Supplementary Materials for

Highly efficient and stable platinum film deposited via a mixed metal-imidazole casting method as a benchmark cathode for electrocatalytic hydrogen evolution

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Table S1. Comparison of the Pt(*w*-MeIm) electrode with the state-of-the-art Pt-based cathodes with the excellent η^{10} values less than 200 mV for HER performance in 1.0 M KOH media (pH 14).

Catalysts	Current collectors	Γ_{Pt} / mg cm^{-2}	Fabrication methods	η^{10} / mV	Tafel slope / mV dec^{-1}	Stability	Ref.
Pt(*w*-MeIm)	GC	0.098	Drop casting/calcination	60	62	η^{10} increased from 60 to 131 mV after 20 h.	TW
Pt	NF	~1.0	Electrophoretic	22	31	At η = 22 mV, *j* decreased from 10 to 6 mA cm^{-2} after 48h.	S1
Pt/NiO/Ni	NF	0.092	Electrodeposition	34	39	η^{10} increased from 34 to 55 mV after 24 h.	S2
Pt/Ni	NF	0.113	Electrodeposition	50	56	NA	S2
Pt-Ni_3N	Ni mesh	0.300	LSV deposition	50	36.5	At η = 50 mV, *j* decreased from 10 to 8 mA cm^{-2} after 24h.	S3
PtO_2-$Ni(OH)_2$ NS	Ti	0.0755	Hydrothermal	80 70	89	η^{20} remained constant at ~300 mV for 100 h.	S4
PtCo-Co	Ti mesh	0.0430	Hydrothermal/calcination	28	35	*j* remained constant at 20 mA cm^{-2} for 50 h, but η^{20} was not mentioned.	S5
Pt(111) modified by $Ni(OH)_2$	Pt	NA	Electrodeposition	138	100-130	At η = 80 and 110 mV, *j* remained constant at 3.7 and 7.7 mA cm^{-2} for 2.25 h.	S6
Pt modified by $Ni(OH)_2$	Pt	NA	Chemical deposition	~45 (η^5)	75	NA	S7
Pt_{SA}-$Co(OH)_2$	Ag	0.059	Cyclic voltametric deposition	29	35.7	η^{20} remained constant at 65 mV for 50 h and η^{100} increased from 140 to 150 after 20 h.	S8
$Pt_{13}Cu_{73}Ni_{14}$/CNF	C felt	6.653	Impregnation/galvanic displacement	67	54	At η = 100 mV, *j* decreased by 18% of its initial value after 0.28 h	S9
Pt_3Ni_2 NW-S/C	GC	0.077	Drop casting with Nafion	51	NA	η^5 increased from 30 to 40 after 5h.	S10
Pt-Ni AS	GC	0.0170	Drop casting with Nafion	27.7	27	NA	S11
Pt_3Ni/C nanoframs/$Ni(OH)_2$	GC	0.0100	Drop casting with Nafion	63 (η^5)	NA	NA	S12
Pt NW/SL-$Ni(OH)_2$	GC	0.0161	Drop casting with Nafion	70 ($\eta^{2.5}$)	NA	NA	S13

Pt-BP/GR	GC	0.0143	Drop casting with Nafion	21	46.9	NA	S14
Pt_{SA}-NiO/Ni/Ag NW	Flex. cloth	0.0054	Hydrothermal/ electrodeposition	26	27.1	η^{20} remained constant at 40 mV for 30 h.	S15
Pt_{SA}-$MoSe_2$	GC	0.0029	Drop casting with Nafion	29	41	NA	S16
Pt_{SA}-N-C	GC	0.0063	Drop casting with Nafion	46	36.8	η^{10} remained constant at 46 mV for 20 h.	S17
Pt_{SA}/AG	GC	0.0311	Drop casting with Nafion	12	30.6	NA	S18
In-Pt_{SA} NW/C	GC	0.0128	Drop casting with Nafion	46	32.4	η^{10} remained constant at 46 mV for 5 h.	S19
Hcp-Pt-Ni/C	GC	0.0076	Drop casting with Nafion	65	78	At η = 65 mV, j decreased from 10 to 5 mA cm^{-2} after 1h.	S20
PtNi-O/C	GC	0.0051	Drop casting with Nafion	40	78.8	η^{10} increased from 40 to 100 mV after 10 h.	S21
$Pt_{3.6}$Ni-S NW/C	GC	0.0153	Drop casting with Nafion	20	114.8	η^{5} increased from 15 to 33 mV after 5 h.	S22
PtNi NP/Ni NSA	C cloth	0.0693	Drop casting with Nafion	38	42	η^{20} increased from 50 to 90 mV after 90 h.	S23
Pt_3Ni_3 NW/C-air	GC	0.0153	Drop casting with Nafion	40	NA	η^{5} increased from 30 to 40 mV after 3 h.	S24, 25
$Pt_1Ru_{1.54}$ NC/BP	GC	0.0148	Drop casting with Nafion	22	19	At η = 22 mV, j decreased from 10 to 7 mA cm^{-2} after 20 h.	S26
Pt-$Co(OH)_2$	C cloth	0.3900	Electrodeposition	32	70	η^{5} increased from 32 to 90 mV after 20 h,	S27
Pt-Ni octahedra/C	GC	0.0062	Drop casting with Nafion	70	59	η^{4} increased from 25 to 50 mV after 1 h,	S28
Pt-2D-$(NiOH)_2$/C	GC	0.00113	Drop casting with Nafion	180	72	At η = 100 mV, j decreased by 49% of its initial value after 5.56 h.	S29
PtNiCo alloy nanohexapod/C	GC	0.0100	Drop casting with Nafion	22 (η^{5})	NA	NA	S30

GC: Grassy carbon, TW: this work, NA: not available, NS: nanosheet, Pt_{SA}: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: the Pt_3Ni_3 NW/C catalyst is thermally annealed in air, NC: nanocrystals, , 2D: two dimensional.

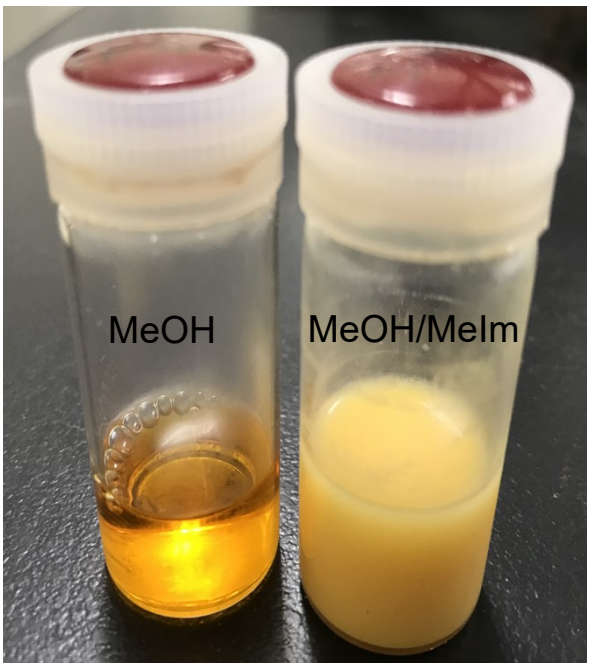

Figure S1. Photos of H_2PtCl_6 (50 mM) precursor solutions in MeOH (left) and MeOH/MeIm (right).

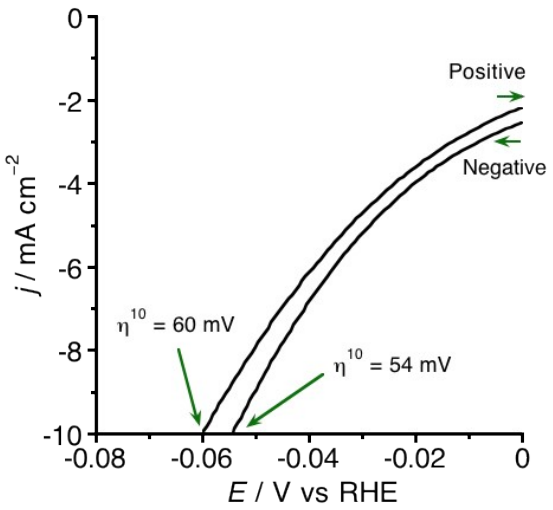

Figure S2. Comparison of LSVs (*iR* corrected) of the Pt(*w*-MeIm) film on a GC electrode (1.0 cm^2 of geometric area, Γ_{Pt} = 0.5 μmol (97.5 μg) cm^{-2}) in 1.0 M KOH solutions (pH 14.0) as measured between positive and negative potential scan directions at a scan rate of 5 mV s^{-1}. The η^{10} value (54 mV) of the Pt(*w*-MeIm) electrode for HER measured in the negative scan direction was lower than (60 mV) in the positive scan direction due to contribution of the current by reduction of the material itself.

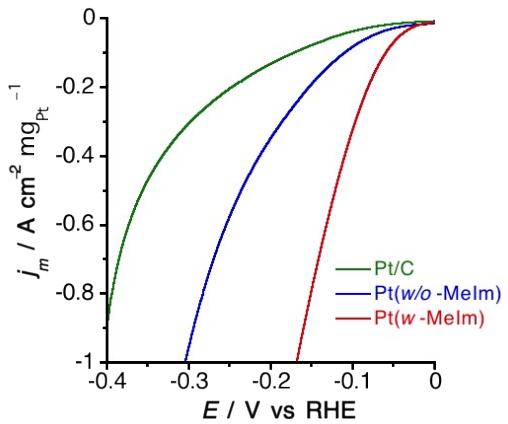

Figure S3. LSVs based on mass activities of the Pt(*w*-MeIm), Pt(*w/o*-MeIm), and Pt/C films on GC electrodes. (1.0 cm^2 of geometric area, Γ_{Pt} = 0.5 μmol cm^{-2}) in 1.0 M KOH solutions (pH 14.0) at a scan rate of 5 mV s^{-1} (The films are shown by different colors in the figure).

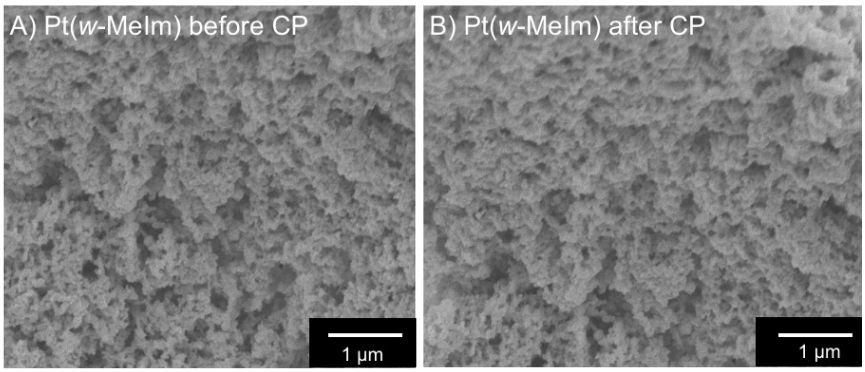

Figure S4. SEM images (top view) of Pt(*w*-MeIm) on GC (A) before and (B) after chronopotentiometry (CP) at 10 mA cm^{-2} for 20 h.

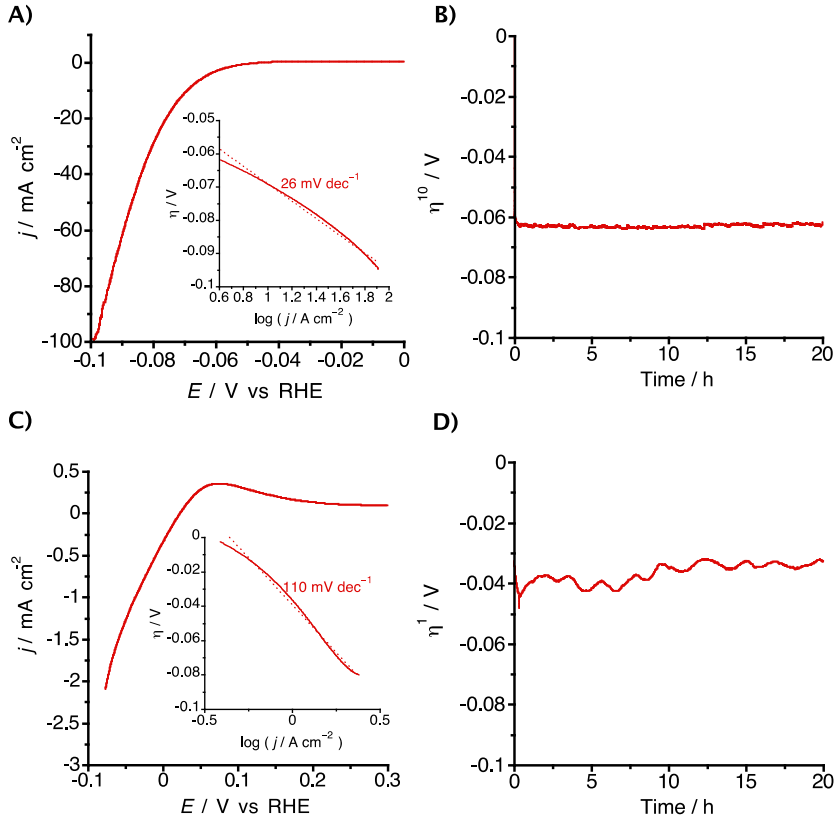

Figure S5. LSVs (*iR* corrected) of the Pt(*w*-MeIm) film on GC electrodes (1.0 cm^2 of geometric area, Γ_{Pt} = 0.5 µmol (97.5 µg) cm^{-2}) in (A) a H_2SO_4 (pH 0.5) and (C) phosphate buffer (0.1 M, pH = 7.0) solutions as measured in the positive potential scan direction at a scan rate of 5 mV s^{-1}. Insets show Tafel plots (solid lines), Tafel slopes (dashed lines), and values based on the LSV data. Chronopotentiograms of η^{10} and η^{1} for -10 and -1.0 mA cm^{-2} of current densities for HER in (B) a H_2SO_4 (pH 0.5) and (D) phosphate buffer (0.1 M, pH = 7.0) solutions, respectively.

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