## Interfacial Coordination Assembly of Tannic Acid with Metal

## ions on Three-Dimensional Nickel Hydroxide Nanowalls for

## **Efficient Water Splitting**

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Fig. S1. Molecular structure of tannic acid.



Fig. S2. (a, b) SEM images and (c, d) TEM images of  $Ni(OH)_2$  nanosheet array.



Fig. S3. The dominant complexation state of TA-Fe at pH > 7.



Fig. S4. Digital photo of  $Ni(OH)_2/NF$ , TA-Fe@Ni(OH)\_2/NF and TA-Ni@Ni(OH)\_2/NF electrodes.



Fig. S5. (a, b) SEM and (c, d) TEM images of TA-Fe@Ni(OH)<sub>2</sub>/NF.



Fig. S6. XRD patterns.



Fig. S7. FTIR spectra of tannic acid.



Fig. S8. Raman spectrum of TA-Ni@Ni(OH)<sub>2</sub>/NF.



Fig. S9. SEM images of TA-Ni@Ni(OH)<sub>2</sub>/NF.



Fig. S10. SEM images of CoOHF nanowires on nickel foam.



Fig. S11. SEM images of TA-Fe@Co(OH)F hybrid structure.



Fig. S12. TEM images of (a, b) Co(OH)F nanowire and (c, d) TA-Fe complex decorated Co(OH)F nanowires.



Fig. S13 (a) Digital photo of Co(OH)F/NF, TA-Fe@Co(OH)F/NF and TA-Co@Co(OH)F/NF. (b) XRD patterns. (c) Raman spectra and (d) FTIR spectra.

Co(OH)F nanoarrays on nickel foam were synthesized via hydrothermal method and served as the supporting matrix. With the formation of TA-metal complex layer, the color of Co(OH)F electrode changes obviously from purple to slight brown color (Fig. S13a). However, no obvious XRD diffraction peak of TA-metal is observed for the hybrid electrode (Fig. S13b), suggesting the amorphous nature. In the Raman spectra (Fig. S13c), the obvious peaks located at 535 and 599 cm<sup>-1</sup> are ascribed to the bidentate complex of TA with Fe<sup>3+</sup> ions. In addition, the additional peaks at 1711 and 1204 cm<sup>-1</sup> would also be ascribed to the complex layer (Fig. S13d).



Fig. S14. SEM images of TA-Co@Co(OH)F/NF.



Fig. S15. Raman spectrum of TA-Fe/NF.



Fig. S16. SEM images of TA-Fe coating on nickel foam.



Fig. S17. The CV curves of different electrocatalysts at the scan rates from 10 mV s<sup>-1</sup> to 120 mV s<sup>-1</sup>: (a) NF, (b) TA-Fe/NF, (c) Ni(OH)<sub>2</sub>/NF, (d) TA-Fe@Ni(OH)<sub>2</sub>/NF. (e) The linear plots of current density changes versus scan rates.



Fig. S18. Multi-step chronoamperometric curves of TA-Fe@Ni(OH)<sub>2</sub>/NF electrode at different current densities from 10 to 180 mA cm<sup>-2</sup> and an increment of 20 mA cm<sup>-2</sup> per 500 s, then back to 20 mA cm<sup>-2</sup>.



Fig. S19. Tafel plots at various electrodes.

Electroatalyst	Electrolyte	Overpotential	Ref.
TA-Fe@Ni(OH) <sub>2</sub> /NF	1 M KOH	280 mV at 100 mA cm <sup>-2</sup>	This work
TA-Fe@Ni(OH) <sub>2</sub> /NF	1 M KOH	306 mV at 350 mA cm <sup>-2</sup>	This work
TA-Ni@Ni(OH) <sub>2</sub> /NF	1 M KOH	373 mV at 100 mA cm <sup>-2</sup>	This work
Ni(OH) <sub>2</sub> /NF	1 M KOH	380 mV at 100 mA cm <sup>-2</sup>	This work
TA-Fe/NF	1 M KOH	340 mV at 100 mA cm <sup>-2</sup>	This work
Ni <sub>3</sub> Se <sub>2</sub> /NF	1 M KOH	315 mV at 100 mA cm <sup>-2</sup>	1
Ni-Co hydroxide/Ni <sub>2</sub> P <sub>2</sub> O <sub>7</sub>	1 M KOH	357 mV at 100 mA cm <sup>-2</sup>	2
CuFe Oxide/CF	1 M KOH	294 mV at 10 mA cm <sup>-2</sup>	3
nPBA@Co(OH)2/NF	1 M KOH	270 mV at 20 mA cm <sup>-2</sup>	4
Ni-S/MIL-53(Fe)/NF	1 M KOH	298 mV at 100 mA cm <sup>-2</sup>	5
NFN-MOF/NF	1 M KOH	335mV at 250 mA cm <sup>-2</sup>	6
BA-NiFe-LDHs/CP	1 M KOH	293 mV at 100 mA cm <sup>-2</sup>	7
V-Ni <sub>3</sub> S <sub>2</sub> @NiFe LDH/NF	1 M KOH	286 mV at 100 mA cm <sup>-2</sup>	8
FeNiOH/NF	1 M KOH	318 mV at 100 mA cm <sup>-2</sup>	9
CoS <sub>2</sub> /Ni <sub>3</sub> S <sub>2</sub> /CoNiOx	1 M KOH	300 mV at 100 mA cm <sup>-2</sup>	10

Table S1. The OER performance comparison of different catalyst

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