Electronic Supplementary Material (ESI) for Journal of Materials Chemistry A.

Hierarchical Ni/$NiTiO_3$ derived from NiTi LDHs: a bifunctional electrocatalyst for overall water splitting †

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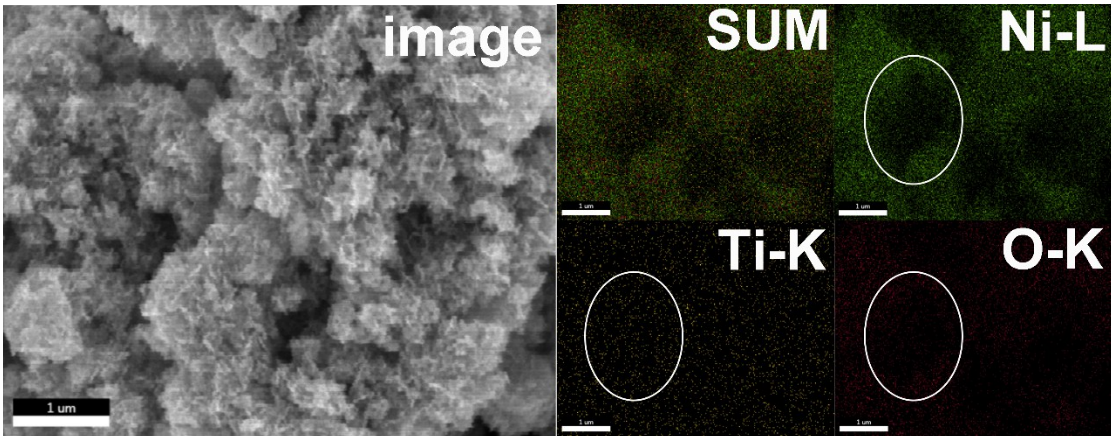


Figure S1. SEM-EDX elemental mapping of NT-15.

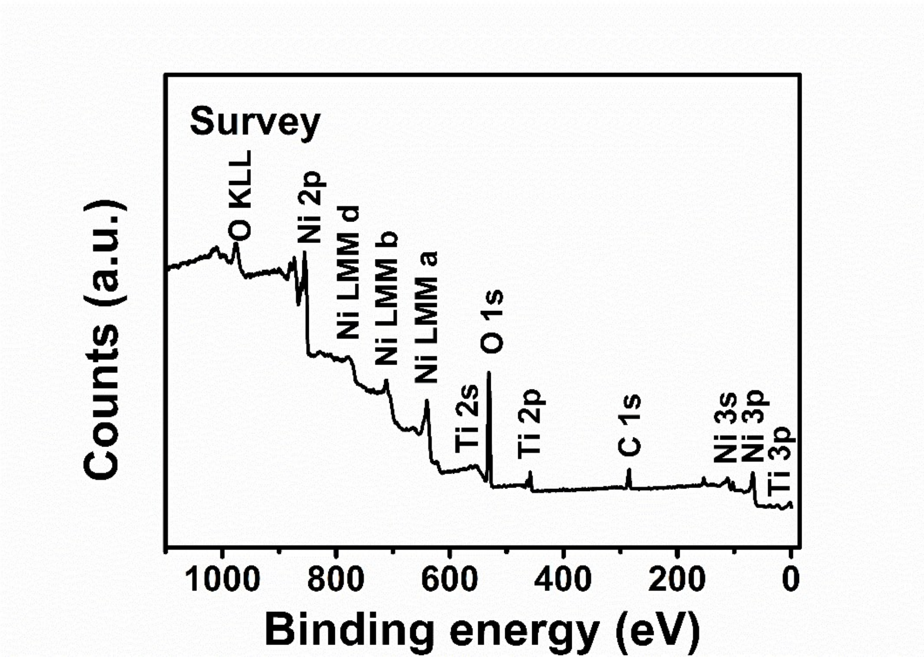


Figure S2. XPS survey of NT-15.

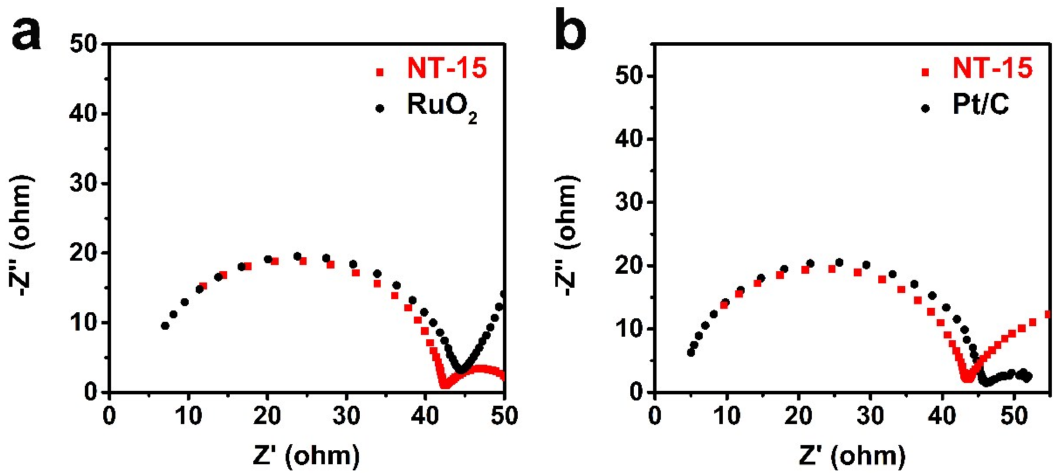


Figure S3. (a) Nyquist plots of NT-15 and RuO_2; **(b)** Nyquist plots of NT-15 and Pt/C.

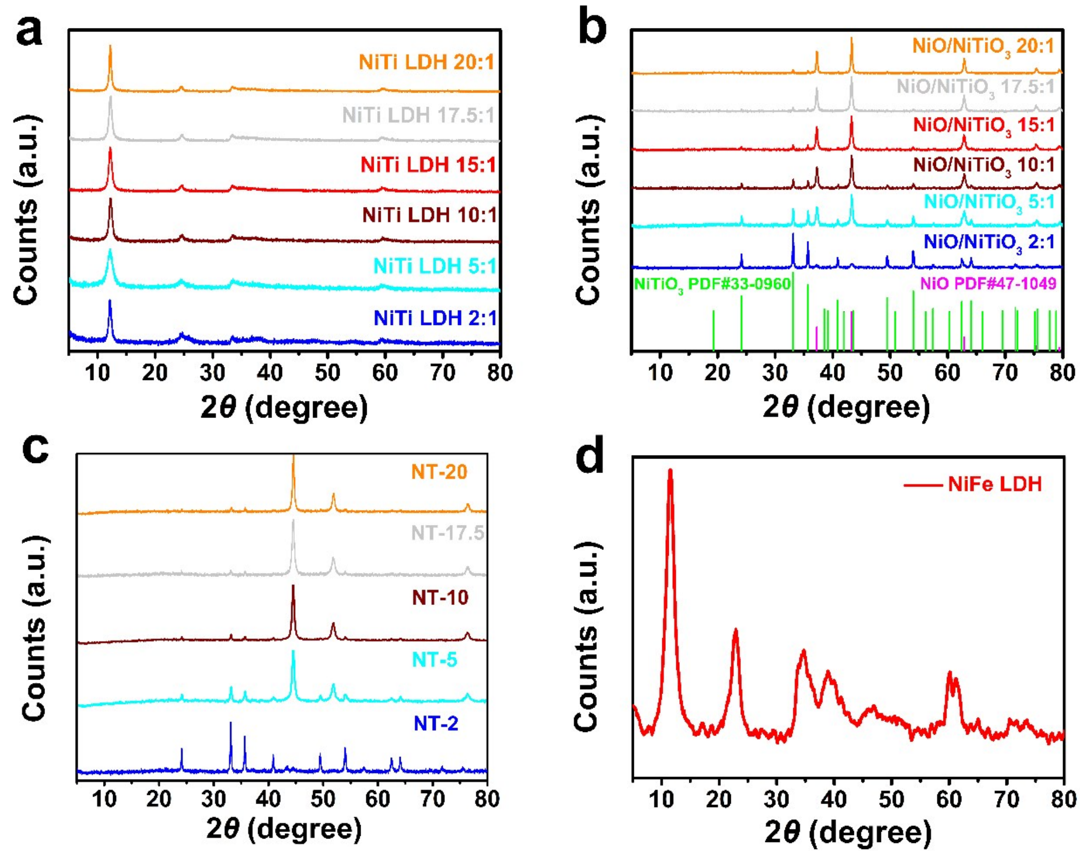
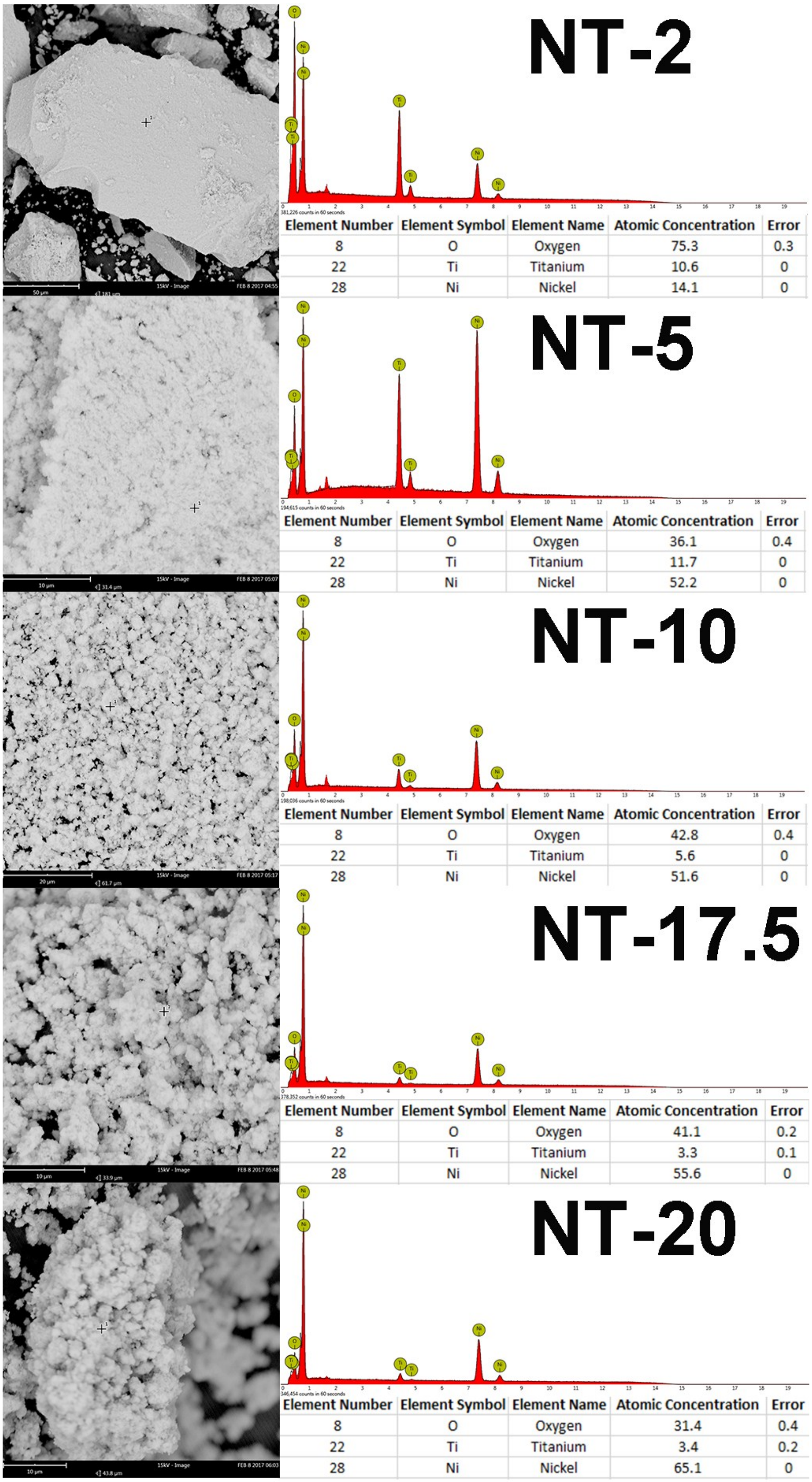


Figure S4. (a, b&c) The XRD patterns Ni/$NiTiO_3$ and their precursors with different ratios of Ni to Ti; **(d)** the XRD pattern of reference catalyst (NiFe LDH).



Element Number	Element Symbol	Element Name	Atomic Concentration	Error
8	O	Oxygen	75.3	0.3
22	Ti	Titanium	10.6	0
28	Ni	Nickel	14.1	0

Element Number	Element Symbol	Element Name	Atomic Concentration	Error
8	O	Oxygen	36.1	0.4
22	Ti	Titanium	11.7	0
28	Ni	Nickel	52.2	0

Element Number	Element Symbol	Element Name	Atomic Concentration	Error
8	O	Oxygen	42.8	0.4
22	Ti	Titanium	5.6	0
28	Ni	Nickel	51.6	0

Element Number	Element Symbol	Element Name	Atomic Concentration	Error
8	O	Oxygen	41.1	0.2
22	Ti	Titanium	3.3	0.1
28	Ni	Nickel	55.6	0

Element Number	Element Symbol	Element Name	Atomic Concentration	Error
8	O	Oxygen	31.4	0.4
22	Ti	Titanium	3.4	0.2
28	Ni	Nickel	65.1	0

Figure S5. The SEM-EDX patterns of Ni/$NiTiO_3$ with different ratios of Ni to Ti.

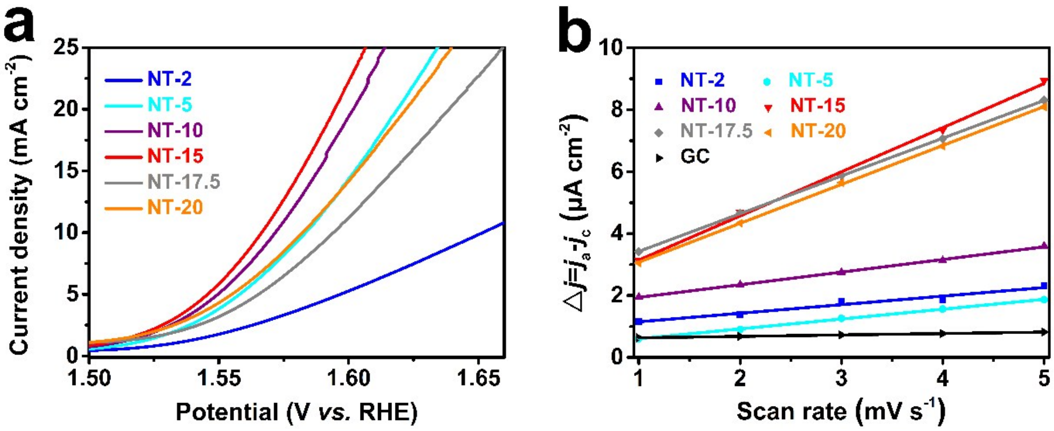


Figure S6. (a) Polarization curves of Ni/$NiTiO_3$ with different ratios of Ni to Ti collected at 5 mV s^{-1} and 2,000 rpm in O_2-saturated 0.1 M KOH; **(b)** differences in current density ($\Delta j = j_a - j_c$) plotted against scan rates. The linear slope is equivalent to twice of C_{dl}.

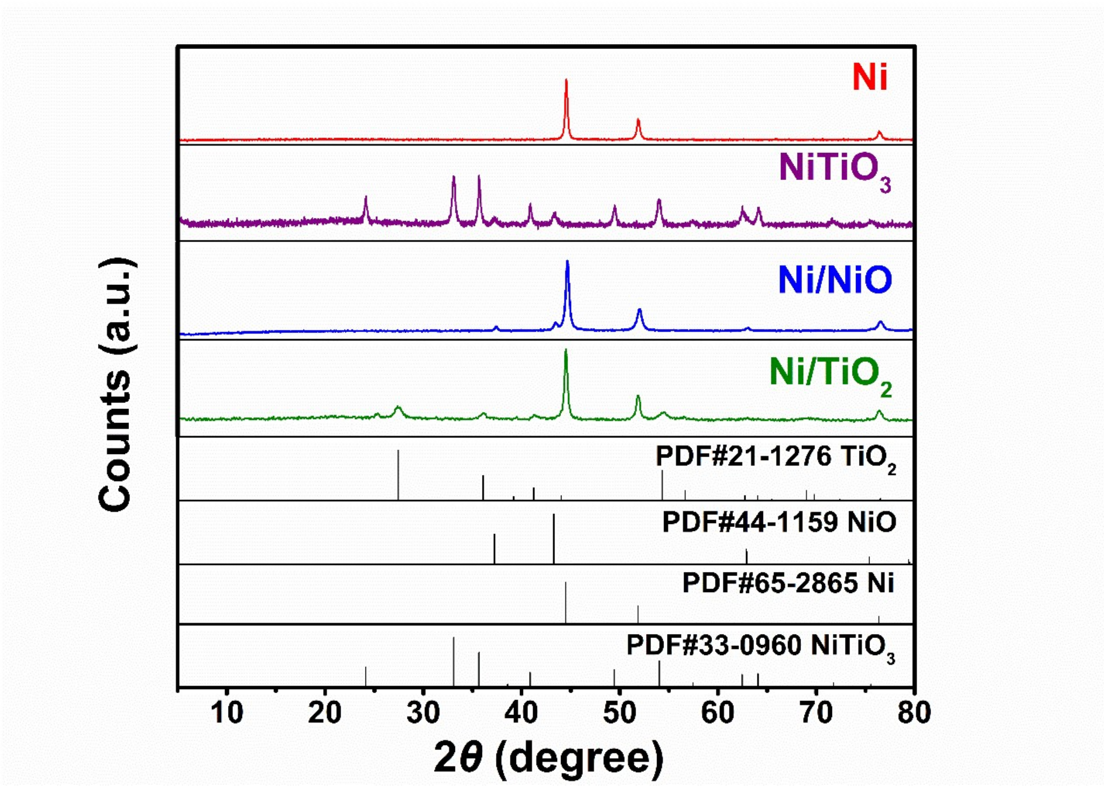


Figure S7. XRD patterns of bare Ni, bare $NiTiO_3$, Ni/NiO and Ni/TiO_2.

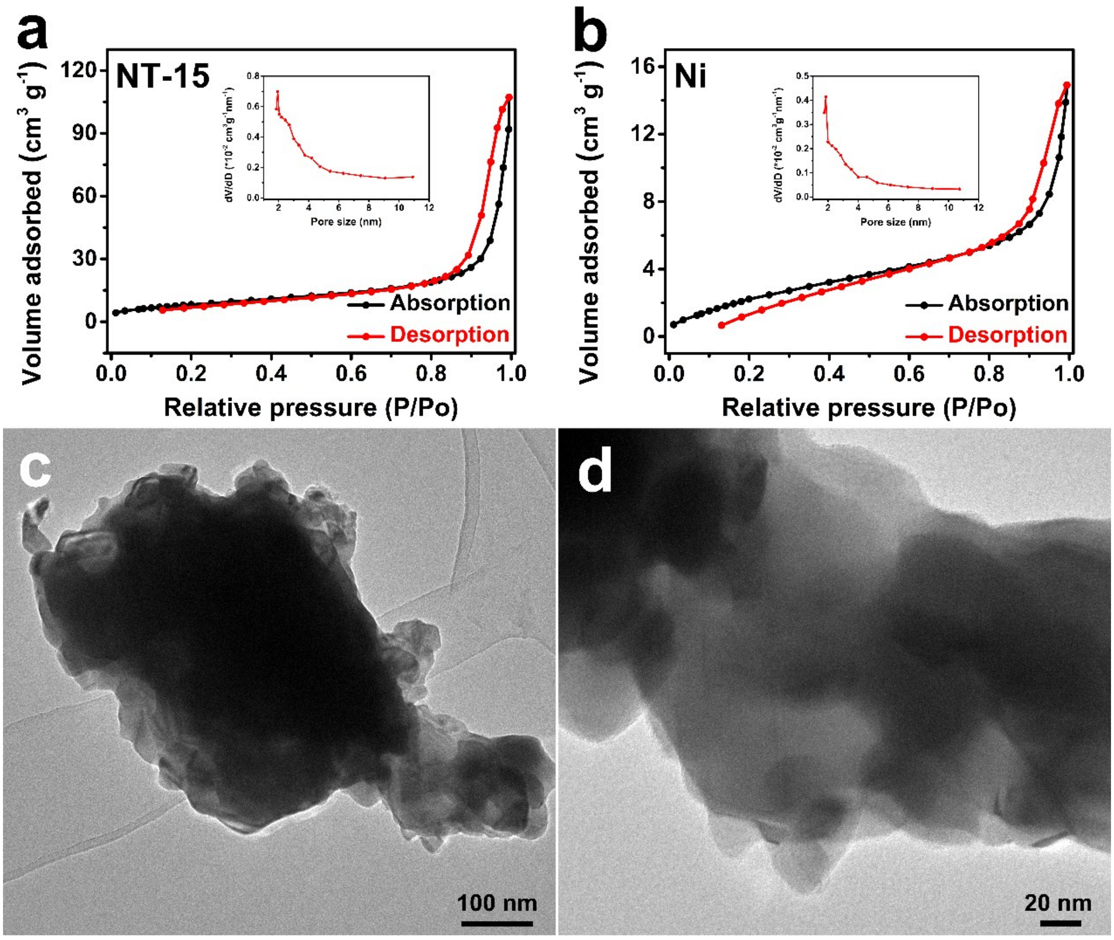


Figure S8. N_2 sorption isotherm of **(a)** NT-15 and **(b)** Ni and pore distribution in the inset; **(c&d)** TEM image of bare aggregated Ni nanoparticles.

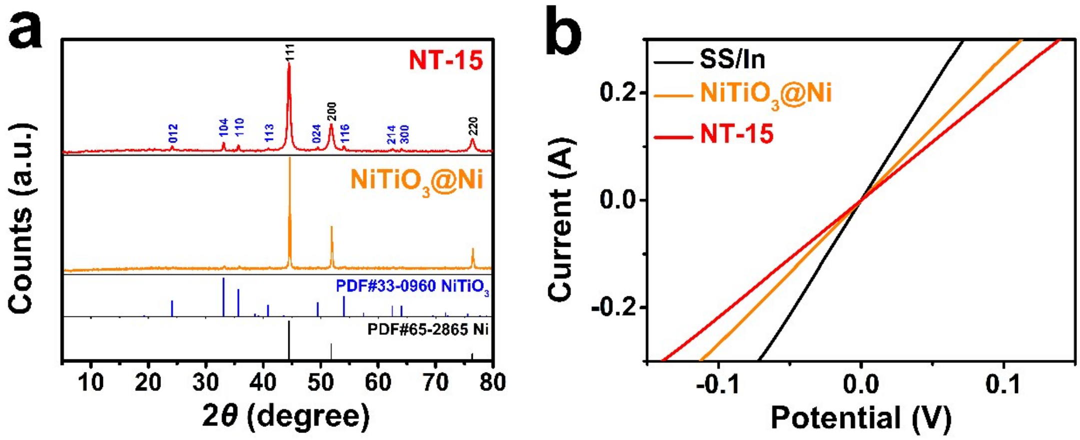


Figure S9. (a) XRD patterns and **(b)** I-V curves of $NiTiO_3$@Ni and NT-15.

Detail calculations: The resistance of SS/In is 0.2367 Ω. The resistances deducting SS/In of $NiTiO_3$@Ni and NT-15 are 0.3724 and 0.4607 Ω, respectively. *k* (conductivity) = GL/A, where G (conductance) = 1/R ($NiTiO_3$@Ni: 7.369 S and NT-15: 4.464 S), L is length ($NiTiO_3$@Ni: 0.66 mm and NT-15: 0.67 mm) and A is area ($2.826*10^{-5}$ m^2).

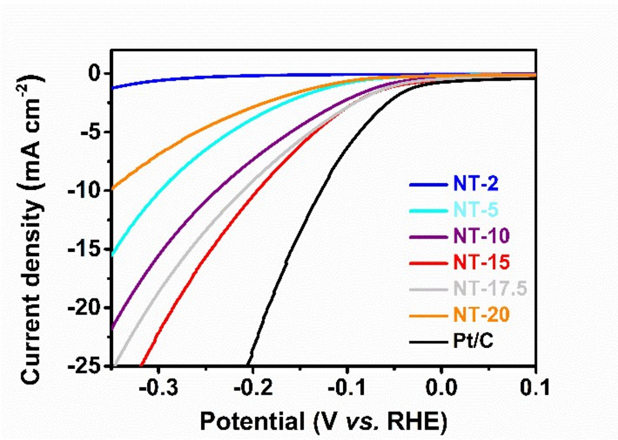


Figure S10. Polarization curves of Ni/$NiTiO_3$ with different ratios of Ni to Ti collected at 5 mV s^{-1} and 2,000 rpm in N_2-saturated 0.1 M KOH;

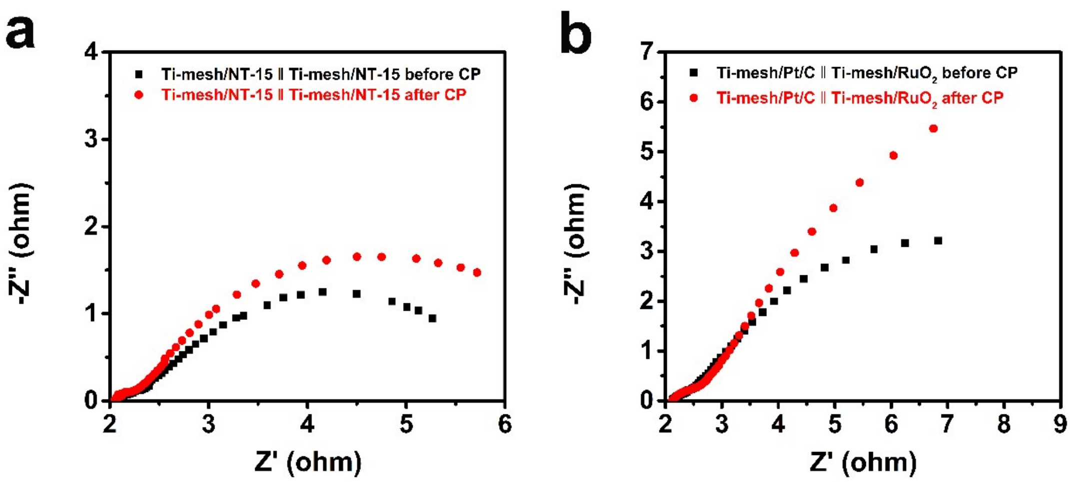


Figure S11. (a) Nyquist plots of Ti-mesh/NT-15 || Ti-mesh/NT-15; **(b)** Nyquist plots of Ti-mesh/Pt/C || Ti-mesh/RuO_2.

Table S1. Comparison with some reported Ni-based OER catalysts.

Catalyst	Onset overpotential (mV)	Overpotential (mV)@ 10 mA/cm^2	Tafel slope (mV dec^{-1})	Electrolyte	reference
Ni/NiTiO$_3$	270	336	62.2	0.1 M KOH	This work
NiCo$_{2.7}$(OH)$_x$	250	350	65	1 M KOH	1
m-NiFe/CN$_x$	~220	360	59.1	0.1 M KOH	2
α-Ni(OH)$_2$	310	331	42	0.1 M KOH	3
Ni$_2$P NPs	220	290	47	1 M KOH	4
NiS nanosheet	270	~290	47	0.1 M KOH	5
NiSe nanowalls/G	370	430	83.4	1 M KOH	6
NiCo$_2$S$_4$ /Ni foam	270	340	—	0.1 M KOH	7

Table S2. Comparison with some reported Ni-based HER catalysts.

Catalyst	Onset overpotential (mV)	Overpotential (mV)@ 10 mA/cm^2	Tafel slope (mV dec^{-1})	Electrolyte	reference
Ni/NiTiO$_3$	50	196	118	0.1 M KOH	This work
Ni$_2$P/CNT	88	124	53	0.5 M H_2SO_4	8
NiSe/NF	~25	96	120	1 M KOH	9
Ni-C-N NS	34.7	60.9	32	0.5 M H_2SO_4	10
Ni/NiO/Ni foam	~0	145	43	1 M KOH	11
TiN@Ni$_3$N	15	—	42.1	1 M KOH	12
NiS$_2$ NA/CC	70	149	69	1.0 M KOH	13

Table S3. Comparison with some reported bifunctional electrocatalysts.

Catalyst	Onset potential (V)	Overpotential (V) @10 mA/cm^2	Electrolyte	OER onset potential (V)	HER onset potential (V)	Electrolyte	reference
Ni/NiTiO$_3$	1.55	1.63	1 M KOH	1.50	–0.05	0.1 M KOH	This work
CoP/Cu foil	~1.56	1.65	1 M KOH	1.52	–0.05	1 M KOH	14
Co$_9$S$_8$/WS$_2$/Ti plate	1.50	1.65	1 M KOH	1.40	–0.90	1 M KOH	15
NiSe/Ni foam	1.50	1.63	1 M KOH	—	~–0.025	1 M KOH	9
TiN@Ni$_3$N/Ti foil	1.57	1.67	1 M KOH	1.52	~–0.015	1 M KOH	12

Table S4. The ICP-AES data of NT-15 in the electrolyte before and after long-term working measurement.

Sample ID	Line	Mean	Units	RSD
OER before long-term working	Ni 231.604	< 0.0000	ug/mL	0.8698
OER before long-term working	Ni 221.648	< 0.0000	ug/mL	0.5181
OER after long-term working-1	Ni 231.604	< 0.0000	ug/mL	0.8742
OER after long-term working-1	Ni 221.648	< 0.0000	ug/mL	0.5716
OER after long-term working-2	Ni 231.604	< 0.0000	ug/mL	0.9321
OER after long-term working-2	Ni 221.648	< 0.0000	ug/mL	0.1648
HER before long-term working	Ni 231.604	< 0.0000	ug/mL	1.4593
HER before long-term working	Ni 221.648	< 0.0000	ug/mL	0.1792
HER after long-term working-1	Ni 231.604	< 0.0000	ug/mL	2.3155
HER after long-term working-1	Ni 221.648	< 0.0000	ug/mL	3.263
HER after long-term working-2	Ni 231.604	< 0.0000	ug/mL	0.482
HER after long-term working-2	Ni 221.648	< 0.0000	ug/mL	2.6068

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