

## Supporting Information

### Extremely efficient and stable hydrogen evolution by a Pt/NiO<sub>x</sub> composite film deposited on a nickel foam using a mixed metal-imidazole casting method

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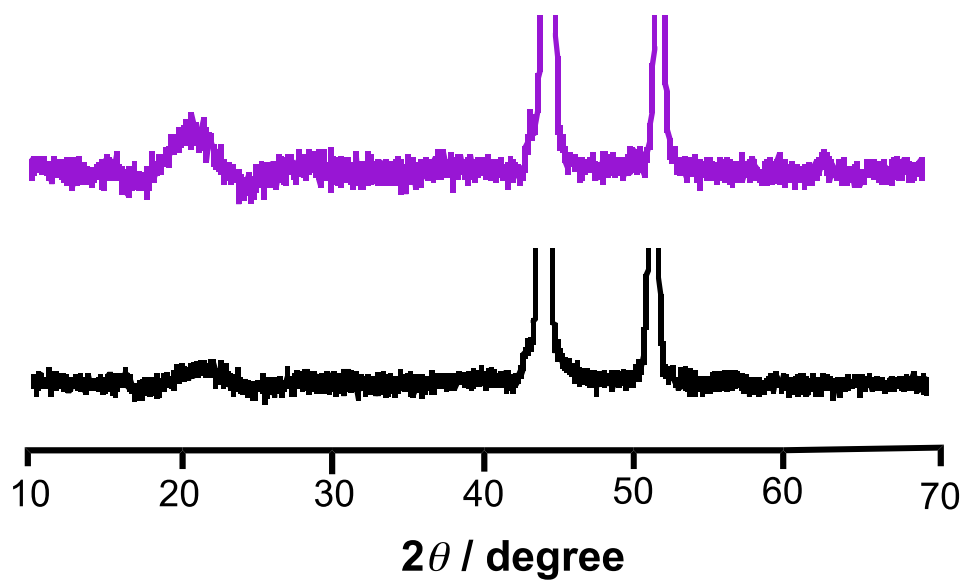
**Table S1.** Comparison of the Pt-film(*w*) electrode with the state-of-the-art Pt-based electrocatalysts with the excellent  $\eta^{10}$  values less than 200 mV for HER performances in 1.0 M KOH media (pH 14) <sup>a)</sup>.

Catalysts	Current collectors	$\Gamma_{\text{Pt}}$ (mg cm <sup>-2</sup> )	Fabrication methods	$\eta^{10} / \eta^{100}$ (mV)	Tafel slope (mV dec <sup>-1</sup> )	Stability	$j_m$ at $\eta = 50$ mV (A cm <sup>-2</sup> mg <sub>Pt</sub> <sup>-1</sup> )	Ref.
Pt-film( <i>w</i> )	NF	1.08	Drop casting/calcination	4.2/26.6	23	CP: $\eta^{10}$ and $\eta^{100}$ remained constant at $\sim 5.0$ and 27 mV for 60 h.	0.42	TW
Pt	GC	0.098	Drop casting/calcination	60/170	62	CP: $\eta^{10}$ increased from 60 to 131 mV after 20 h.	0.073	S1
Pt	FTO	0.098	Drop casting/calcination	198/NA	142	CP: $\eta^{10}$ increased from 150 to 230 mV after 20 h.	NA	S2
Pt <sub>3</sub> Ni-NiS	NF	$\sim 1.0$	Electrophoretic	12/73	24	CA: At $\eta = 12$ mV, $j$ remained constant at 10 mA cm <sup>-2</sup> for 192 h	0.075	S3
Pt	NF	$\sim 1.0$	Electrophoretic	22/107	31	CA: At $\eta = 22$ mV, $j$ decreased from 10 to 6 mA cm <sup>-2</sup> after 48h.	0.035	S3
Pt/NiO/Ni	NF	0.092	Electrodeposition	34/110	39	CP: $\eta^{10}$ increased from 34 to 55 mV after 24 h.	0.220	S4
Pt/Ni	NF	0.113	Electrodeposition	50/150	56	NA	0.090	S4
Pt-Ni <sub>3</sub> N	Ni mesh	0.300	LSV deposition	50/110	36.5	CA: At $\eta = 50$ mV, $j$ decreased from 10 to 8 mA cm <sup>-2</sup> after 24h.	0.033	S5
PtO <sub>2</sub> -Ni(OH) <sub>2</sub> NS	Ti	0.0755	Hydrothermal	80/280 70/250	89	CP: $\eta^{20}$ remained constant at $\sim 300$ mV for 100 h.	0.093 0.132	S6
PtCo-Co	Ti mesh	0.0430	Hydrothermal/calcination	28/105	35	CA: $j$ remained constant at 20 mA cm <sup>-2</sup> for 50 h, but $\eta$ was not shown.	0.51	S7
Pt(111) modified by Ni(OH) <sub>2</sub>	Pt	NA	Electrodeposition	138/NA	100-130	CA: At $\eta = 80$ and 110 mV, $j$ remained constant at 3.7 and 7.7 mA cm <sup>-2</sup> for 2.25 h, respectively.	NA	S8
Pt modified by Ni(OH) <sub>2</sub>	Pt	NA	Chemical deposition	$\sim 45$ ( $\eta^5$ )/NA	75	NA	NA	S9
Pt <sub>SA</sub> -Co(OH) <sub>2</sub>	Ag	0.059	Cyclic voltametric deposition	29/104	35.7	CP: $\eta^{20}$ remained constant at 65 mV for 50 h, but $\eta^{100}$ increased from 140 to 150 after 20 h.	0.42	S10
Pt <sub>13</sub> Cu <sub>73</sub> Ni <sub>14</sub> /CNF	C felt	6.653	Impregnation/galvanic displacement	67/NA	54	CA: At $\eta = 100$ mV, $j$ decreased by 18% of its initial value after 0.28 h	0.013	S11
Pt <sub>3</sub> Ni <sub>2</sub> NW-S/C	GC	0.077	Drop casting with	51/NA	NA	CP: $\eta^5$ increased from 30 to 40 after	0.120	S12

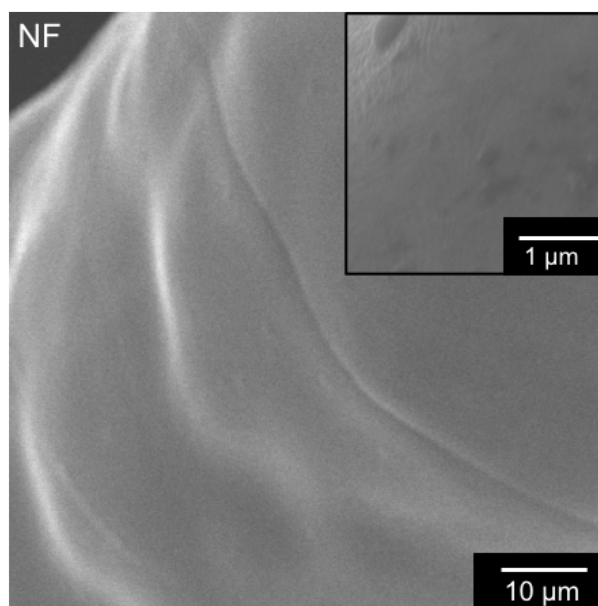
Pt-Ni AS	GC	0.0170	Nafion Drop casting with Nafion	27.7/NA	27	5h. NA	1.50	S13
Pt <sub>3</sub> Ni/C	GC	0.0100	Drop casting with Nafion	63 ( $\eta^5$ )/NA	NA	NA	0.31	S14
nanoframs/Ni(OH) <sub>2</sub> Pt NW/SL-Ni(OH) <sub>2</sub>	GC	0.0161	Drop casting with Nafion	70 ( $\eta^{2.5}$ )/N A	NA	NA	0.09	S15
Pt-BP/GR	GC	0.0143	Drop casting with Nafion	21/80	46.9	NA	3.2	S16
Pt <sub>SA</sub> -NiO/Ni/Ag NW	Flex. cloth	0.0054	Hydrothermal/ electrodeposition	26/85	27.1	CP: $\eta^{20}$ remained constant at 40 mV for 30 h.	6.85	S17
Pt <sub>SA</sub> -MoSe <sub>2</sub>	GC	0.0029	Drop casting with Nafion	29/110	41	NA	5.17	S18
Pt <sub>SA</sub> -N-C	GC	0.0063	Drop casting with Nafion	46/220	36.8	CP: $\eta^{10}$ remained constant at 46 mV for 20 h.	1.90	S19
Pt <sub>SA</sub> /AG	GC	0.0311	Drop casting with Nafion	12/NA	30.6	NA	0.80	S20
In-Pt <sub>SA</sub> NW/C	GC	0.0128	Drop casting with Nafion	46/NA	32.4	CP: $\eta^{10}$ remained constant at 46 mV for 5 h.	0.86	S21
Hcp-Pt-Ni/C	GC	0.0076	Drop casting with Nafion	65/NA	78	CA: At $\eta = 65$ mV, $j$ decreased from 10 to 5 mA cm <sup>-2</sup> after 1h.	1.60	S22
PtNi-O/C	GC	0.0051	Drop casting with Nafion	40/105	78.8	CP: $\eta^{10}$ increased from 40 to 100 mV after 10 h.	3.92	S23
Pt <sub>3,6</sub> Ni-S NW/C	GC	0.0153	Drop casting with Nafion	20/NA	114.8	CP: $\eta^5$ increased from 15 to 33 mV after 5 h.	2.61	S24
PtNi NP/Ni NSA	C cloth	0.0693	Drop casting with Nafion	38/NA	42	CP: $\eta^{20}$ increased from 50 to 90 mV after 90 h.	NA	S25
Pt <sub>3</sub> Ni <sub>3</sub> NW/C-air	GC	0.0153	Drop casting with Nafion	40/NA	NA	CP: $\eta^5$ increased from 30 to 40 mV after 3 h.	0.65	S26, 27
Pt <sub>1</sub> Ru <sub>1.54</sub> NC/BP	GC	0.0148	Drop casting with Nafion	22/75	19	CP: At $\eta = 22$ mV, $j$ decreased from 10 to 7 mA cm <sup>-2</sup> after 20 h.	3.38	S28
Pt-Co(OH) <sub>2</sub>	C cloth	0.3900	Electrodeposition	32/115	70	CP: $\eta^5$ increased from 32 to 90 mV after 20 h,	0.05	S29
Pt-Ni octahedra/C	GC	0.0062	Drop casting with	70/NA	59	CP: $\eta^4$ increased from 25 to 50 mV	1.81	S30

Pt-2D-(NiOH) <sub>2</sub> /C	GC	0.0011	Nafion Drop casting with 3 Nafion	180/NA	72	after 1 h, CA: At $\eta = 100$ mV, $j$ decreased by 49% of its initial value after 5.56 h.	0.88	S31
PtNiCo alloy nanohexapod/C	GC	0.0100	Drop casting with Nafion	22( $\eta^5$ ) /NA	NA	NA	NA	S32

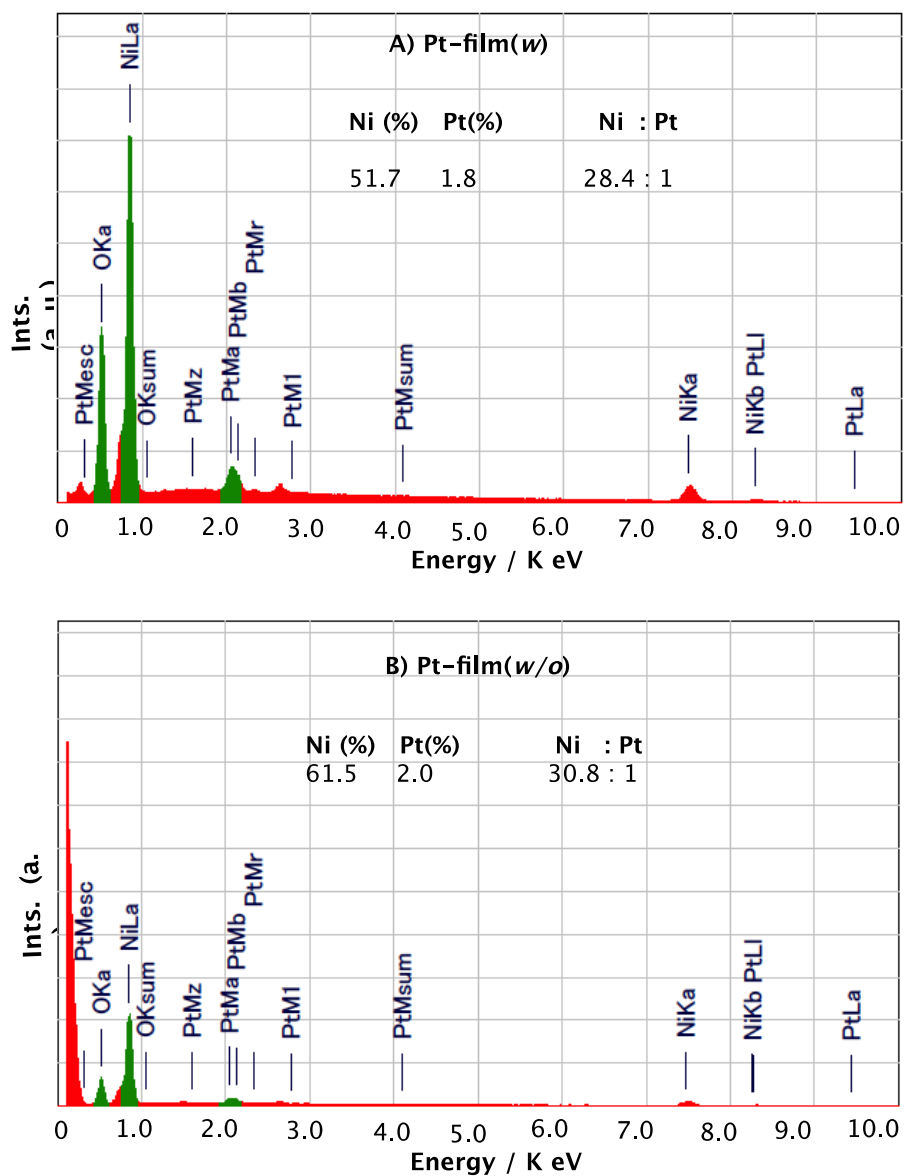
<sup>a)</sup> GC: Grassy carbon, TW: this work, NA: not available, CP: chronopotentiometry, CA: chronoamperometry, NS: nanosheet, Pt<sub>SA</sub>:Pt single atom, CNF: carbon nanofiber, NW: nanowires, AS: anisotropic, SL: single layered, BP: black phosphorous, GR: graphite, N-C: nitrogen doped carbon, AG: aniline-stacked graphene, Hcp: hexagonal close-packed superstructure, NP nanoparticle, NSA: nanosheet array, air: thermally annealed in air, NC: nanocrystals, 2D: two dimensional.



**Figure S1.** X-ray diffraction patterns (XRD) of the bare NF substrate before (black, below) and after (violet, above) calcination at 450 °C.



**Figure S2.** SEM image (top view) of the bare NF surface.



**Figure S3.** EDS spectra of (A) Pt-film(*w*) and (B) Pt-film(*w/o*) on the NF substrate, as measured with SEM observation.

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