Supplementary information

for

Interface-engineered hybrid electrocatalysts of Ti@holey-TiN/layered-double-hydroxides for efficient seawater electrolysis

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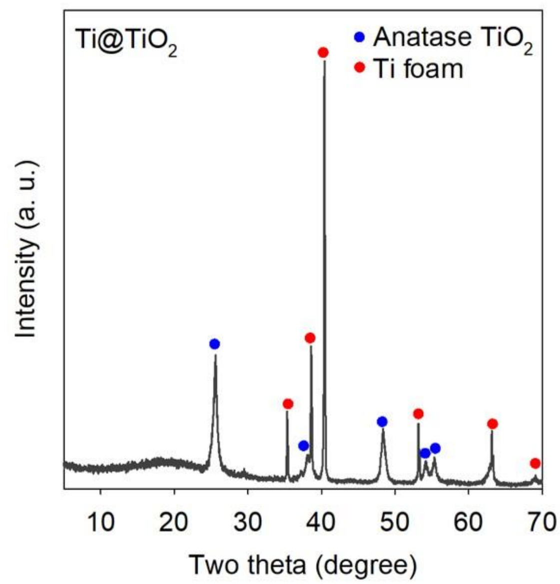

Fig. S1. Powder X-ray diffraction (XRD) patterns of Ti@TiO$_2$ material.

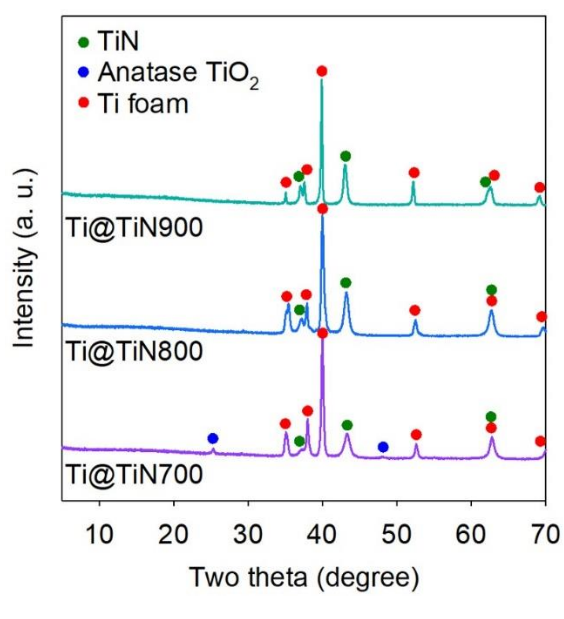

Fig. S2. Powder XRD patterns of Ti@TiN700–900 materials.

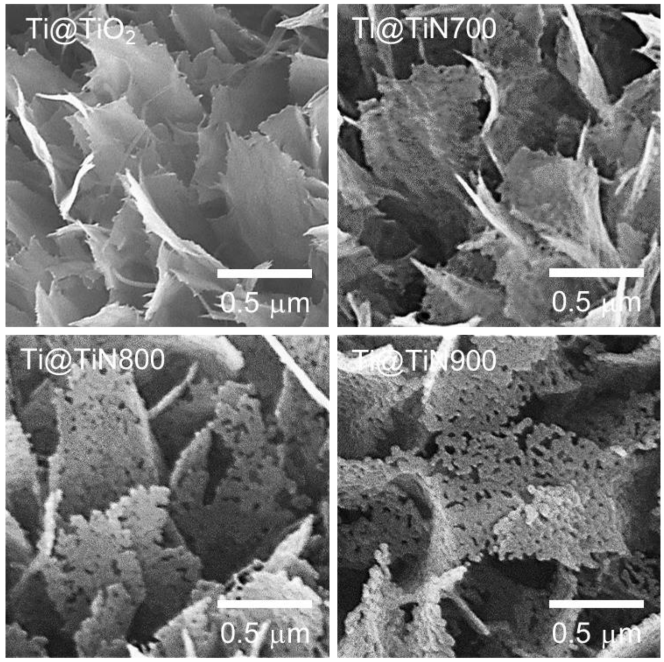

Fig. S3. Field emission-scanning electron microscopy (FE-SEM) images of Ti@TiO_2 and Ti@TiN700–900 materials.

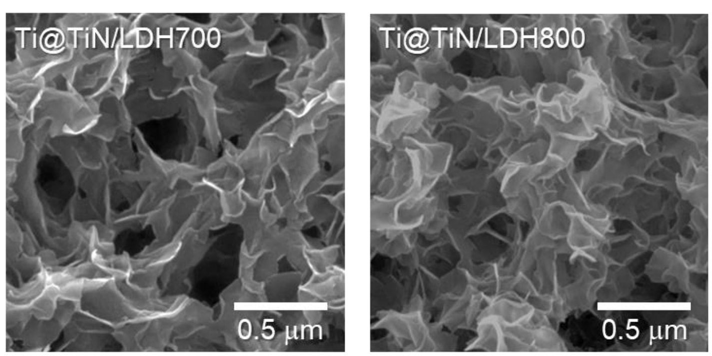

Fig. S4. FE-SEM images of Ti@TiN/LDH700 and Ti@TiN/LDH800 materials.

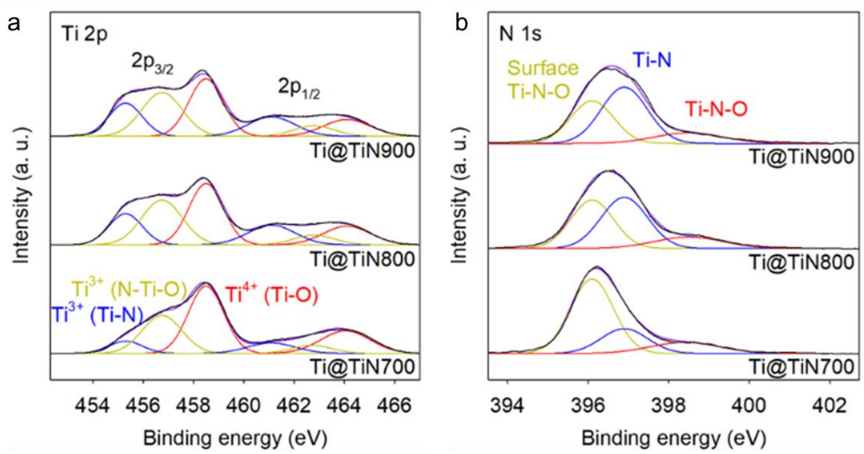

Fig. S5. (a) Ti 2p X-ray photoelectron spectroscopy (XPS) data and (b) N 1s XPS data of Ti@TiN700–900 materials.

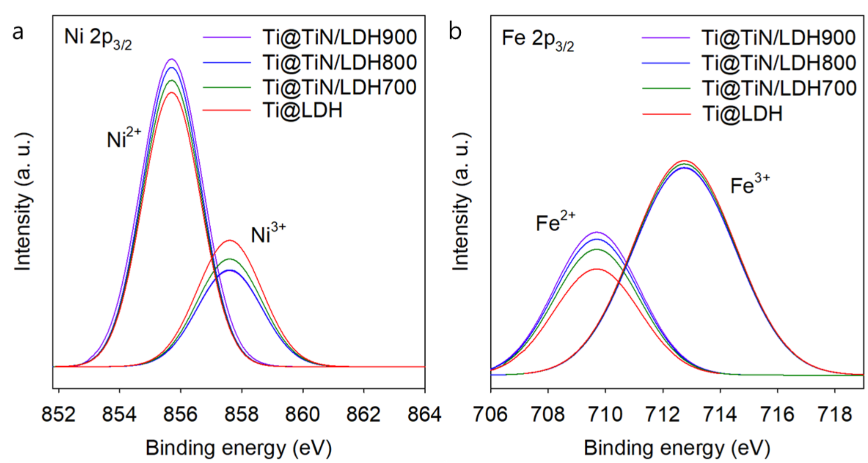

Fig. S6. (a) Overlapped XPS data of deconvoluted Ni 2p peaks and (b) Fe 2p peaks of Ti@LDH and Ti@TiN700–900 materials.

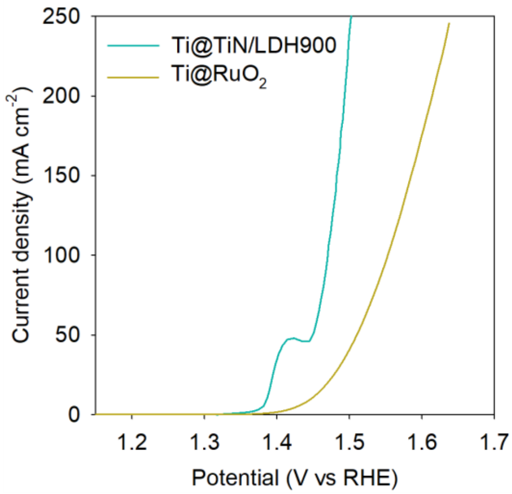

Fig. S7. Linear sweep voltammetry (LSV) data of Ti@TiN/LDH900 and Ti@RuO_2.

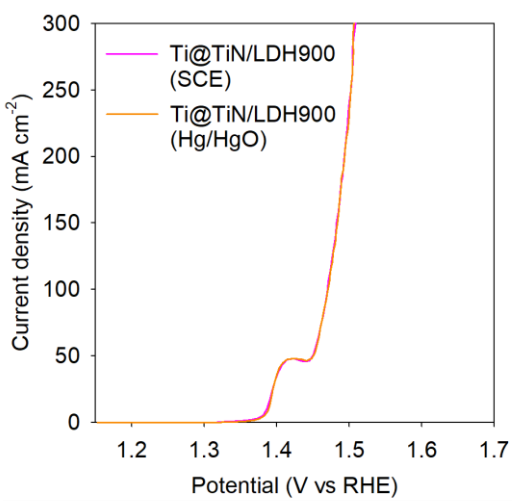

Fig. S8. LSV data of Ti@TiN/LDH900 measured using the SCE and Hg/HgO reference electrodes.

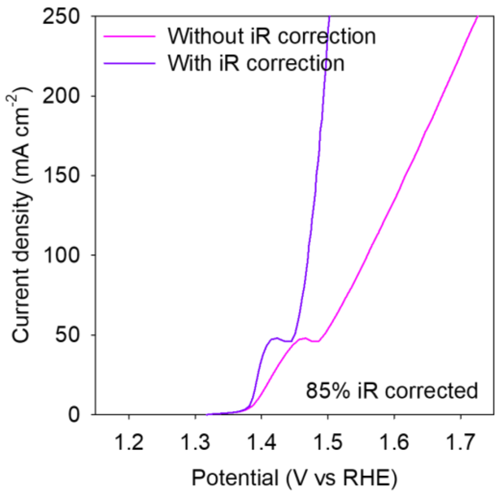

Fig. S9. LSV data of Ti@TiN/LDH900 without and with *iR* correction.

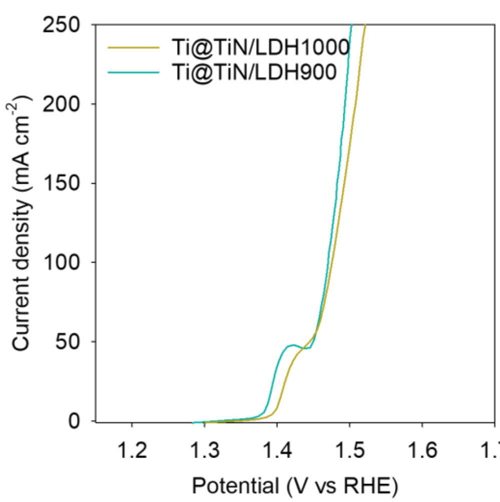

Fig. S10. LSV data of Ti@TiN/LDH900 and Ti@TiN/LDH1000 (synthesis of Ti@TiN/LDH at 1000 °C).

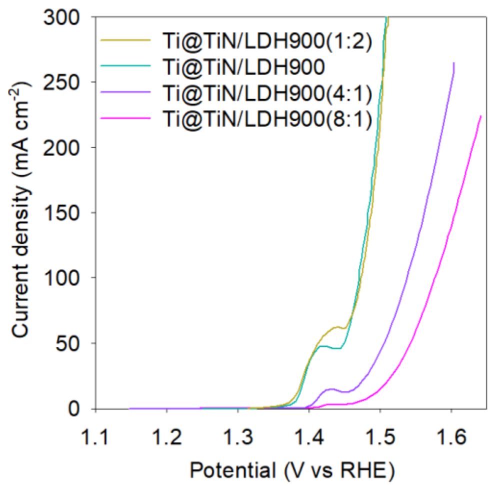

Fig. S11. LSV data of Ti@TiN/LDH900 nanohybrids with different TiN/LDH ratios (x:y).

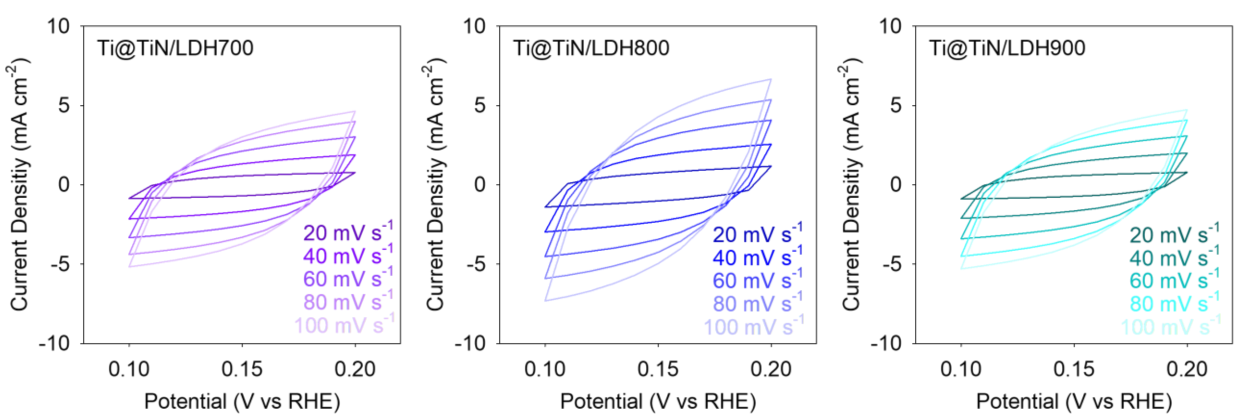

Fig. S12. Cyclic voltammetry (CV) curves of Ti@TiN/LDH700–900 materials measured at various scan rates.

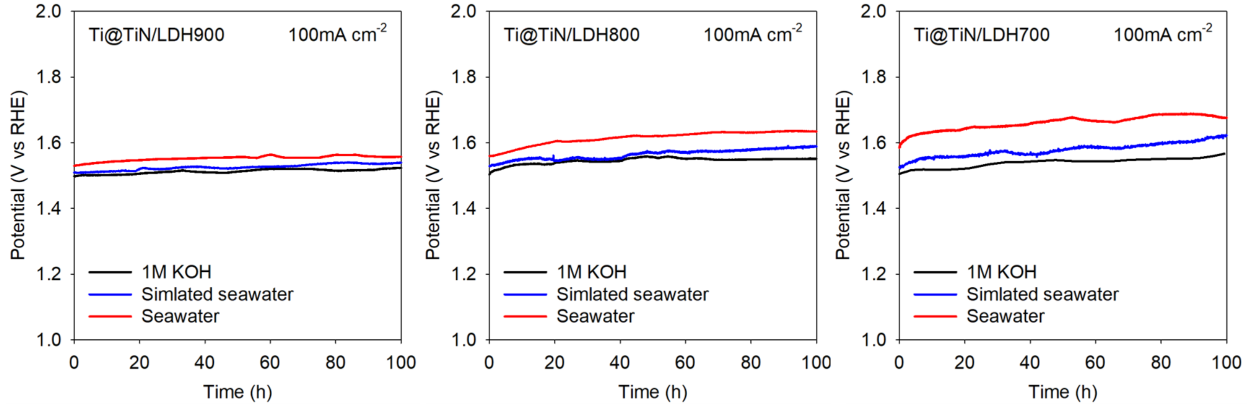

Fig. S13. Durability data of Ti@TiN/LDH900, Ti@TiN/LDH800 and Ti@TiN/LDH700 measured at 1 M KOH, simulated seawater and real seawater electrolytes.

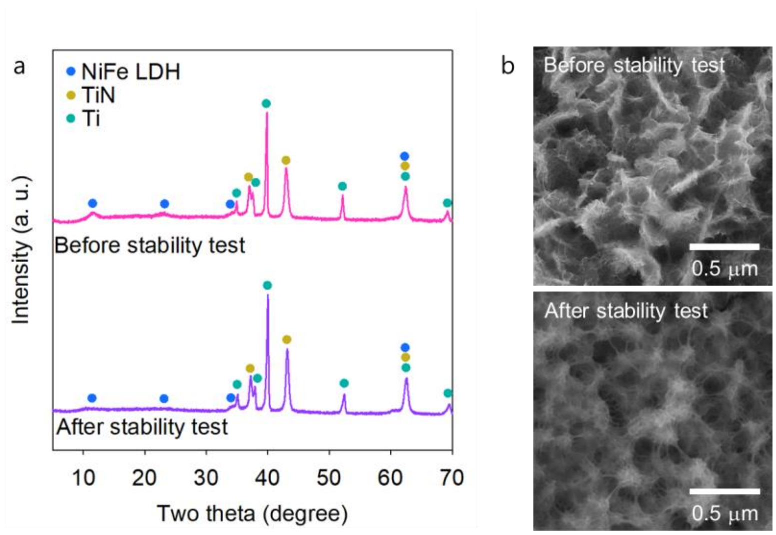

Fig. S14. (a) Powder XRD and (b) FE-SEM data of Ti@TiN/LDH900 before and after stability test.

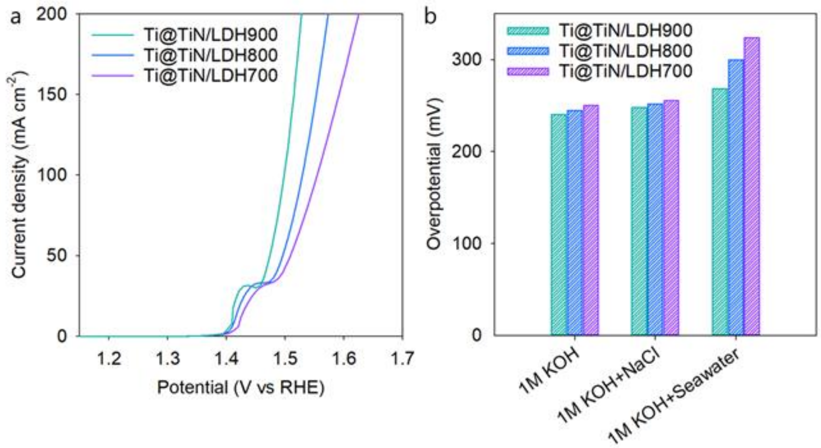

Fig. S15. (a) LSV of OER, and (b) overpotentials of Ti@TiN/LDH700, Ti@TiN/LDH800 and Ti@TiN/LDH900 measured at seawater electrolyte.

Table S1. Energy dispersive spectroscopy (EDS) data of Ti@TiN/LDH900 nanohybrids with different TiN/LDH ratios.

Material	molar ratio (Ni–Fe-LDH/TiN)
Ti@TiN/LDH900	2.03/1

Table S2. Relative concentrations of Ni^{2+}/Ni^{3+} and Fe^{2+}/Fe^{3+} in Ti@TiN/LDH materials.

Material	Ni^{2+}/Ni^{3+}	Fe^{2+}/Fe^{3+}
Ti@LDH	2.442	0.569
Ti@TiN/LDH700	2.720	0.613
Ti@TiN/LDH800	2.954	0.665
Ti@TiN/LDH900	3.066	0.681

Table S3. Results of non-linear least-squares Ni/Fe K-edge EXAFS fitting analysis.

Material	Bonding pair	Coordination number	Bond distance (Å)	ΔE (eV)	σ^2 (Å^2)
Ti@TiN/LDH700	Ni–O	6	2.038	2.51	0.0039
	Fe–O	6	1.973	0.94	0.0053
Ti@TiN/LDH800	Ni–O	6	2.039	2.43	0.0040
	Fe–O	6	1.975	0.10	0.0043
Ti@TiN/LDH900	Ni–O	6	2.040	1.77	0.0044
	Fe–O	6	1.984	1.03	0.0083
Ni–Fe-LDH	Ni–O	6	2.023	–3.64	0.0058
	Fe–O	6	1.968	–5.14	0.0079

Table S4. Comparison of OER activity for Ti@TiN/LDH in 1 M KOH and real seawater electrolytes with recently reported OER electrocatalysts.

Catalyst	Support	Electrolyte	Overpotential @ 100 mV @ mA cm^{-2}	Reference
Ti@TiN/NiFe LDH	Ti foam	1 M KOH 1 M KOH + Seawater	240 268	This Work
NiFe LDH@ $FeNi_2S_4$	Ni foam	1 M KOH 1 M KOH + Seawater	240 271	*Adv. Funct. Mater.*, 2022, **32**, 2200951
N-CDs/NiFe LDH	Ni foam	1 M KOH 1 M KOH + Seawater	260 340	*Nano Res.*, 2022, **15**, 7063–7070
BZ-NiFe LDH	Carbon cloth	1 M KOH 1 M KOH + Seawater	230 300	*Nano Res. Energy*, 2022, **1**, 9120028
CoP_x@FeOOH	Ni foam	1 M KOH 1 M KOH + Seawater	254 283	*Appl. Catal., B*, 2021, **294**, 120256
NiCoS	Ni foam	1 M KOH 1 M KOH + Seawater	270 360	*Appl. Catal., B*, 2021, **291**, 120071
Mo–Ni_3S_2	Ni foam	1 M KOH 1 M KOH + Seawater	280 291	*Energy Fuels*, 2022, **36**, 2910–2917
Ni_3S_2@Fe-NiP_x	Ni foam	1 M KOH 1 M KOH + Seawater	240 290	*Adv. Sci.*, 2022, **9**, 2104846
Ni_2P-Fe_2P	Ni foam	1 M KOH 1 M KOH + Seawater	261 305	*Adv. Funct. Mater.*, 2021, **31**, 2006484
S,P-(Ni,Mo,Fe) NiMoP	Wood aerogel	1 M KOH 1 M KOH + Seawater	279 286	*Appl. Catal., B*, 2021, **293**, 120215
1D-Cu@Co-CoO/Rh	Cu foam	1 M KOH 1 M KOH + Seawater	380 400	*Small*, 2021, **17**, 2103826

Table S5. Charge transfer resistance (R_{ct}) values calculated from electrochemical impedance spectroscopy (EIS) fitting analysis.

Material	R_{ct} (Ω)
Ti@TiN	36.7
Ti@LDH	152.6
Ti@TiN/LDH700	2.5
Ti@TiN/LDH800	2.3
Ti@TiN/LDH900	1.5