

## Supplementary Information

### **High-Conductivity NiFe@Ni@Cu Composite Electrodes for Durable, Efficient Industrial Oxygen Evolution**

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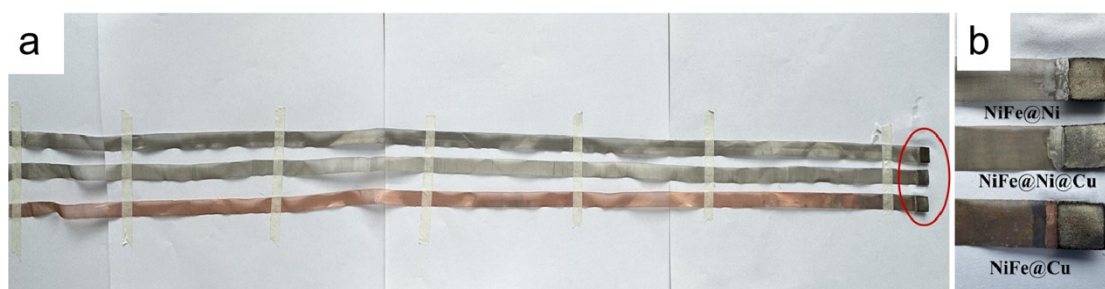
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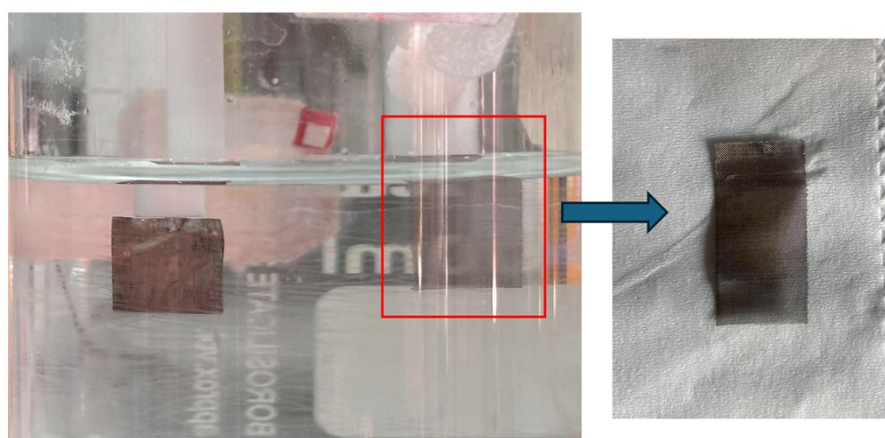
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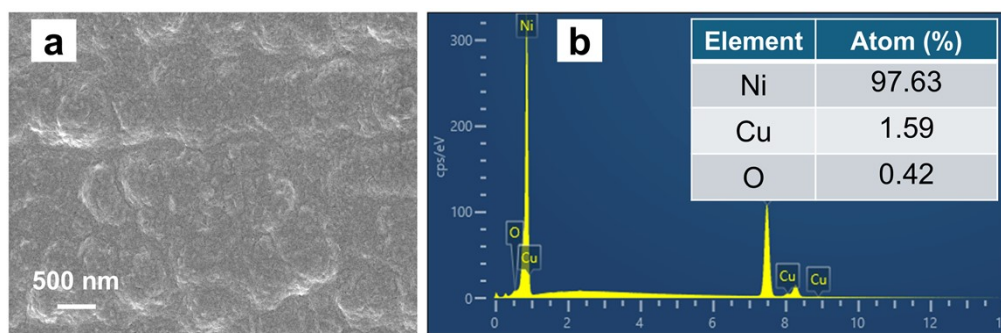
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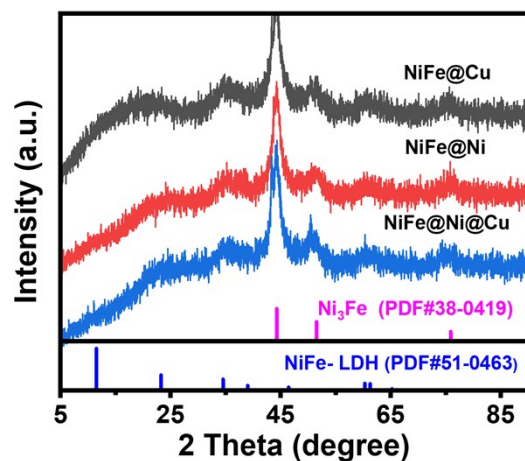
**Figure S1.** (a) Digital photograph of NiFe@Cu, NiFe@Ni, and NiFe@Ni@Cu. (b) The locally magnified digital photograph of NiFe coating.



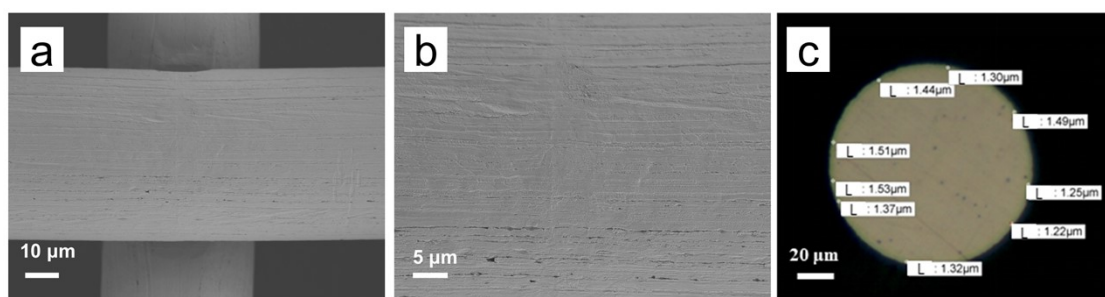
**Figure S2.** Digital photographs of Ni@Cu after 80-hour stability test.



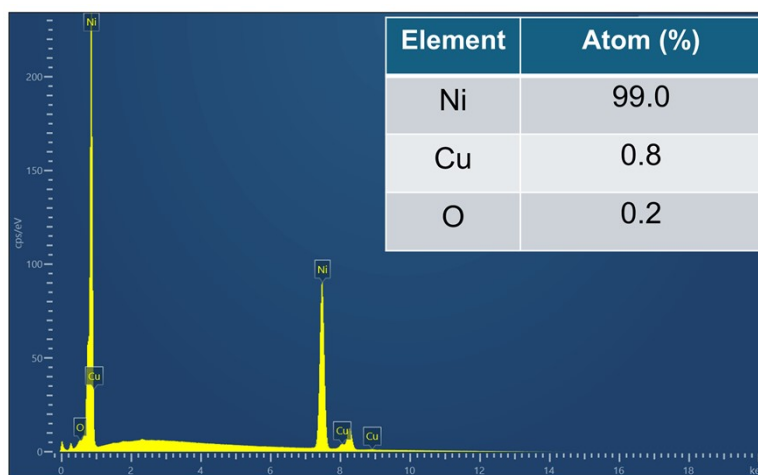
**Figure S3.** (a) SEM image and (b) EDS spectrum of Ni@Cu after 80-hour stability test.



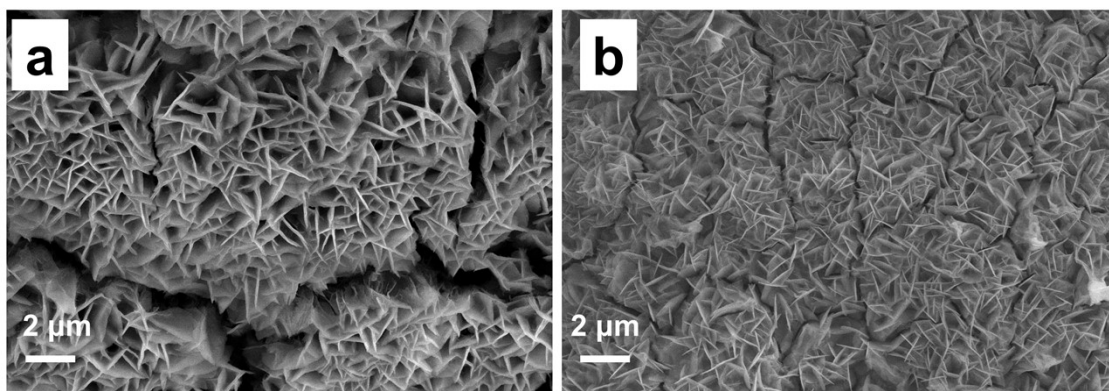
**Figure S4.** XRD patterns of NiFe@Cu, NiFe@Ni, and NiFe@Ni@Cu powder samples.



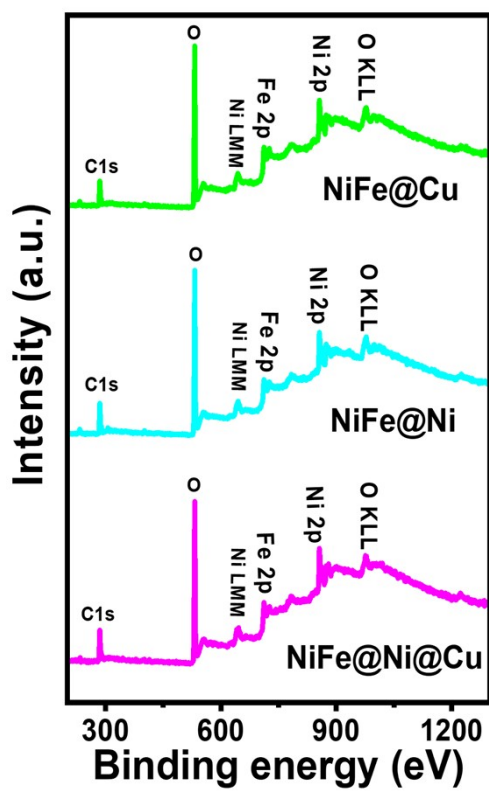
**Figure S5.** (a, b) SEM images of Cu, (c) Cross-section metallographic diagram of Ni@Cu.



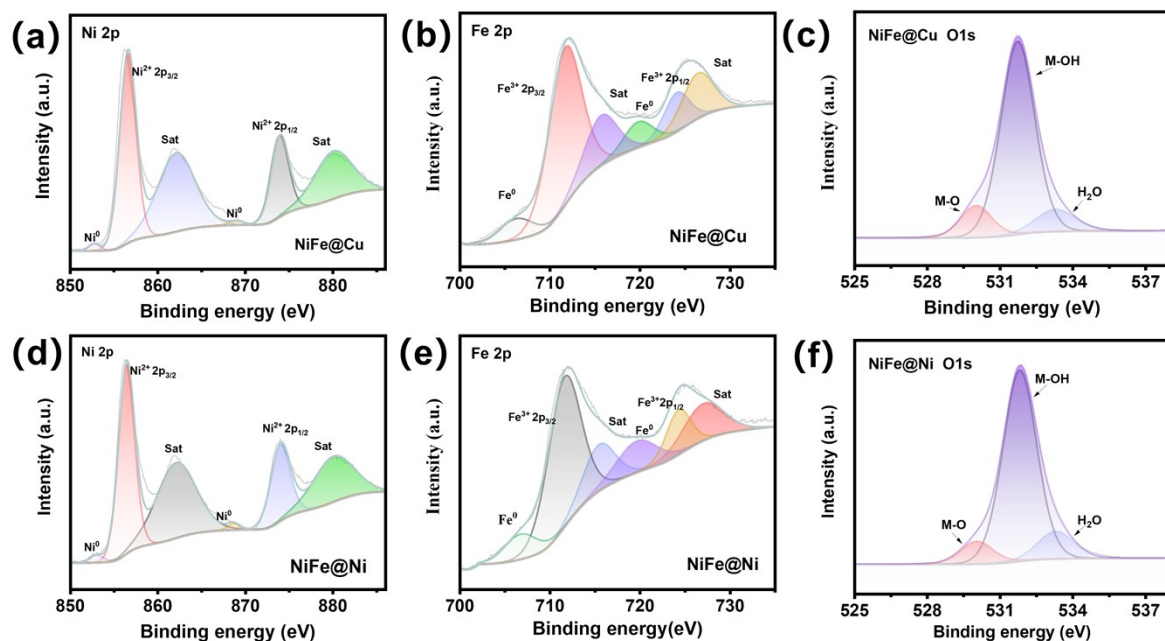
**Figure S6.** EDS spectrum of Ni@Cu.



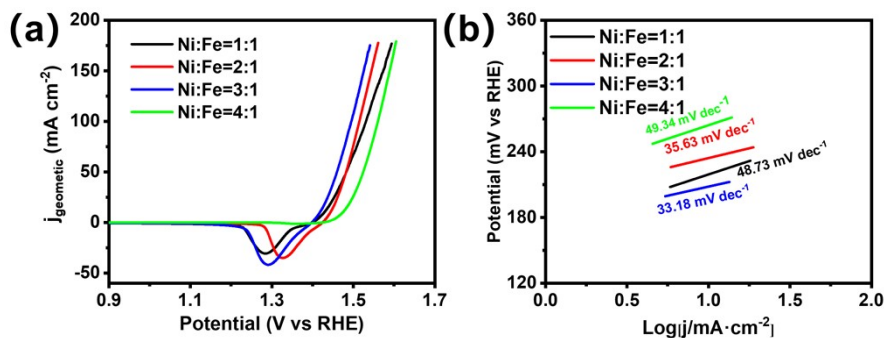
**Figure S7.** SEM images of (a) NiFe@Cu, and (b) NiFe@Ni.



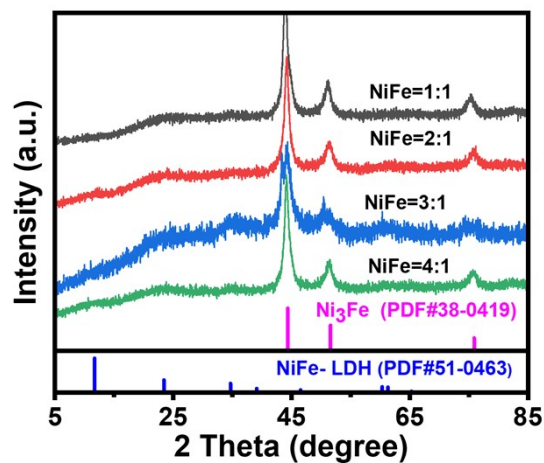
**Figure S8.** XPS survey spectra.



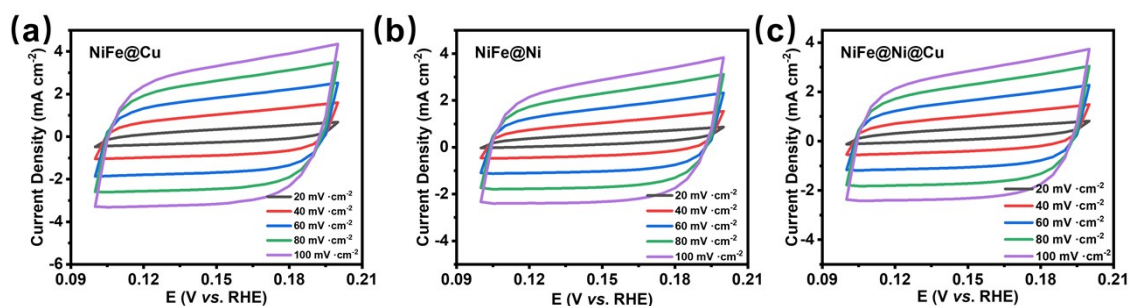
**Figure S9.** XPS spectra of NiFe@Cu (a) Ni 2p, (b) Fe 2p, (c) O 1s. XPS spectra of NiFe@Ni (d) Ni 2p, (e) Fe 2p, (f) O 1s.



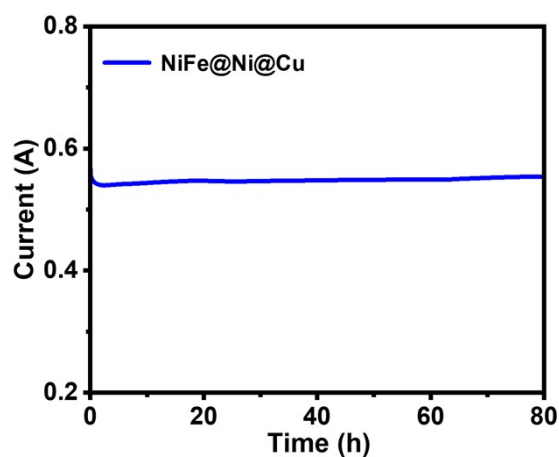
**Figure S10.** (a) LSV curves and (b) Tafel plots of samples with different  $\text{Ni}^{2+}/\text{Fe}^{2+}$  molar ratios.



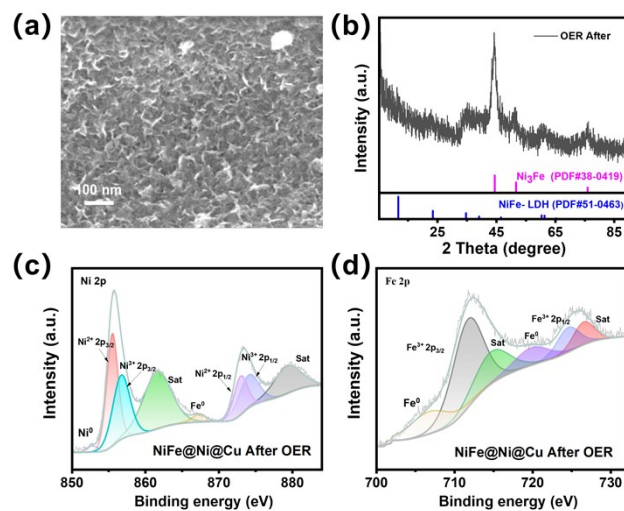
**Figure S11.** XRD patterns of samples with different  $\text{Ni}^{2+}/\text{Fe}^{2+}$  molar ratios.



**Figure S12.** CV curves at different scan rates of (a) NiFe@Cu, (b) NiFe@Ni, and (c) NiFe@Ni@Cu, measured in the non-Faradaic potential region to evaluate double-layer capacitance.



**Figure S13.** Stability test of NiFe@Ni@Cu.



**Figure S14.** (a) SEM image, (b) XRD pattern, (c) Ni 2p and (d) Fe 2p spectra of NiFe@Ni@Cu after the stability test.

## Supporting Tables

**Table S1.** Tafel fitting results

| Sample | $E_{\text{corr}}$ (V) | $I_{\text{corr}}$ (A cm <sup>-2</sup> ) | Corrosion Rate (mm / y) |
|--------|-----------------------|-----------------------------------------|-------------------------|
| Cu     | -0.48                 | $1.034 \times 10^{-4}$                  | 0.338                   |
| Ni     | -0.0313               | $8.785 \times 10^{-6}$                  | 0.0575                  |
| Ni@Cu  | 0.0133                | $8.855 \times 10^{-6}$                  | 0.0288                  |

**Table S2.** Comparison table of OER electrocatalysts in 1 M KOH: this work vs. literature

| Catalyst                                                                    | Voltage / mV<br>(10 mA cm <sup>-2</sup> ) | Tafel slope<br>(mV dec <sup>-1</sup> ) | Reference |
|-----------------------------------------------------------------------------|-------------------------------------------|----------------------------------------|-----------|
| NiFe@Ni@Cu                                                                  | 208                                       | 36.1                                   | This work |
| NiFe LDH-D1                                                                 | 241                                       | 54.9                                   | 1         |
| NiFeMo LDH/NF                                                               | 220                                       | 45.6                                   | 2         |
| NiFe-LDH/AISI 304                                                           | 230                                       | 52                                     | 3         |
| NiFe-BTC@ CNTs                                                              | 230                                       | 36                                     | 4         |
| NiFe LDH@ITO                                                                | 240                                       | 45                                     | 5         |
| NiFe ER 5                                                                   | 241                                       | 43                                     | 6         |
| NiFe-20-H                                                                   | 241                                       | 44.2                                   | 7         |
| (Ni <sub>0.75</sub> Fe <sub>0.25</sub> ) <sub>0.05</sub> Al <sub>0.05</sub> | 279                                       | 38                                     | 8         |
| Ni <sub>6</sub> Fe <sub>4</sub> LDH/NF                                      | 280                                       | 116                                    | 9         |
| hcp-NiFe@NC                                                                 | 226                                       | 41                                     | 10        |
| NiFe-35-120                                                                 | 242                                       | 56.64                                  | 11        |
| NiFe-LDH/SnS                                                                | 310                                       | 53.6                                   | 12        |
| Mo <sub>0.1</sub> -NiFe-LDH/N,S-rGO<br>(10 wt%)                             | 253                                       | 65                                     | 13        |
| SmNi <sub>0.02</sub> Fe-LD/NF                                               | 230                                       | 76.13                                  | 14        |

## REFERENCES

- (1) Zhang, B.; Qiu, S.; Xing, Y.; Zhao, G.; Liao, W.; Mu, L.; Zhao, N. Introduction of Cationic Vacancies into NiFe LDH by In Situ Etching To Improve Overall Water Splitting Performance. *Langmuir*. **2023**, *39* (49), 18152-18160.
- (2) Tang, X.; Li, Y.; Jiang, X.; Li, H.; Wang, Y.; Wang, X. High-valent metal molybdenum-doped NiFe LDH nanosheet for efficient oxygen evolution reaction. *J. Electroanal. Chem.* **2025**, *982*.
- (3) Patiño López, J. J.; Vasquez-Montoya, M. F.; Velásquez, C. A.; Cartagena, S.; Montoya, J. F.; Martinez-Puente, M. A.; Ramírez, D.; Jaramillo, F. Self-Supported Spray-Coated NiFe-LDH Catalyst on a Stainless Steel Substrate for Efficient Oxygen Evolution Reaction. *ACS Appl. Mater. Interfaces*. **2023**, *15* (48), 56547-56555.
- (4) Cho, S.; Oh, Y.; Nguyen, H. T.; Chae, K.; Tran, N. A. T.; Lee, Y.-W.; Hong, J.; Shin, D.; Cho, H.-S.; Cho, Y. Enhanced oxygen evolution reaction in alkaline water electrolysis using bimetallic NiFe metal-organic frameworks integrated with carbon nanotubes. *Int. J. Hydrogen Energy*. **2024**, *90*, 747-756.
- (5) Xu, J.; Li, Z.; Chen, D.; Yang, S.; Zheng, K.; Ruan, J.; Wu, Y.; Zhang, H.; Chen, J.; Xie, F.; et al. Porous Indium Tin Oxide-Supported NiFe LDH as a Highly Active Electrocatalyst in the Oxygen Evolution Reaction and Flexible Zinc-Air Batteries. *ACS Appl. Mater. Interfaces*. **2021**, *13* (41), 48774-48783.
- (6) Huang, X.; Kim, K. H.; Jang, H.; Luo, X.; Yu, J.; Li, Z.; Ao, Z.; Wang, J.; Zhang, H.; Chen, C.; O'Hare, D. Intrabasal Plane Defect Formation in NiFe Layered Double Hydroxides

Enabling Efficient Electrochemical Water Oxidation. *ACS Appl. Mater. Interfaces*. **2023**, *15* (46), 53815-53826.

(7) Wang, S.; Wang, H.; Chen, S.; Cheung, K. K. K.; Wong, H. F.; Leung, C. W.; Zapien, J. A. Hydrochloric acid etching induced flower-like NiFe-layered double hydroxide as efficient electrocatalyst for oxygen evolution reaction. *Int. J. Hydrogen Energy*. **2023**, *48* (45), 17045-17054.

(8) Lee, B.-J.; Jung, S.-M.; Yu, G.; Kim, H.-Y.; Kwon, J.; Kim, K.-S.; Kwak, J.; Lee, W.; Mok, D. H.; Back, S.; Kim, Y.-T. Highly Active and Stable Al-Doped NiFe Self-Supported Oxygen Evolution Reaction Electrode for Alkaline Water Electrolysis. *ACS Catal.* **2025**, *15* (2), 1123-1134.

(9) Sreenivasulu, M.; Hiremath, N. K. V.; Alshehri, M. A.; Shetti, N. P. A Green Solvent-Free Approach Synthesis for Rational Designing of a NiFe-Layered Double Hydroxide [NiFe-LDH] Electrocatalyst for Hydrogen Generation. *Energy Fuels*. **2024**, *38* (21), 20791-20806.

(10) Wang, C.; Yang, H.; Zhang, Y.; Wang, Q. NiFe Alloy Nanoparticles with hcp Crystal Structure Stimulate Superior Oxygen Evolution Reaction Electrocatalytic Activity. *Angew. Chem. Int. Ed.* **2019**, *58* (18), 6099-6103.

(11) Li, X.; Chen, Y.; Zheng, X.; Zhu, Y.; Wang, Z.; Wang, Y. One-step electrodeposition of composition-controllable dendritic NiFe alloy electrocatalysts for oxygen evolution reaction. *J. Alloys Compd.* **2023**, *968*, 172313.

(12) Sun, Y.; Cai, Q.; Wang, Z.; Li, Z.; Zhou, Q.; Li, X.; Zhao, D.; Lu, J.; Tian, S.; Li, Y.;

Wang, S. Two-Dimensional SnS Mediates NiFe-LDH-Layered Electrocatalyst toward Boosting OER Activity for Water Splitting. *ACS Appl. Mater. Interfaces*. **2024**, *16* (18), 23054-23060.

(13) Kumar, K. R.; Sri, N. D.; Kale, V. N.; Maiyalagan, T. Surface reconstruction of Mo-doped NiFe-LDH nanosheets decorated on nitrogen and sulfur dual-doped reduced graphene oxide as a bifunctional electrocatalyst for overall water splitting. *International Journal of Hydrogen Energy* **2025**, *101*, 837-847.

(14) Nagappan, S.; Gurusamy, H.; Minhas, H.; Karmakar, A.; Ravichandran, S.; Pathak, B.; Kundu, S. Unraveling the Synergistic Role of Sm<sup>3+</sup> Doped NiFe-LDH as High-Performance Electrocatalysts for Improved Anion Exchange Membrane and Water Splitting Applications. *Small Methods* **2024**, e2401655.