

## Supporting Information

### Papillae-like morphology of Ni/Ni(OH)<sub>2</sub> hybrid crystal by stepwise electrodeposition for synergistically improved HER

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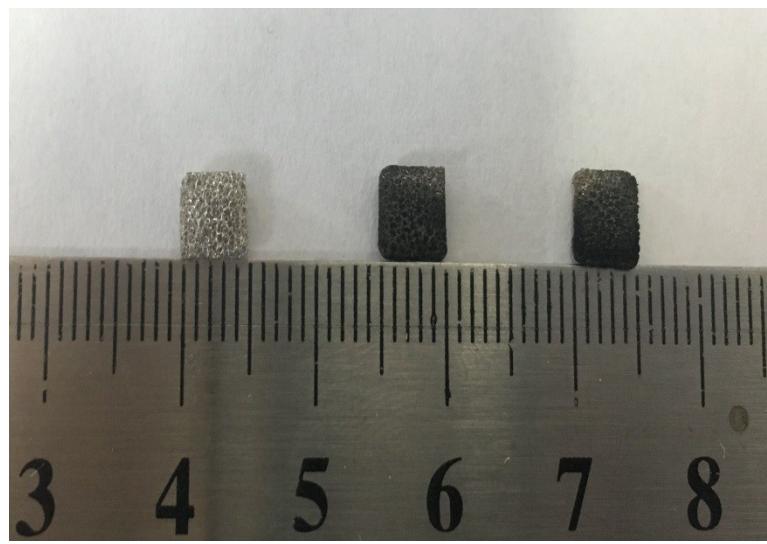


Figure S1. Photograph for the bare Ni foam (left), 1st-Ni/Ni(OH)<sub>2</sub>/NF (middle), and 2nd-Ni/Ni(OH)<sub>2</sub>/NF-500 (right).

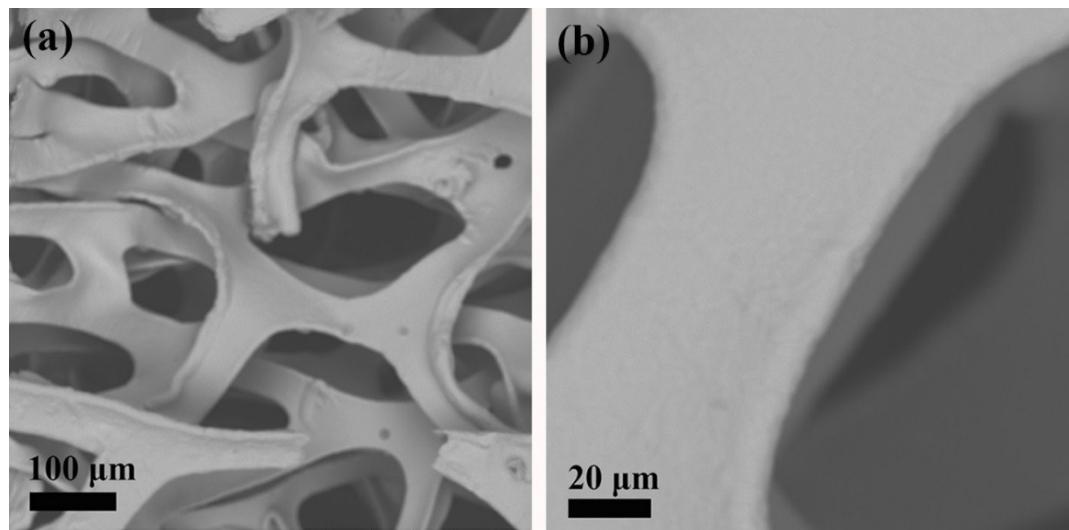


Figure S2. SEM images for bare Ni foam.

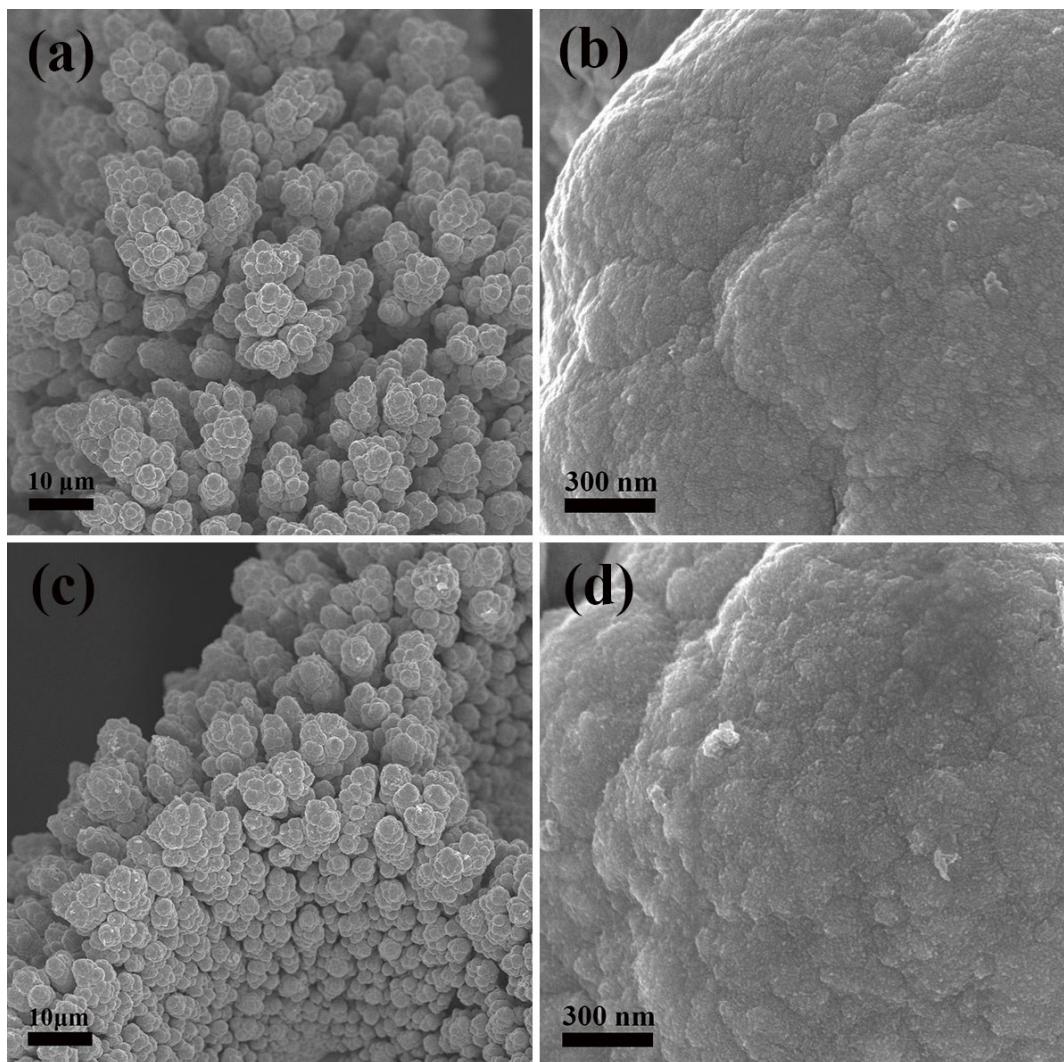


Figure S3. SEM images of 3rd-Ni/Ni(OH)<sub>2</sub>/NF-500 (a, b) and 4th-Ni/Ni(OH)<sub>2</sub>/NF-500 (c, d).

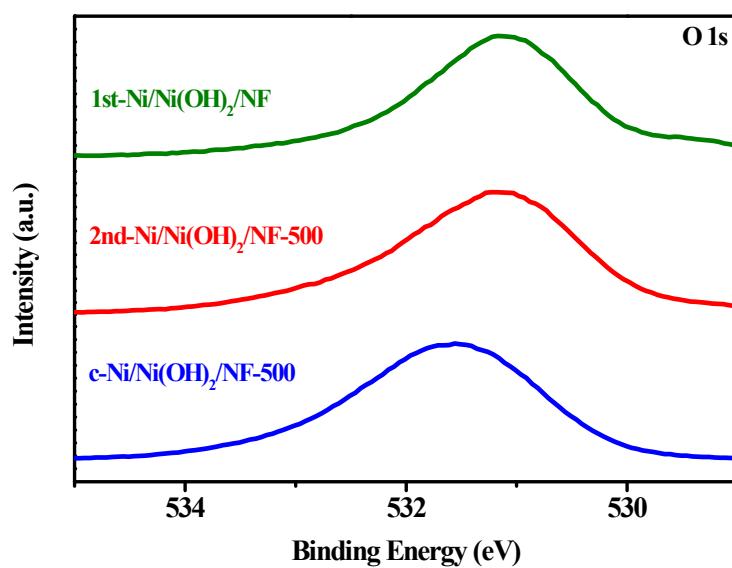


Figure S4. XPS spectra of O 1s region of 1st-Ni/Ni(OH)<sub>2</sub>/NF, 2nd-Ni/Ni(OH)<sub>2</sub>/NF-500 and c-Ni/Ni(OH)<sub>2</sub>/NF-500.

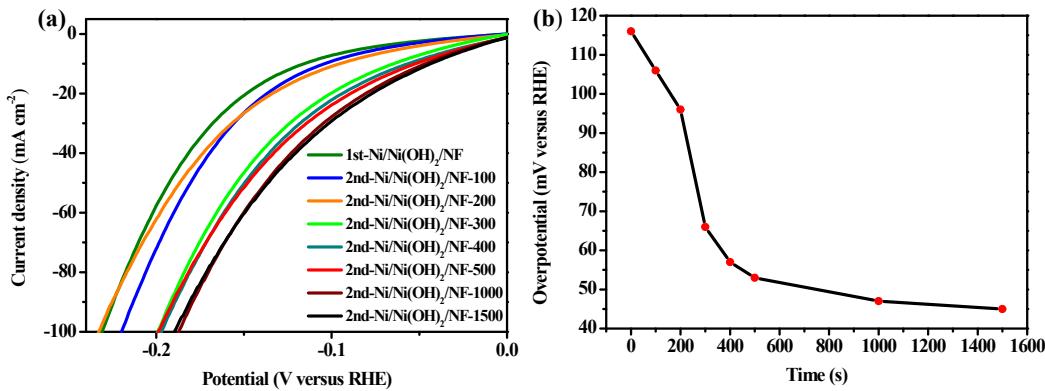


Figure S5. (a) The LSV curves for 1st-Ni/Ni(OH)<sub>2</sub>/NF, 2nd-Ni/Ni(OH)<sub>2</sub>/NF-100, 2nd-Ni/Ni(OH)<sub>2</sub>/NF-200, 2nd-Ni/Ni(OH)<sub>2</sub>/NF-300, 2nd-Ni/Ni(OH)<sub>2</sub>/NF-400, 2nd-Ni/Ni(OH)<sub>2</sub>/NF-500, 2nd-Ni/Ni(OH)<sub>2</sub>/NF-1000, 2nd-Ni/Ni(OH)<sub>2</sub>/NF-1500 at a scan rate of  $2 \text{ mV s}^{-1}$  in  $1.0 \text{ M KOH}$  for HER. (b) The overpotential of HER activity at  $j = 10 \text{ mA cm}^{-2}$  with different second step electrodeposition time.

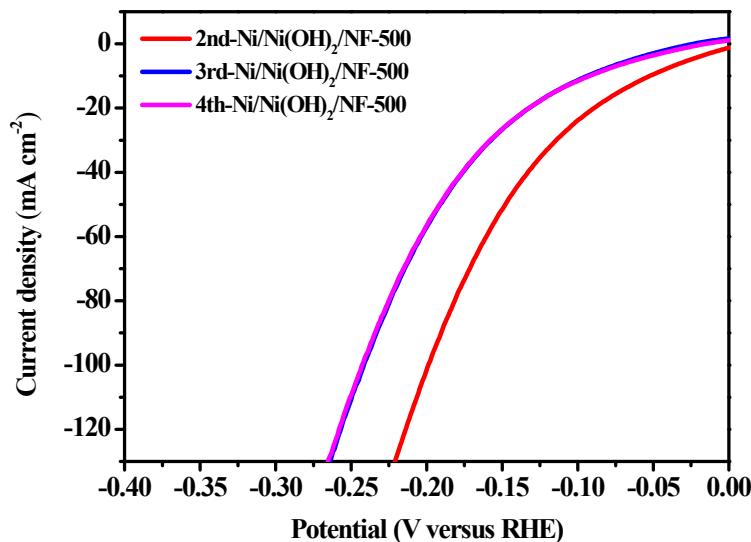


Figure S6. The LSV curves for 2nd-Ni/Ni(OH)<sub>2</sub>/NF-500, 3rd-Ni/Ni(OH)<sub>2</sub>/NF-500, and 4th-Ni/Ni(OH)<sub>2</sub>/NF-500 at a scan rate of  $2 \text{ mV s}^{-1}$  in  $1 \text{ M KOH}$  for HER.

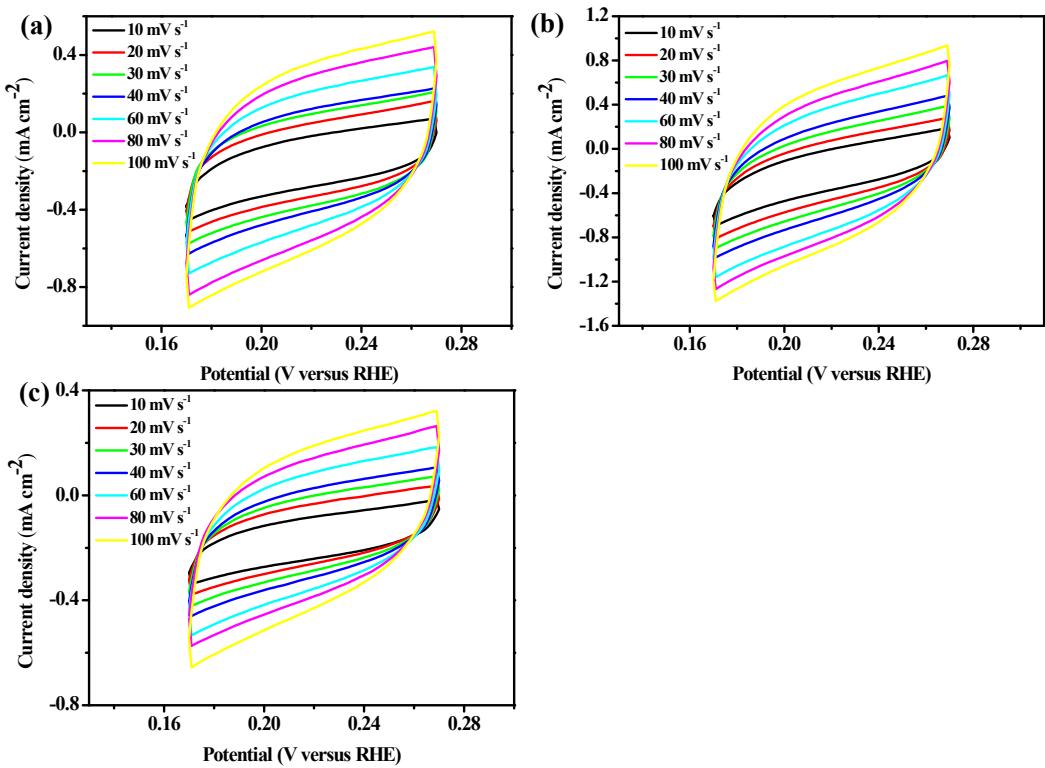


Figure S7. Cyclic voltammograms of (a) 1st-Ni/Ni(OH)<sub>2</sub>/NF, (b) 2nd-Ni/Ni(OH)<sub>2</sub>/NF-500, and (c) c-Ni/Ni(OH)<sub>2</sub>/NF-500.

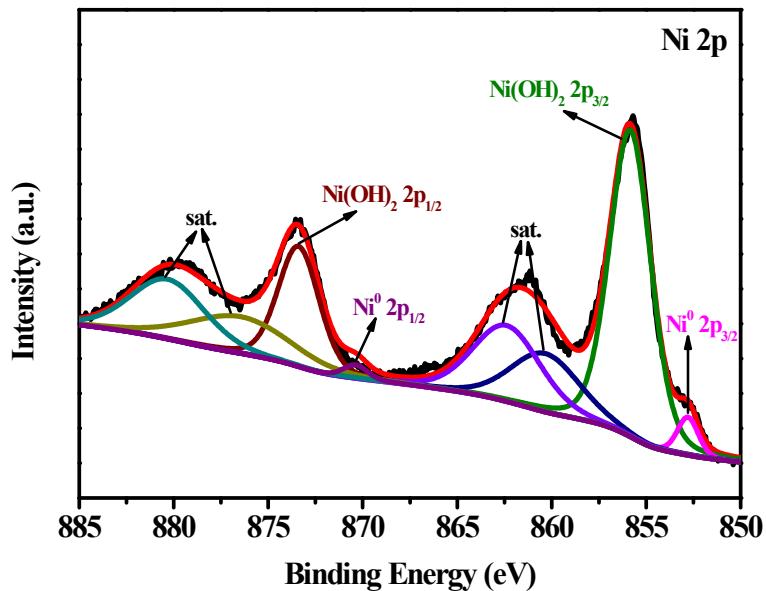


Figure S8. XPS spectrum of Ni 2p for the 2nd-Ni/Ni(OH)<sub>2</sub>/NF-500 after 20 h continuous test.

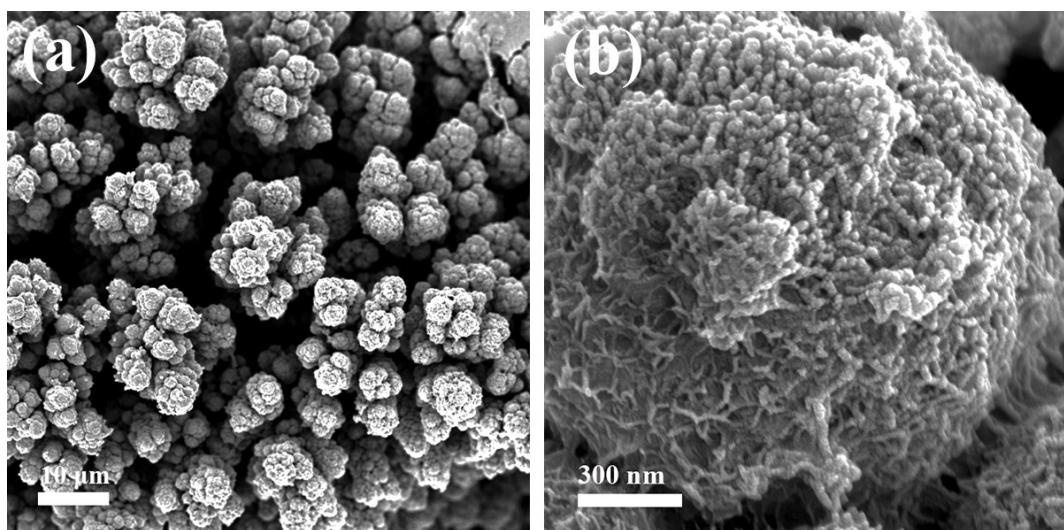


Figure S9. SEM images of 2nd-Ni/Ni(OH)<sub>2</sub>/NF-500 after 20 h continuous test.

Table S1. Comparison of the HER performance of 2nd-Ni/Ni(OH)<sub>2</sub>/NF-500 with the previously reported Ni-based and Ni(OH)<sub>2</sub>-based electrocatalysts.

Catalyst	Overpotential at 10 mA cm <sup>-2</sup> (mV vs RHE)	Electrolyte	Reference
2nd-Ni/Ni(OH) <sub>2</sub> /NF-500	53	1.0 M KOH	This work
Ni(OH) <sub>2</sub> @Ni/CC	68	1.0 M KOH	1
Ni(OH) <sub>2</sub> /NF	127	1.0 M KOH	2
NiO/Ni-CNT	80	1.0 M KOH	3
Ni(OH) <sub>2</sub> -WP/CP	77	1.0 M KOH	4
Ni(OH) <sub>2</sub> -Fe <sub>2</sub> P/TM	76	1.0 M KOH	5
PANI/Ni/NF	72	1.0 M KOH	6

## References

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