Supplementary material

Surface-engineered Ni-Pt Alloys as Robust and Cost-effective Electrocatalysts for High-Performance Proton Exchange Membrane Water Electrolysis

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Table S1. Electrodeposition conditions of the catalysts.

Bulk atomic composition (EDX)	Electrolyte configuration / mM					Deposition potential / V_{SCE}	Deposition time / min
	$NiCl_2$	H_2PtCl_6	H_3BO_3	NH_4Cl	HCl		
Ni_{100}	50	0					
$Ni_{99.8}Pt_{0.2}$	50	0.25					
$Ni_{99.5}Pt_{0.5}$	50	0.50	300	100	10	-3.0	10
$Ni_{98.7}Pt_{1.3}$ (P-$Ni_{98.7}Pt_{1.3}$)	50	1.25					
Pt_{100}	0	1.25					

Table S2. Binding energies for Ni $2p_{3/2}$ and Pt 4f XPS spectra for Ni_{100}, Pt_{100}, P-$Ni_{98.7}Pt_{1.3}$, and HT-$Ni_{98.7}Pt_{1.3}$.

Sample	Ni $2p_{3/2}$	Peak position / eV	Area ratio / %	Pt 4f	Peak position / eV	Area ratio / %	Atomic ratio of Ni : Pt
Ni_{100}	Ni metal	852.15	24.66		-		100 : 0
	NiO	853.27	7.87				
	$Ni(OH)_2$	855.40	67.47				
Pt_{100}		-		Pt	71.54 74.87	80.05	0 : 100
				PtO	72.39 75.71	12.41	
				PtO_2	74.36 77.57	7.54	
P-$Ni_{98.7}Pt_{1.3}$	Ni metal	852.26	19.86	Pt	71.15 74.50	100	75.07 : 24:93
	NiO	853.25	12.01				
	$Ni(OH)_2$	855.51	68.12				
HT-$Ni_{98.7}Pt_{1.3}$	Ni metal	852.31	30.15	Pt	71.09 74.44	100	49.21 : 50.79
	NiO	853.29	13.74				
	$Ni(OH)_2$	855.61	56.11				

Table S3. Comparison of PEMWE performance with Pt-based cathodes and IrO_2 anode.

Membrane electrode assembly (MEA)			Temperature / °C	Current density / A cm^{-2}		Cell voltage / V_{cell}		Reference
Cathode (loading amount)	Membrane	Anode (loading amount)		@ 1.7 V	@ 2.0 V	@ 1 A cm^{-2}	@ 2 A cm^{-2}	
HT-$Ni_{98.7}Pt_{1.3}$ (0.029 mg cm^{-2})	**Nafion 212**	**Com. IrO_x (2.0 mg_{Ir} cm^{-2})**	**90**	**0.783**	**2.731**	**1.748**	**1.914**	**This work**
Pt/C (0.1 mg_{Pt} cm^{-2})	**Nafion 212**	**Com. IrO_x (2.0 mg_{Ir} cm^{-2})**	**90**	**0.916**	**2.633**	**1.714**	**1.883**	**This work**
Pt/C (0.2 mg_{Pt} cm^{-2})	Nafion 212	Com. IrO_x (2.5 mg_{Ir} cm^{-2})	80	1.024	2.267	1.698	2.311	Journal of Power Sources 501 (2021) 230002
PED Pt/CP (0.051 mg_{Pt} cm^{-2})	Nafion 212	ED IrO_2/CP (0.1 mg cm^{-2})	90	0.705	2.031	1.792	1.994	Appl. Surf. Sci., 444 (2018) 303-311
Pt/C (0.04 mg_{Pt} cm^{-2})	Nafion 212	IrO_2-CM (0.5 mg_{Ir} cm^{-2})	80	0.758	1.593	1.806	1.605	Material Today Energy 32 (2023) 101237
$PdPt_{ML}$ (0.5 mg_{Pt} cm^{-2})	Nafion 212	Com. IrO_x (2.0mg_{Ir} cm^{-2})	80	0.330	1.340	1.894	2.163	Small, 20 (2024) 2406935
HEA-QDs/C (0.2 mg_{Pt} cm^{-2})	Nafion 212	Com. IrO_x (1.5 mg_{Ir} cm^{-2})	65	0.959	-	1.701	1.865	Energy Environ. Sci., 17 (2024), 6594

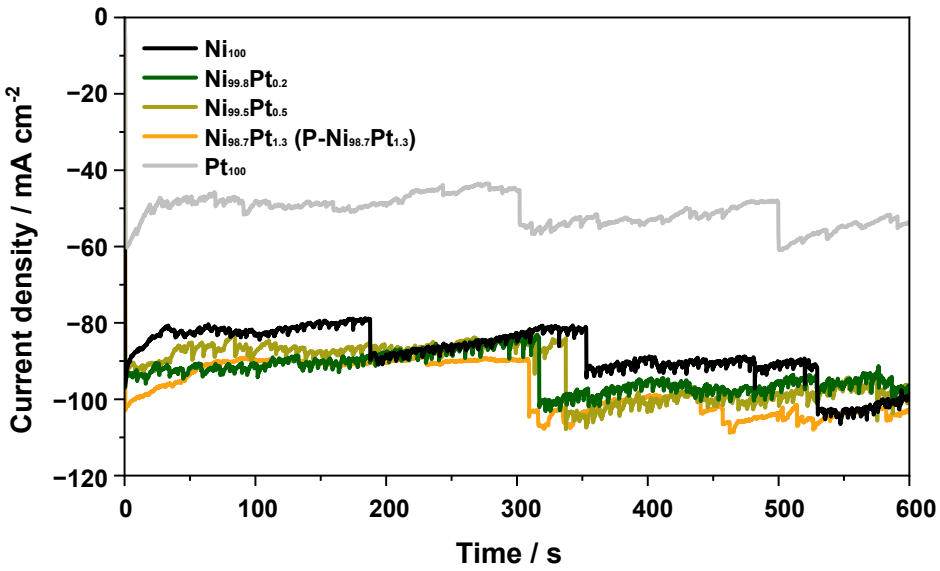

Figure S1. Potentiostatic electrodeposition i-t curve.

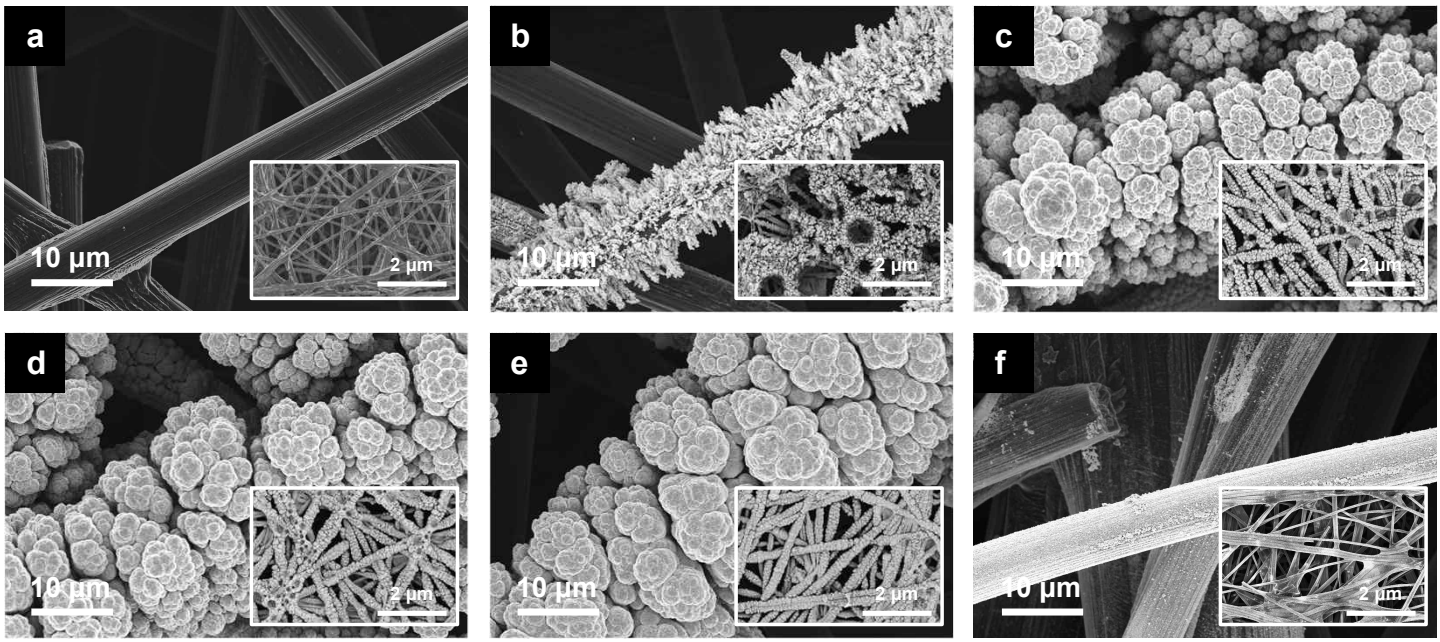

Figure S2. FE-SEM images of (a) bare carbon paper, (b) Ni_{100}, (c) $Ni_{99.7}Pt_{0.3}$, (d) $Ni_{98.9}Pt_{0.5}$, (e) $Ni_{98.7}Pt_{1.3}$(P-$Ni_{98.7}Pt_{1.3}$), and (f) Pt_{100} samples electrodeposited on carbon paper without heat-treatment. The insets show the FE-SEM images at lower magnification.

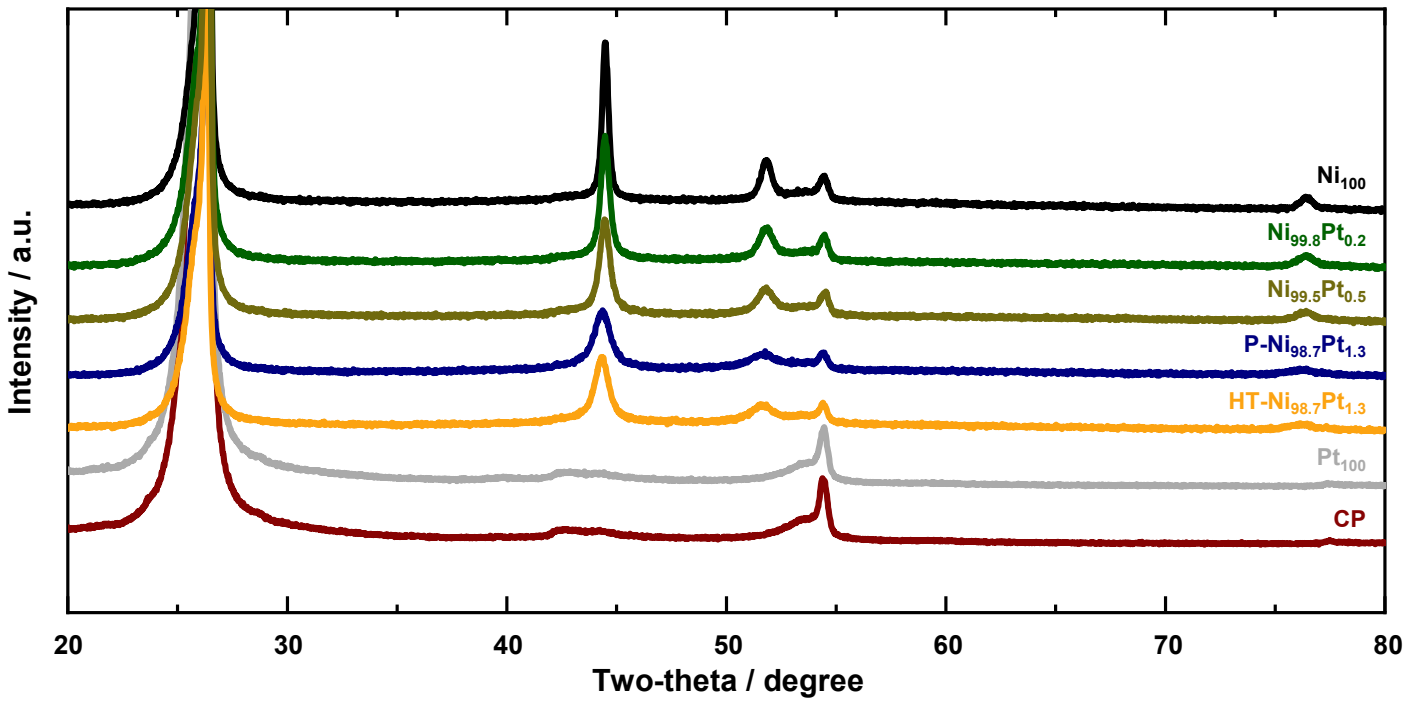

Figure S3. XRD patterns in the two-theta range of 20-80°.

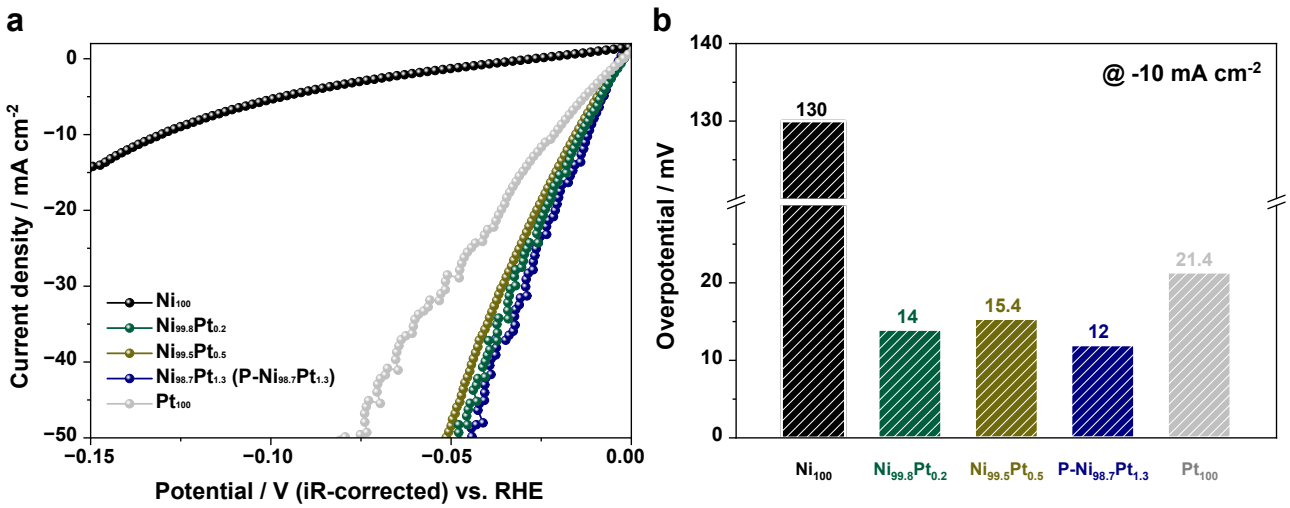

Figure S4. (a) HER polarization curves of Ni_{100}, $Ni_{99.8}Pt_{0.2}$, $Ni_{99.5}Pt_{0.5}$, $Ni_{98.7}Pt_{1.3}$ (P-$Ni_{98.7}Pt_{1.3}$), and Pt_{100} samples. (b) Overpotential at -10 mA cm^{-2} current density.

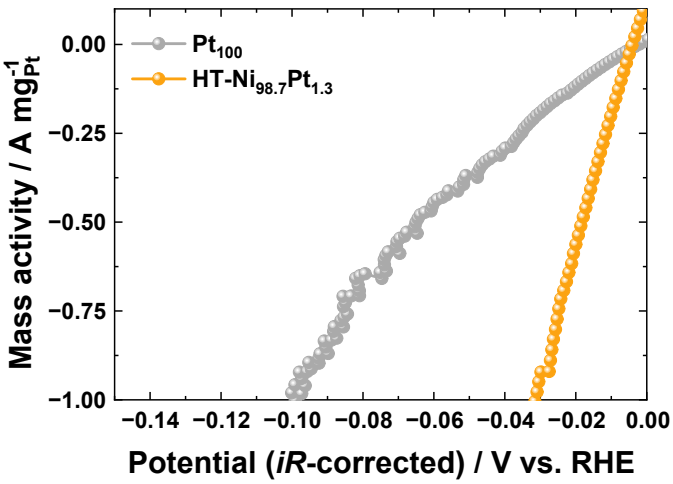

Figure S5. Mass activity normalized by Pt loading amounts.

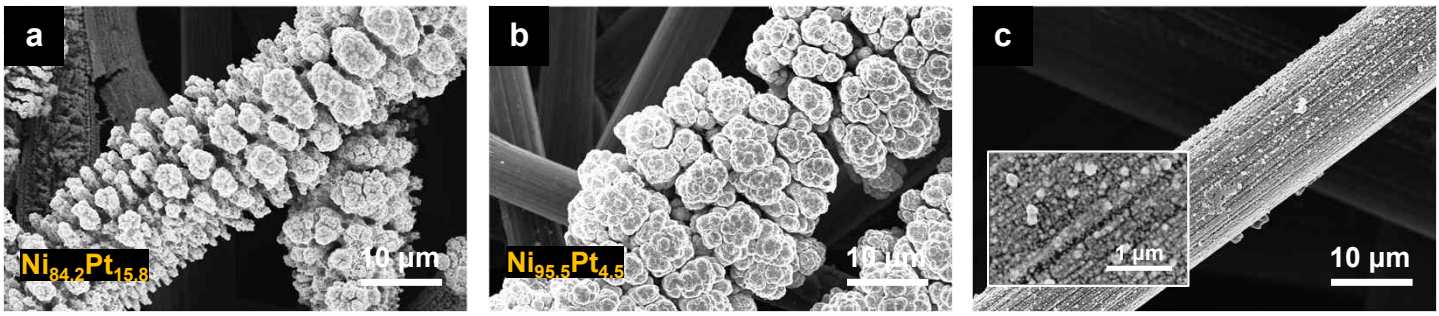

Figure S6. FE-SEM images after ADT 5000 cycles of (a) P-$Ni_{98.7}Pt_{1.3}$, (b) HT-$Ni_{98.7}Pt_{1.3}$, and (c) Pt_{100}. The inset in c shows the FE-SEM image at higher magnification.

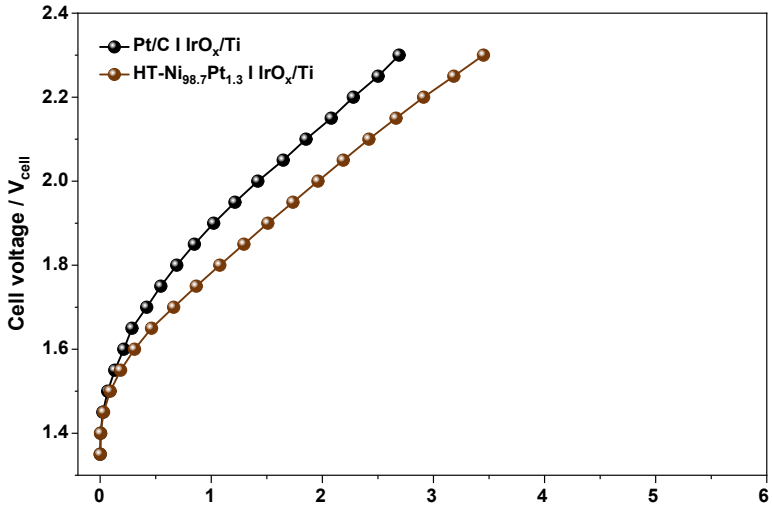

Figure S7. Single-cell polarization curves using HT-$Ni_{98.7}Pt_{1.3}$ (0.029 mg of Pt per cm^2)/CP cathode and IrO_x/Ti anode. A commercial Pt/C cathode(0.1 mg of Pt per cm^2) based PEMWE performance is compared.