# Engineering Cu/NiCu LDH Heterostructure Nanosheet Arrays for Highly-Efficient Water Oxidation

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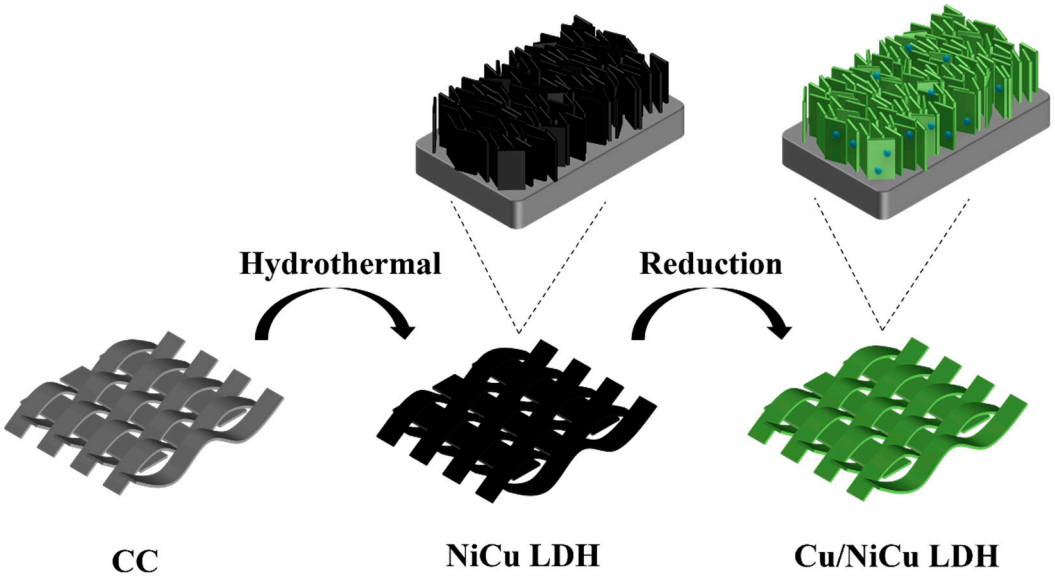


**Figure S1**. Schematic diagram of the preparation process of Cu/NiCu LDH.

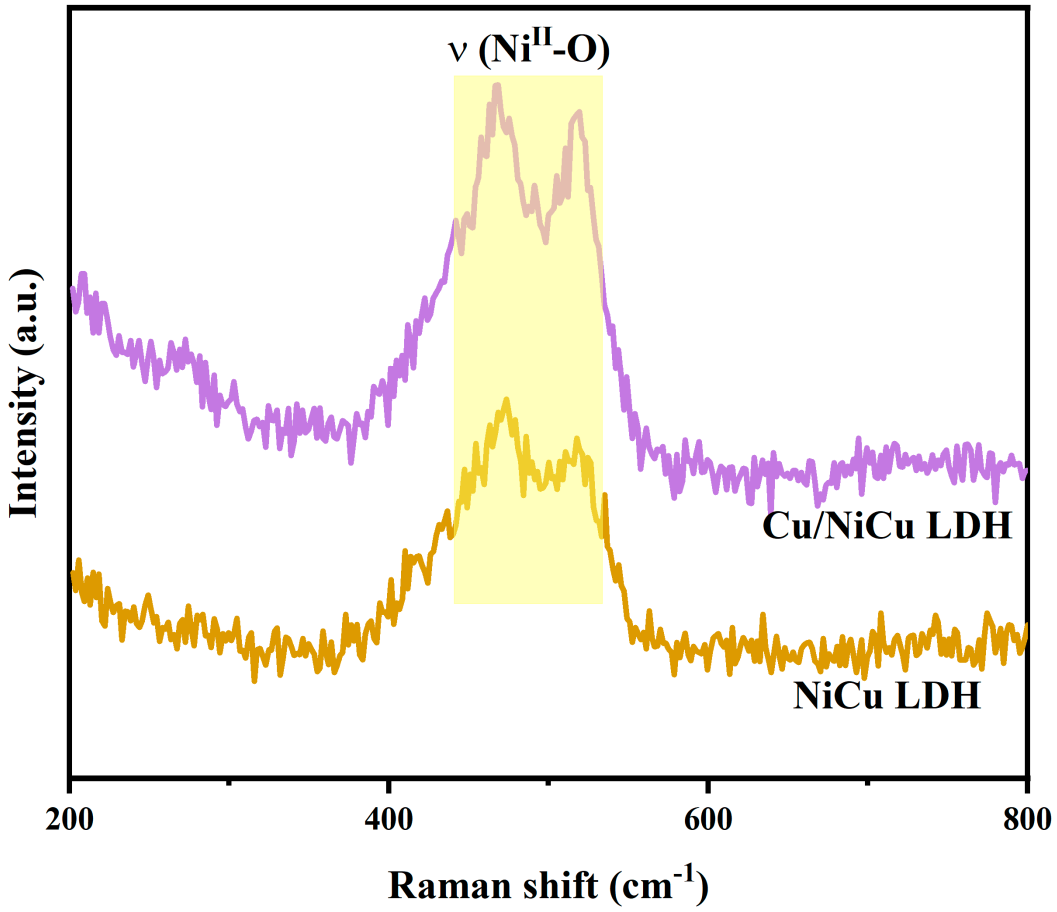


**Figure S2.** Raman spectrum of the synthesized NiCu LDH and Cu/NiCu LDH.

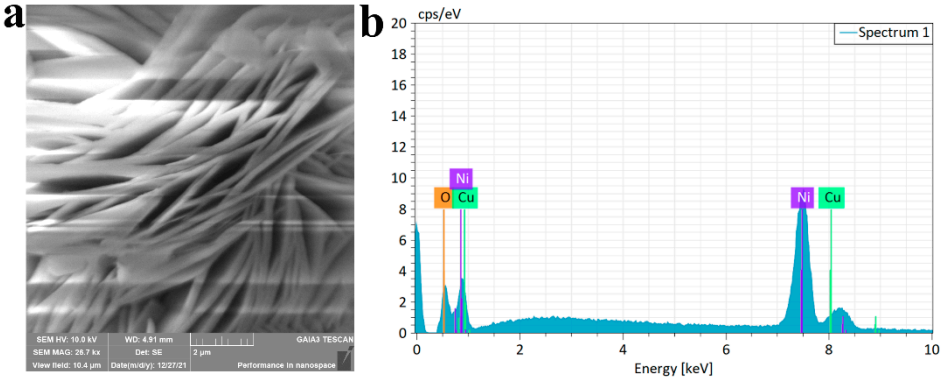


**Figure S3**. (a) SEM image and (b) EDS spectrum of NiCu LDH.

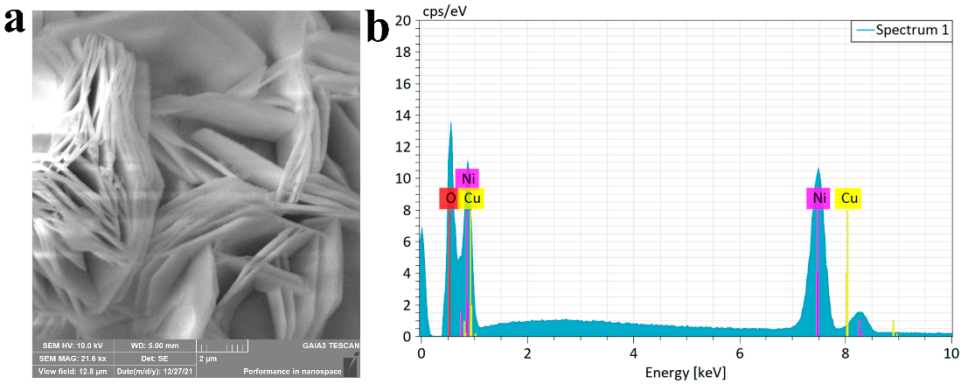


**Figure S4**. (a) SEM image and (b) EDS spectrum of $Cu_{1.25}$/NiCu LDH.

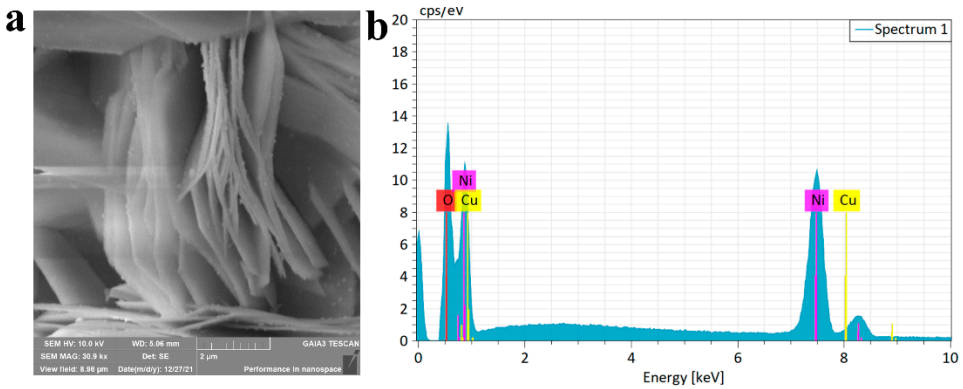


**Figure S5**. EDS spectrum of $Cu_{2.5}$/NiCu LDH.

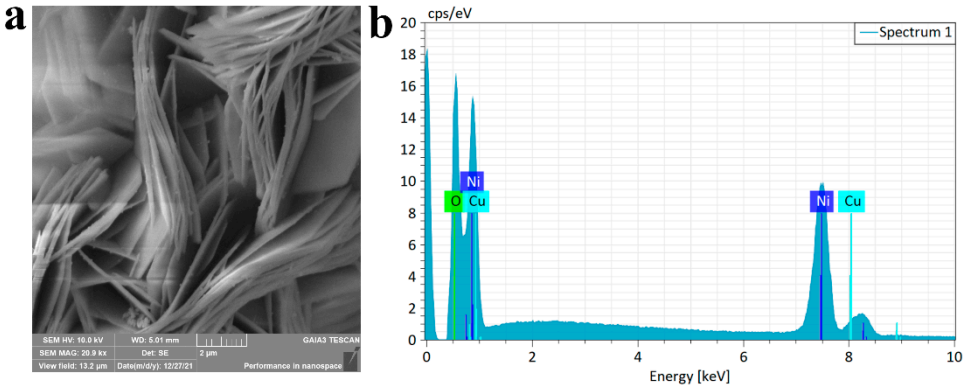


**Figure S6**. (a) SEM image and (b) EDS spectrum of $Cu_5$/NiCu LDH.

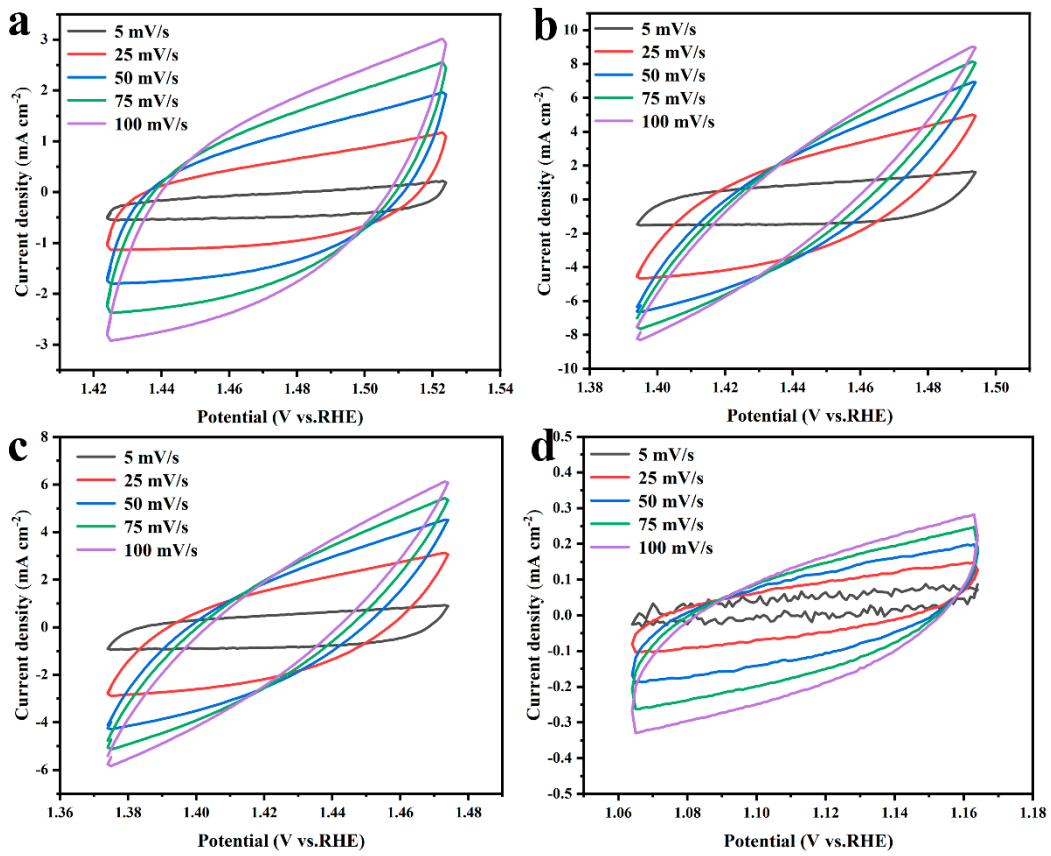


**Figure S7**. CV curves of (a) NiCu LDH, (b) $Cu_{1.25}$/NiCu LDH, (c) $Cu_{2.5}$/NiCu LDH and (d) $Cu_5$/NiCu LDH.

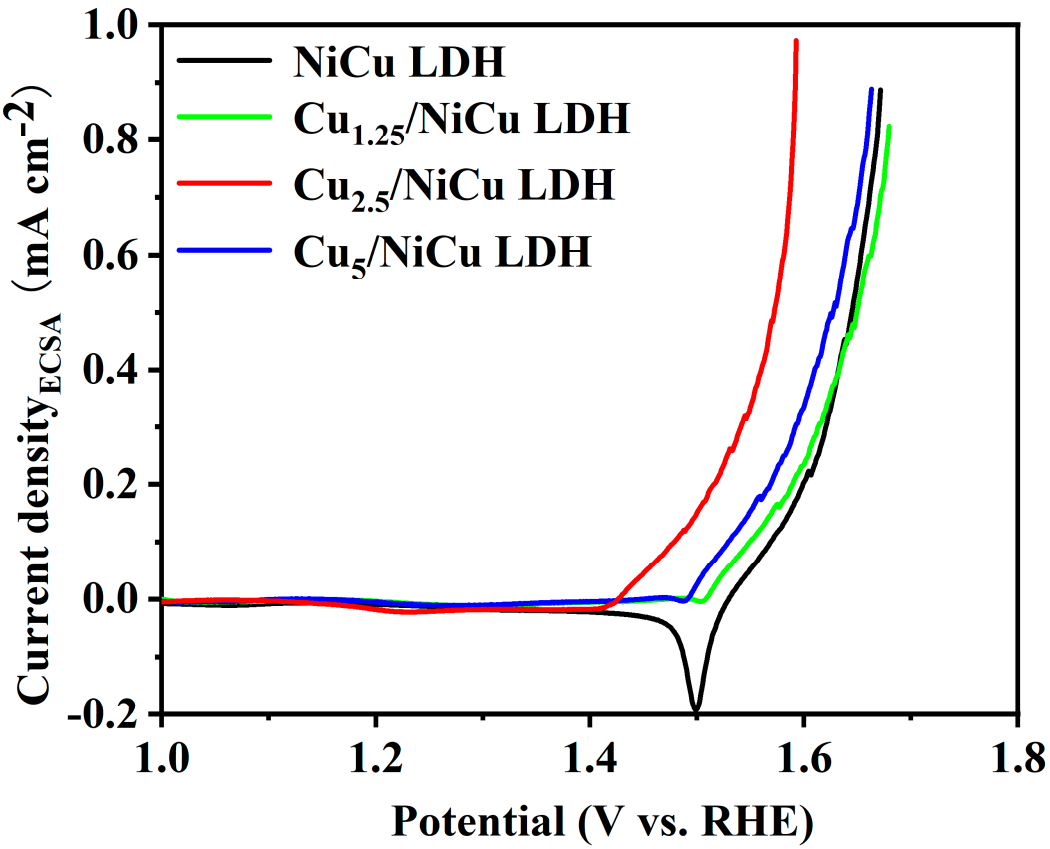


**Figure S8**. Normalized LSV curves using the ECSA value of Cu/NiCu LDH, $Cu_{1.25}$/NiCu LDH, $Cu_{2.5}$/NiCu LDH and $Cu_5$/NiCu LDH.

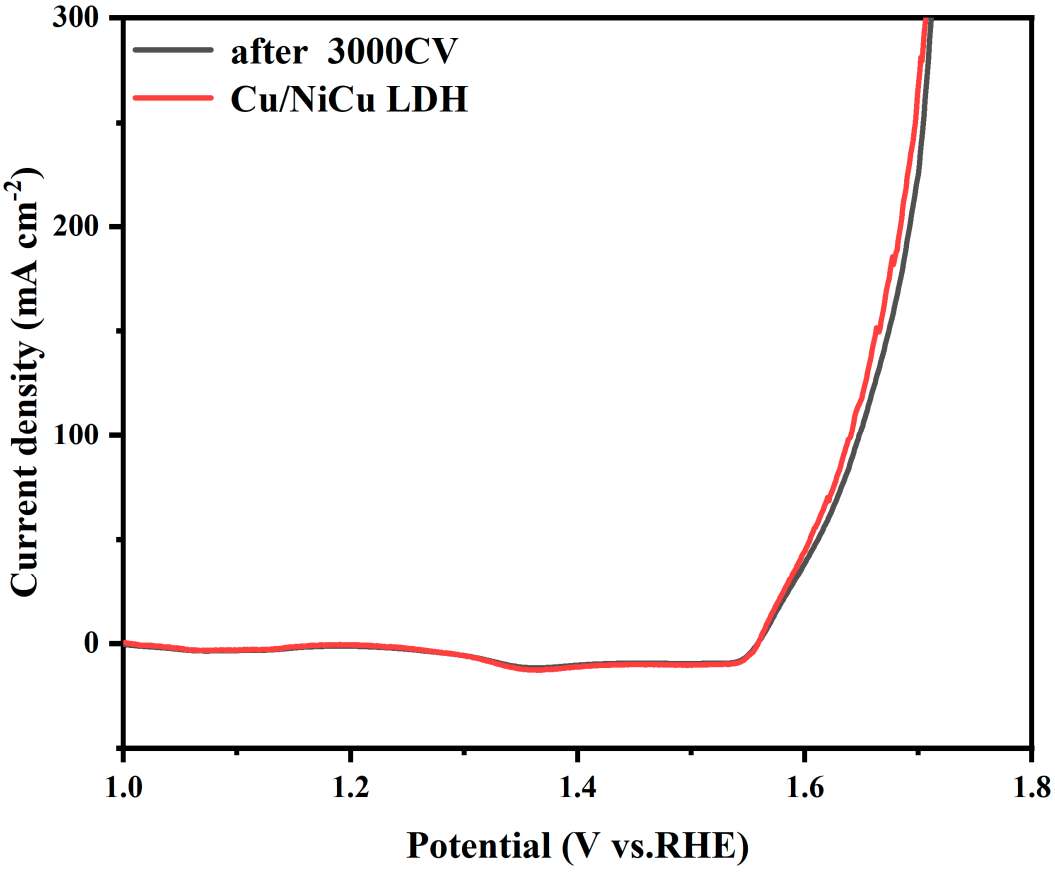


**Figure S9**. The LSV curves of Cu/NiCu LDH before and after 3000 CV tests.

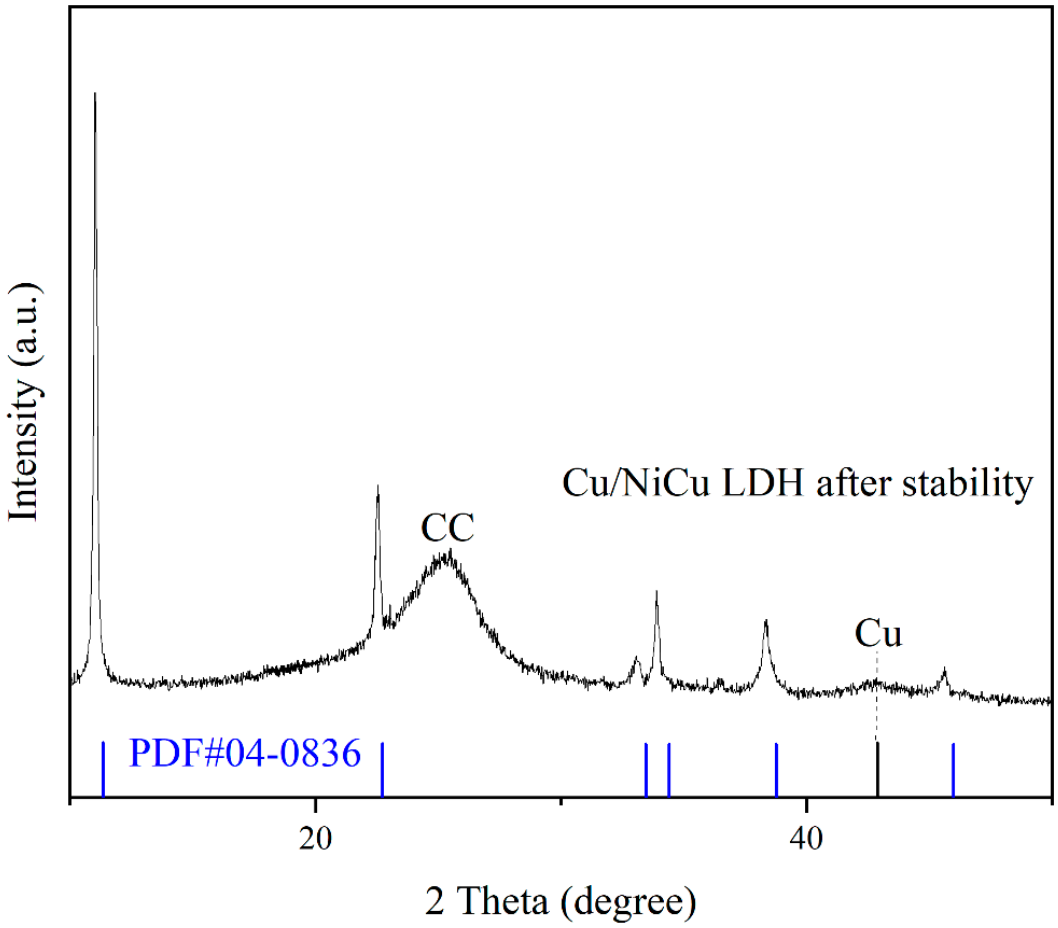


**Figure S10.** XRD pattern of $Cu_{2.5}$/NiCu LDH after stability test.

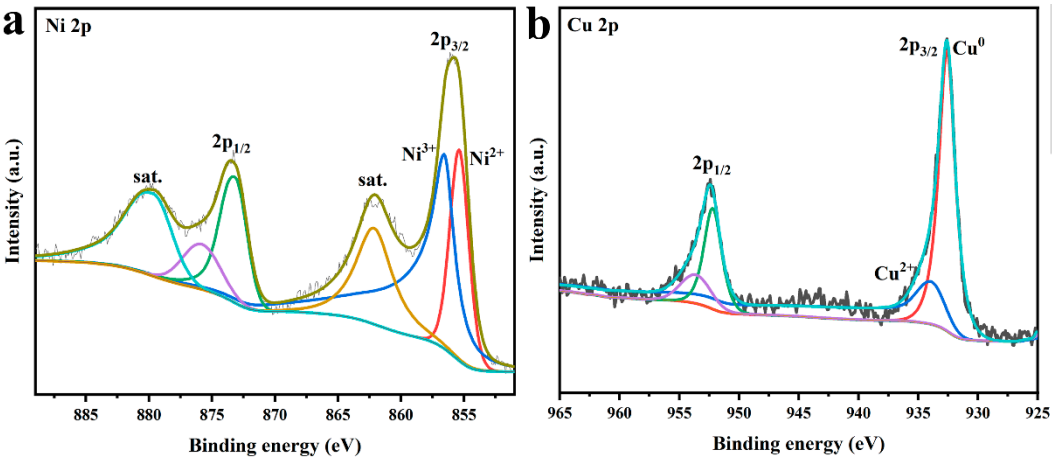


**Figure S11.** (a) Ni 2p and (b) Cu 2p XPS spectra of $Cu_{2.5}$/NiCu LDH after stability.

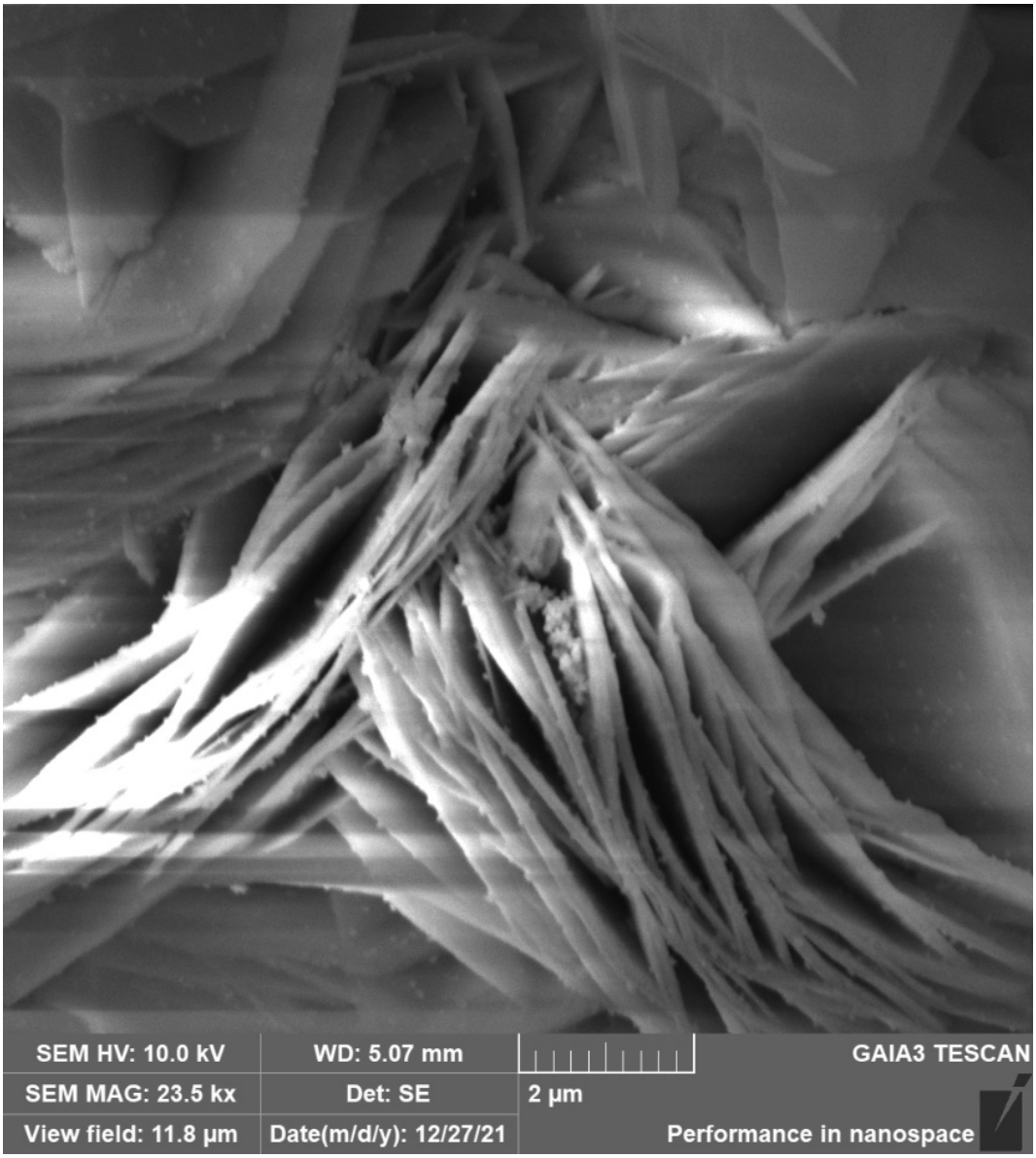


**Figure S12**. SEM image of $Cu_{2.5}$/NiCu LDH after stability test.

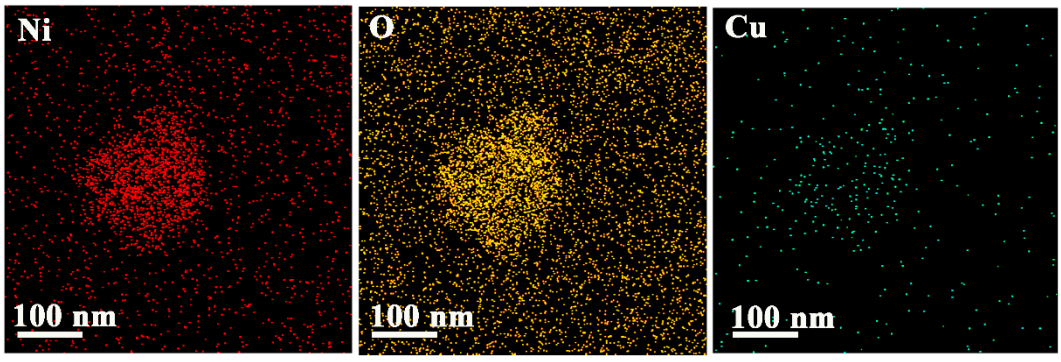


**Figure S13**. Element mapping of $Cu_{2.5}$/NiCu LDH after stability test.

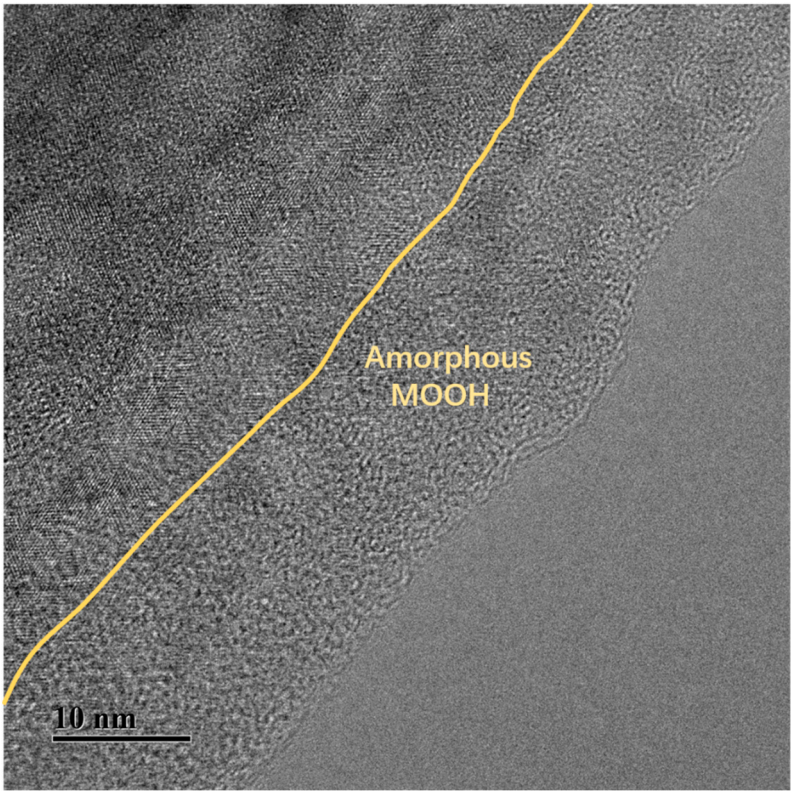


**Figure S14**. HRTEM image of $Cu_{2.5}$/NiCu LDH after stability test.

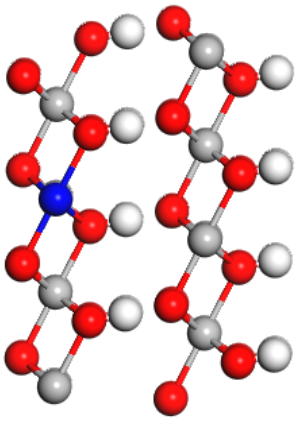

**Figure S15**. Constructed model of NiCuOOH.

**Table S1.** EDS results of NiCu LDH, $Cu_{1.25}$/NiCu LDH, $Cu_{2.5}$/NiCu LDH and $Cu_5$/NiCu LDH.

| | Ni (at%) | Cu(at%) | O (at%) |
|---|---|---|---|
| NiCu LDH | 34.58 | 1.08 | 64.34 |
| $Cu_{1.25}$/NiCu LDH | 25.94 | 1.95 | 72.10 |

| | | | |
|---|---|---|---|
| $Cu_{2.5}$/NiCu LDH | 27.86 | 2.52 | 69.62 |
| $Cu_5$/NiCu LDH | 30.34 | 6.14 | 63.52 |
| $Cu_{2.5}$/NiCu LDH after stability test | 20.7 | 2.31 | 76.99 |

**Table S2.** Comparison of OER performances of $Cu_{2.5}$/NiCu LDH with previously reported well-performed OER electrocatalysts.

| Catalysts | η@10 mA $cm^{-2}$ (mV) | Electrolyte | Substrate | Ref. |
|---|---|---|---|---|
| $Cu_{2.5}$/NiCu LDH | 206 | 1M KOH | CC | This work |
| Mxene/NiFe LDH | 260 | 1M KOH | RDE | [1] |
| GDY/NiFe LDH | 260 | 1M KOH | CF | [2] |
| Pt/NiFe LDH | 230 | 1M KOH | CC | [3] |
| $Ni_2Cr_1$ LDH/NF | 319 | 1M KOH | NF | [4] |
| S–NiFe LDH/CC | 206 | 1M KOH | CC | [5] |
| Ni/LDH-ZnO | 210 | 1M KOH | NF | [6] |
| Exfoliated NiFe LDH | 302 | 1M KOH | GC | [7] |
| NiO/NiFe LDH | 270 | 1M KOH | GC | [8] |
| Fe–Ni LDH/MOFs | 255 | 1M KOH | GC | [9] |

Glassy carbon (GC); Ni foam (NF); carbon cloth (CC); Cu foam (CF); rotating disk electrode (RDE).

**Table S3.** ECSA values of NiCu LDH, $Cu_{1.25}$/NiCu LDH, $Cu_{2.5}$/NiCu LDH and $Cu_5$/NiCu LDH.

| | NiCu LDH | $Cu_{1.25}$/NiCu LDH | $Cu_{2.5}$/NiCu LDH | $Cu_5$/NiCu LDH |
|---|---|---|---|---|
| ECSA values | 324.75 | 447.75 | 523.25 | 415.5 |

**Reference**


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