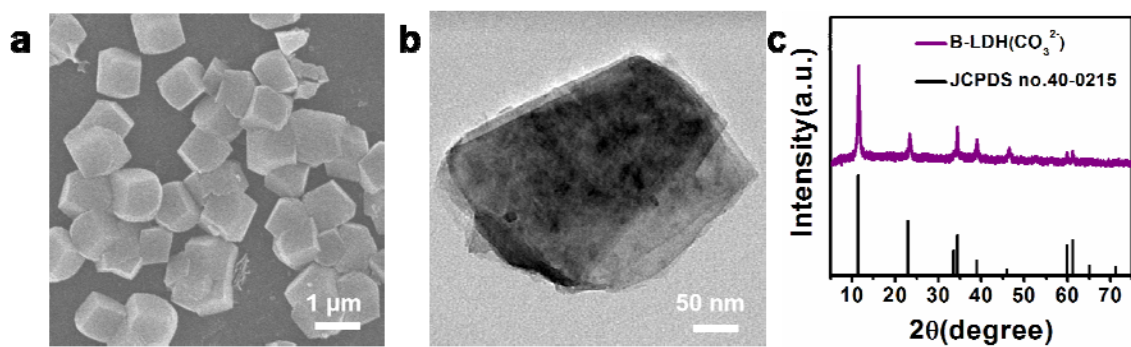


**Fig. S2** (a) TEM image and (b) PXRD patterns of the products obtained under similar conditions of U-LDH( $\text{SO}_4^{2-}$ ) except that only DMAC was used as the reaction solvent.

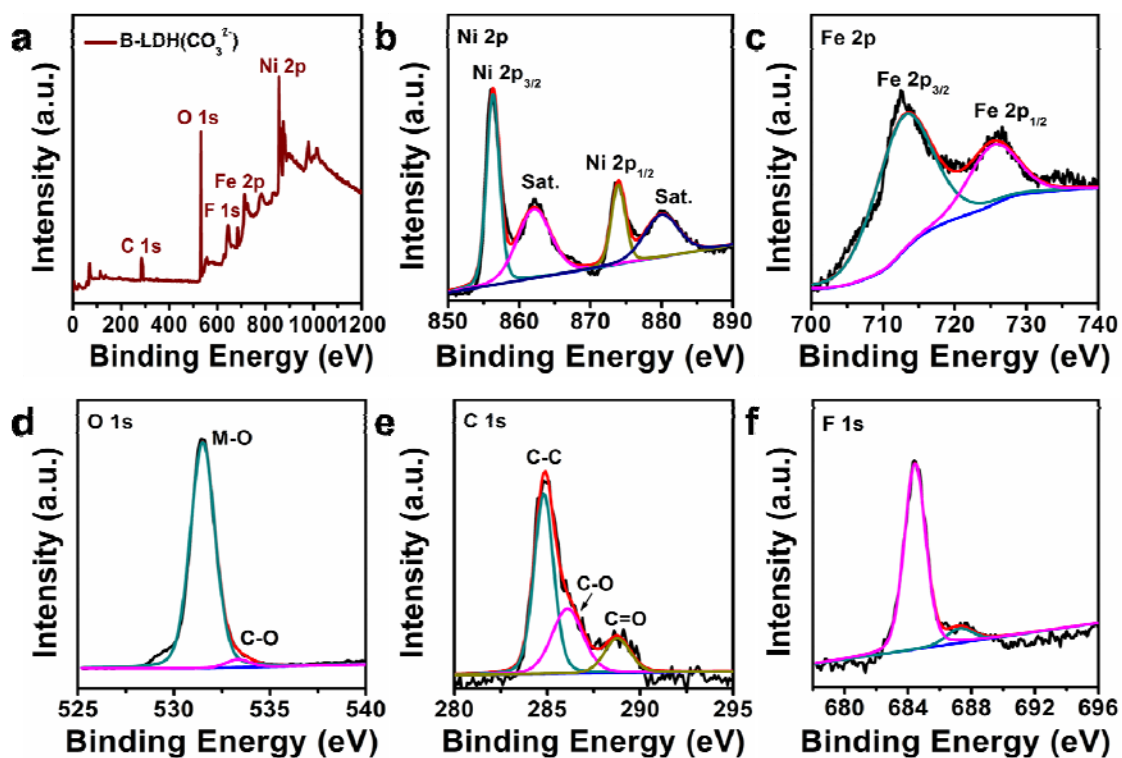


**Fig. S3** EDX spectrum of U-LDH(SO<sub>4</sub><sup>2-</sup>).

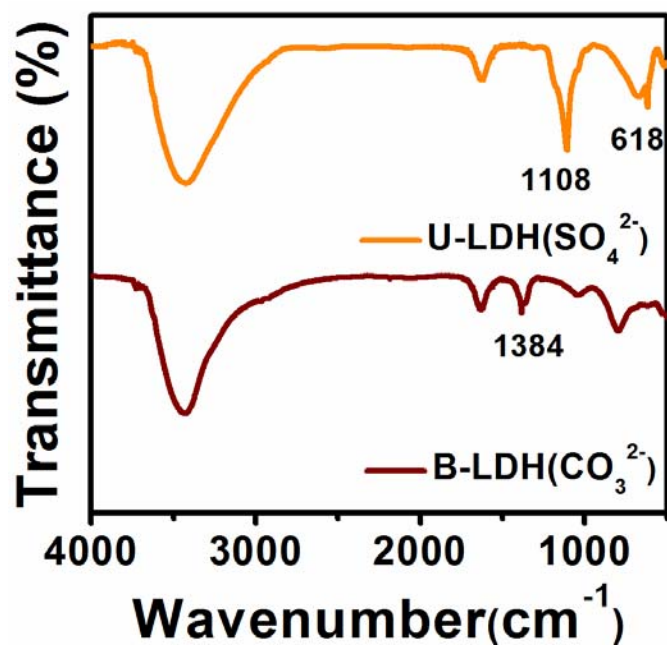
**Noted:**  $n(\text{Ni}) : n(\text{Fe}) : n(\text{S}) = 18.1 : 5.9 : 5.5 = 3 : 1 : 0.93$



**Fig. S4** (a) SEM image, (b) TEM image and (c) PXRD patterns of B-LDH(CO<sub>3</sub><sup>2-</sup>).



**Fig. S5** XPS spectra of B-LDH(CO<sub>3</sub><sup>2-</sup>): (a) survey scan, (b) Ni 2p, (c) Fe 2p, (d) O 1s, (e) C 1s, (f) F 1s.



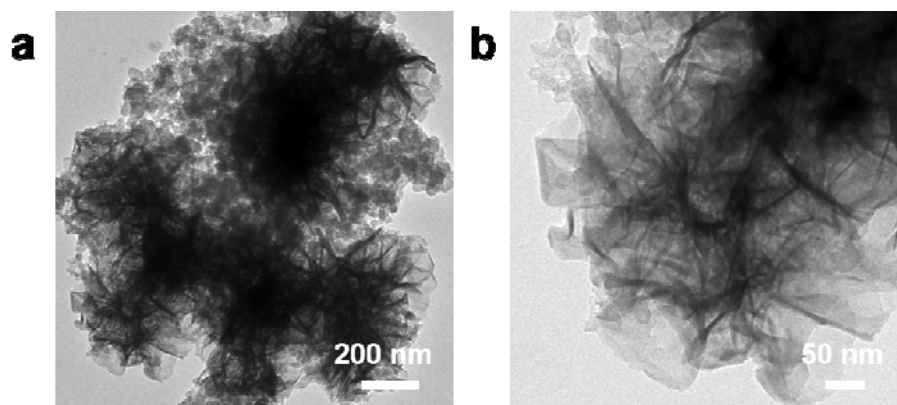
**Fig. S6** FT-IR spectra of U-LDH(SO<sub>4</sub><sup>2-</sup>) and B-LDH(CO<sub>3</sub><sup>2-</sup>).

**Noted:** The peak at 1384 cm<sup>-1</sup> is attributed to ν<sub>3</sub>(CO<sub>3</sub><sup>2-</sup>), while those at 1108 cm<sup>-1</sup> and 618 cm<sup>-1</sup> are attributed to the ν<sub>3</sub>(SO<sub>4</sub><sup>2-</sup>) and ν<sub>4</sub>(SO<sub>4</sub><sup>2-</sup>).

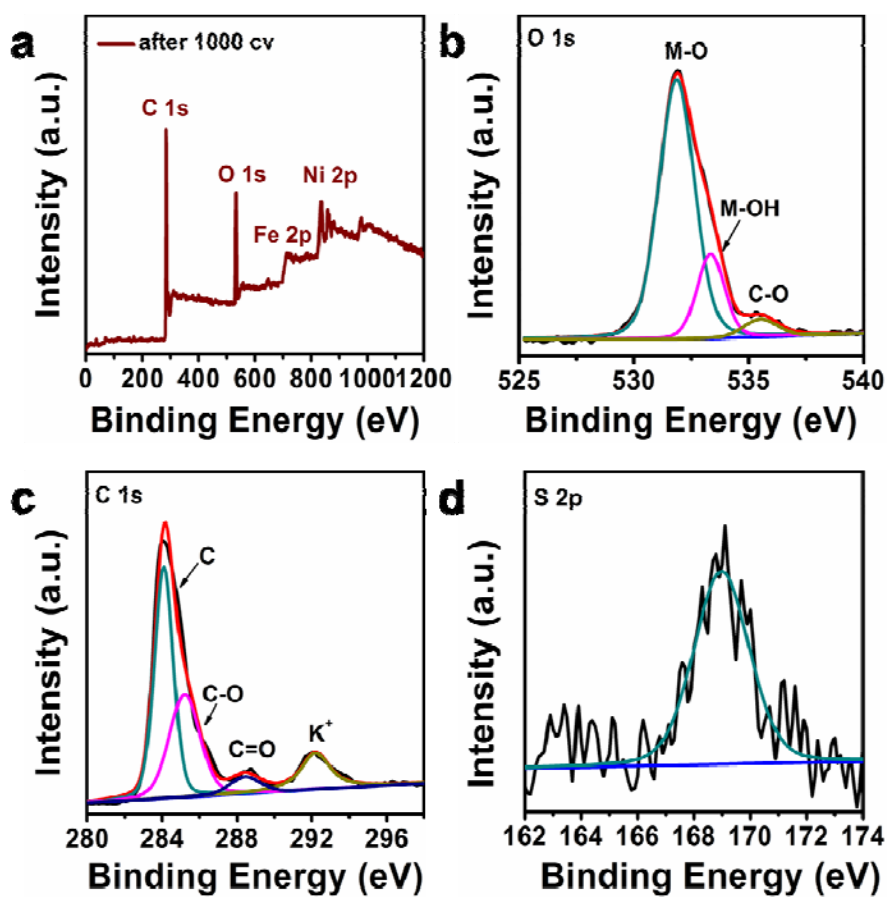
**Table S1** Comparisons of OER performance for Fe/Ni-based and LDH-based electrocatalysts in 1 M KOH.

Catalysts	Electrode	Overpotential (mV) at 10 mA·cm <sup>-2</sup>	Tafel slope (mV·dec <sup>-1</sup> )	Reference
U-LDH(SO <sub>4</sub> <sup>2-</sup> )	GCE	212	65.2	<b>This work</b>
SO <sub>4</sub> <sup>2-</sup> (EG) NiFe LDH	Carbon paper	375	56	<i>Chem. Mater.</i> , 2018, <b>30</b> , 4321-4330.
NiFe-SO <sub>4</sub>	Ni foam	356	93	<i>Chem. Mater.</i> , 2019, <b>31</b> , 6798-6807.
Ni-Fe LDH nanoprisms	GCE	280	49.4	<i>Angew. Chem. Int. Ed.</i> , 2018, <b>57</b> , 172-176.
Ni/NiO@CoFe LDH	Ni/NiO foam	230	34.3	<i>ChemSusChem</i> , 2019, <b>12</b> , 2773-2779.
NiFe/Cu <sub>2</sub> O NWs/CF	Cu foam	284	42	<i>ChemSusChem</i> , 2017, <b>10</b> , 1475-1481.
Fe(OH) <sub>3</sub> @Co-MOF-74	carbon paper	292	44	<i>ChemSusChem</i> , 2019, <b>12</b> , 4623-4628.
NiFe-LDH-UF (UF: Ultrafine)	Graphite paper	254	32	<i>Adv. Energy Mater.</i> , 2018, <b>8</b> , 1703585.
NiFe hydroxide	GCE	270	36.2	<i>Angew. Chem. Int. Ed.</i> , 2019, <b>58</b> , 736-740.
δ-FeOOH NSs/NF	Ni foam	265	36	<i>Adv. Mater.</i> , 2018, <b>30</b> , 1803144.
CoMn-LDH	GCE	325	43	<i>J. Am. Chem. Soc.</i> , 2014, <b>136</b> , 16481-16484.
NiFeRu LDH/Ni foam	Ni foam	225	32.4	<i>Adv. Mater.</i> , 2018, <b>30</b> ,

				1706279.
Ni <sub>0.75</sub> Fe <sub>0.125</sub> V <sub>0.125</sub> -LDHs/ NF	Ni foam	231	39.4	<i>Small</i> , 2018, <b>14</b> , 1703257.
Cu@CoFe LDH	Cu foam	240	44.4	<i>Nano Energy</i> , 2017, <b>41</b> , 327-336.
CoFe LDHs-Ar	GCE	266	37.85	<i>Angew. Chem. Int. Ed.</i> , 2017, <b>56</b> , 5867-5871.

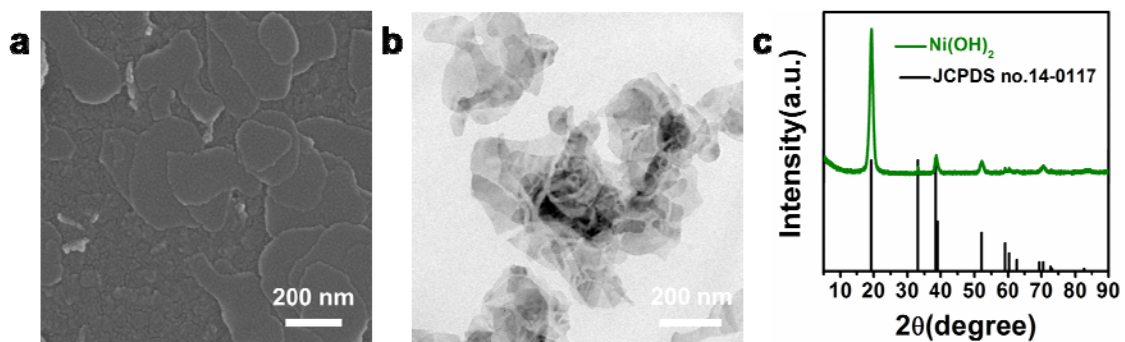


**Fig. S7** TEM images of U-LDH(SO<sub>4</sub><sup>2-</sup>) after 1000 CV cycles.

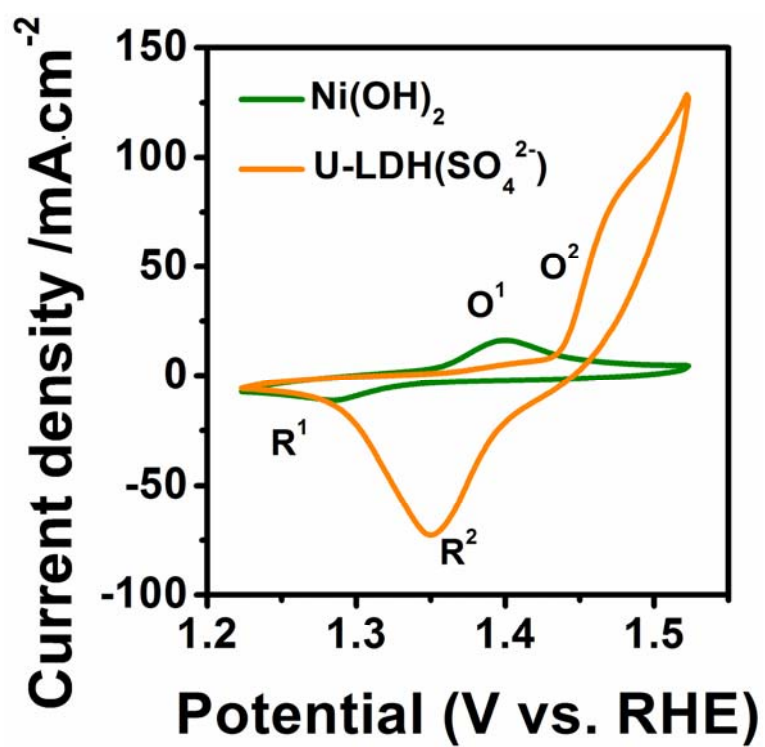


**Fig. S8** XPS spectra of U-LDH(SO<sub>4</sub><sup>2-</sup>) after 1000 CV cycles: (a) survey scan, (b) O 1s, (c) C 1s, (d) S 2p.

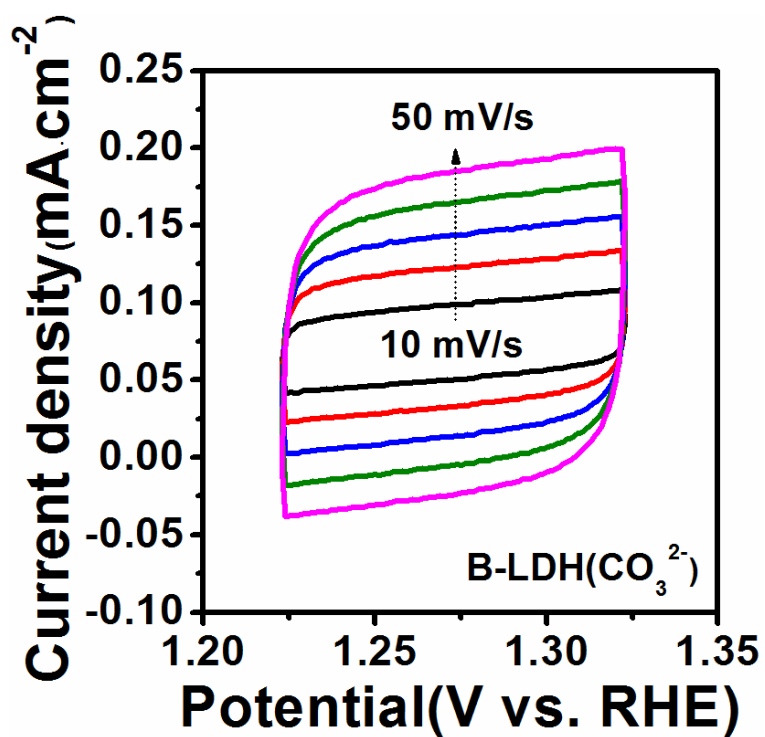




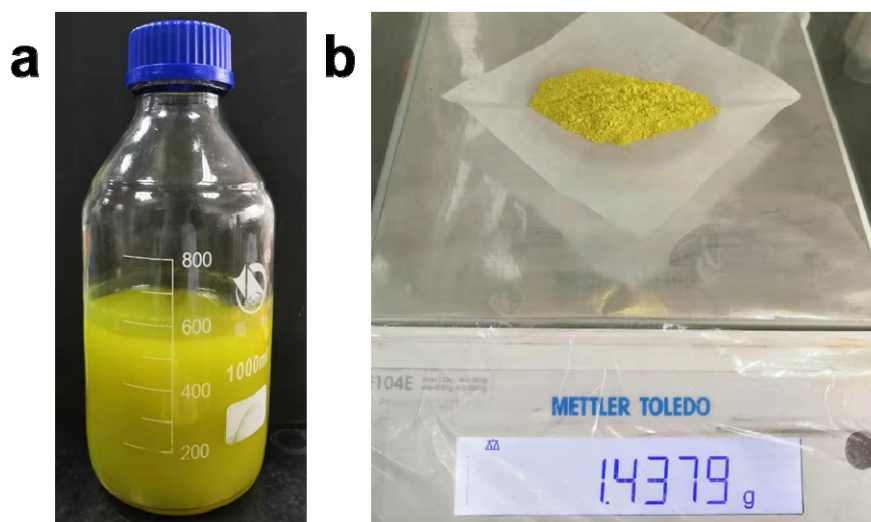
**Fig. S9** (a) SEM image, (b) TEM image, (c) PXRD patterns of  $\text{Ni}(\text{OH})_2$ .



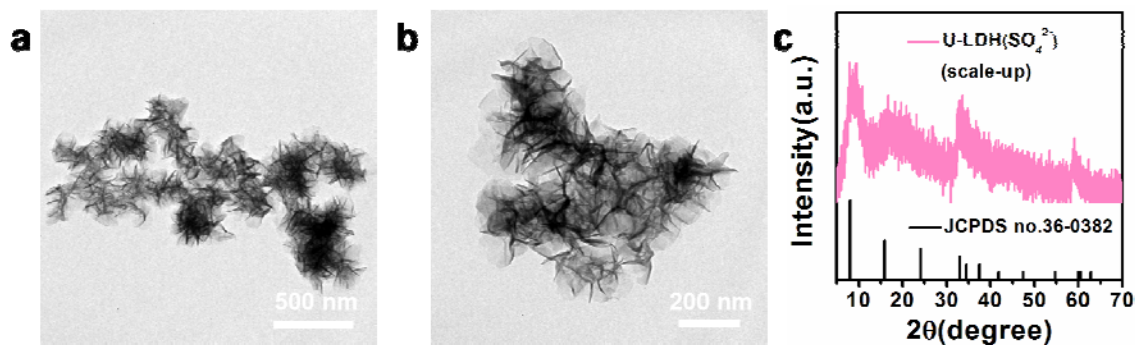
**Fig. S10** CV curves of  $\text{Ni}(\text{OH})_2$  and  $\text{U-LDH}(\text{SO}_4^{2-})$  at a scan rate of  $5 \text{ mV} \cdot \text{s}^{-1}$ .



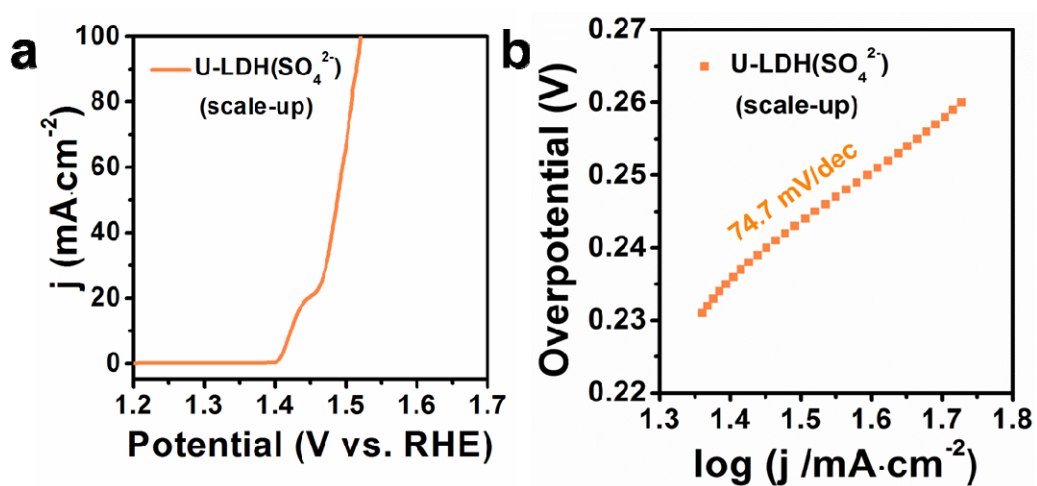
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**Fig. S14** (a) Linear sweep voltammetry OER curve and (b) Tafel plot of U-LDH(SO<sub>4</sub><sup>2-</sup>) prepared in gram-scale.