

Supporting Information

Innovative Approach for Preparing CNT-supported Pt-Nanoparticle Functional Electrocatalyst using Protic Ionic Liquids

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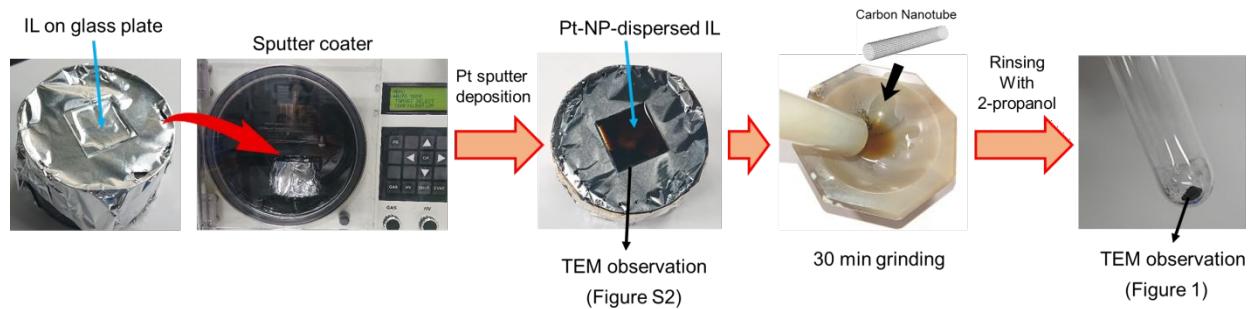


Figure S1. The procedure for the preparation of Pt-NP-modified CNT electrocatalysts.

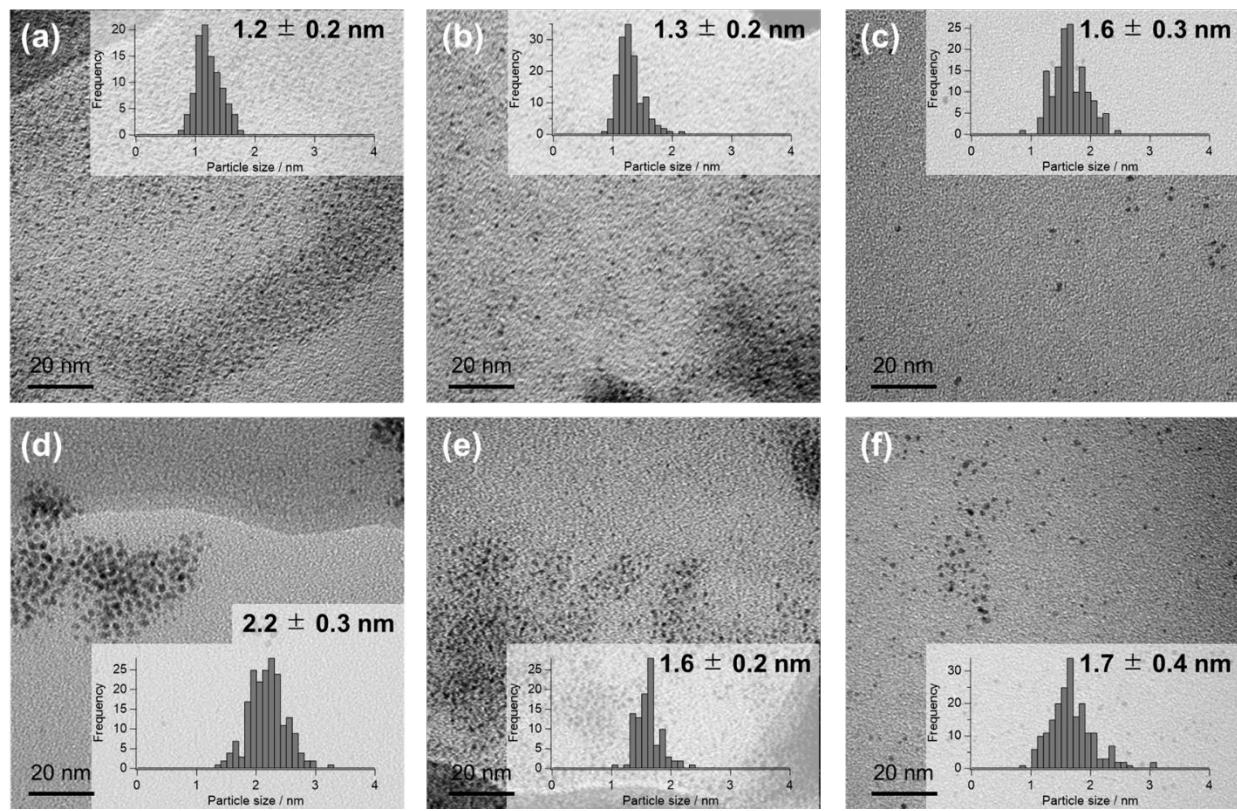


Figure S2. TEM images with particle size distribution of Pt NPs prepared by Pt-sputtering onto different ILs; (a) $[N_{1,1,1,3}][TFSA]$, (b) $[C_4mim][TFSA]$, (c) $[C_4mim][TfO]$, (d) $[dema]HSO_4$, (e) $[dema][TFSA]$, and (f) $[dema][TfO]$.

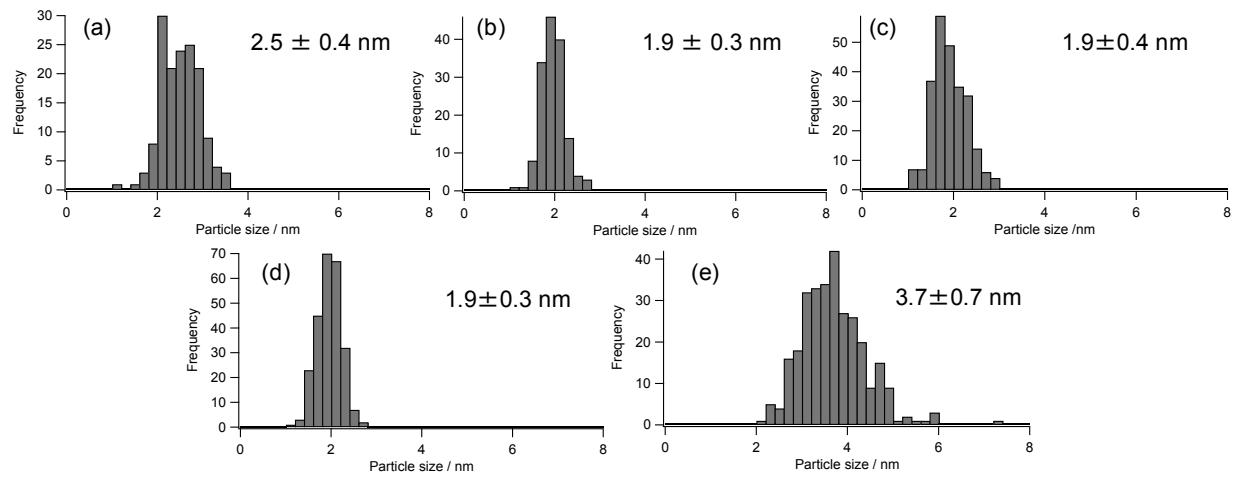


Figure S3. The particle size distributions of Pt-SWCNT prepared using (a) [dema]HSO₄, (b) [dema][TFSA], and (c) [dema][TfO], and those of (d) Pt-MWCNT and (e) commercial Pt–C.

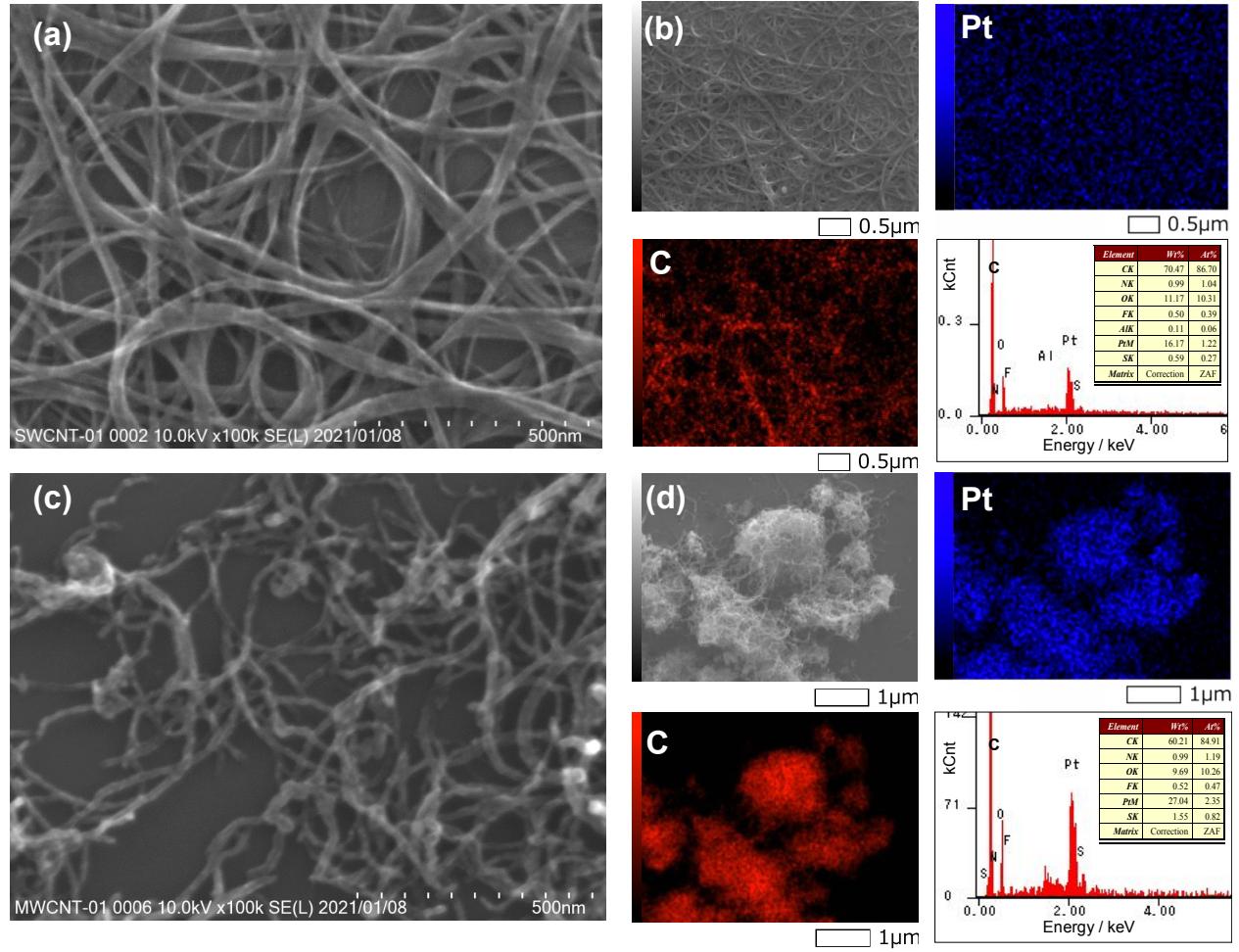


Figure S4. SEM images and SEM-EDX spectra and mapping images for Pt and C of (a,b) Pt-SWCNT and those of (c,d) Pt-MWCNT.

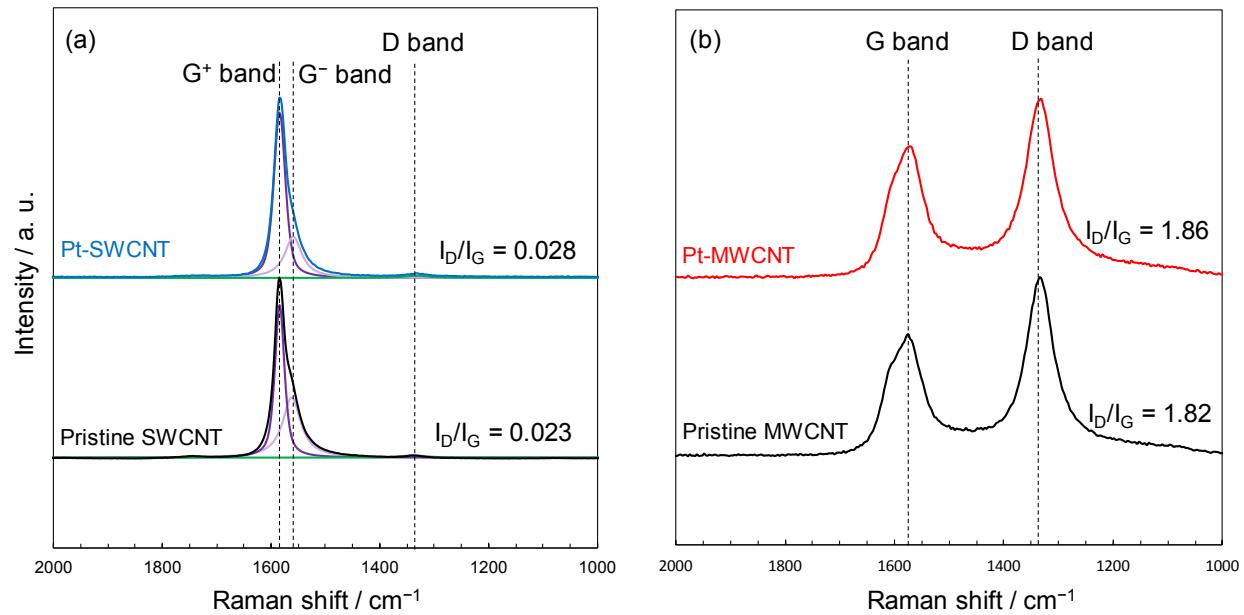


Figure S5. Raman spectra of (a) pristine SWCNT (black), Pt-SWCNT (blue), (b) pristine MWCNT (black), and Pt-MWCNT (red).

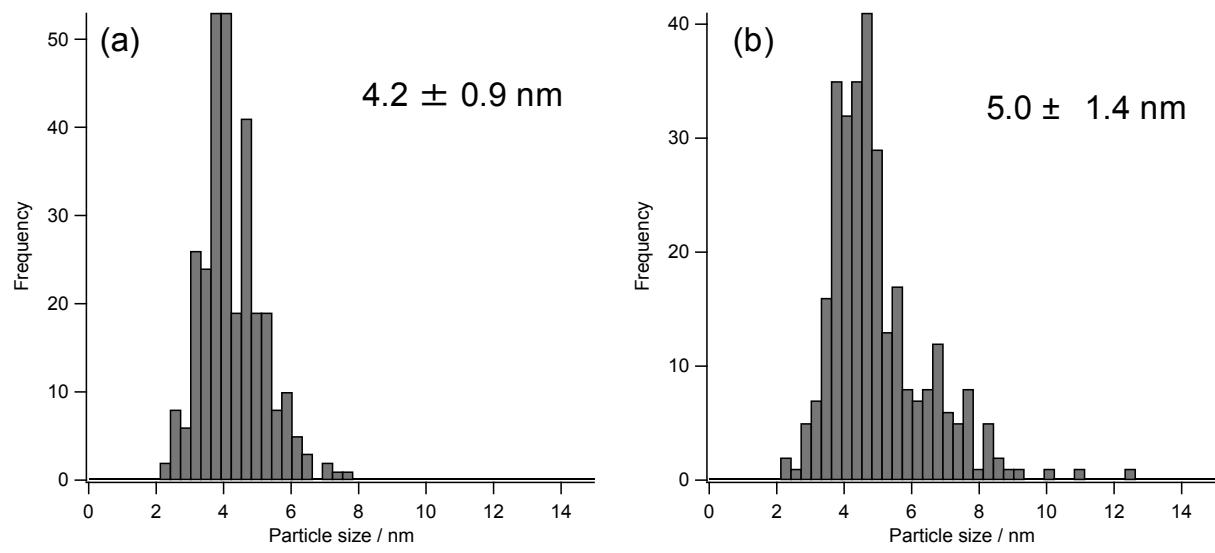


Figure S6. Pt NP size distribution of (a) Pt-MWCNT and (b) Pt–C subjected to 20,000-cycle ADT.

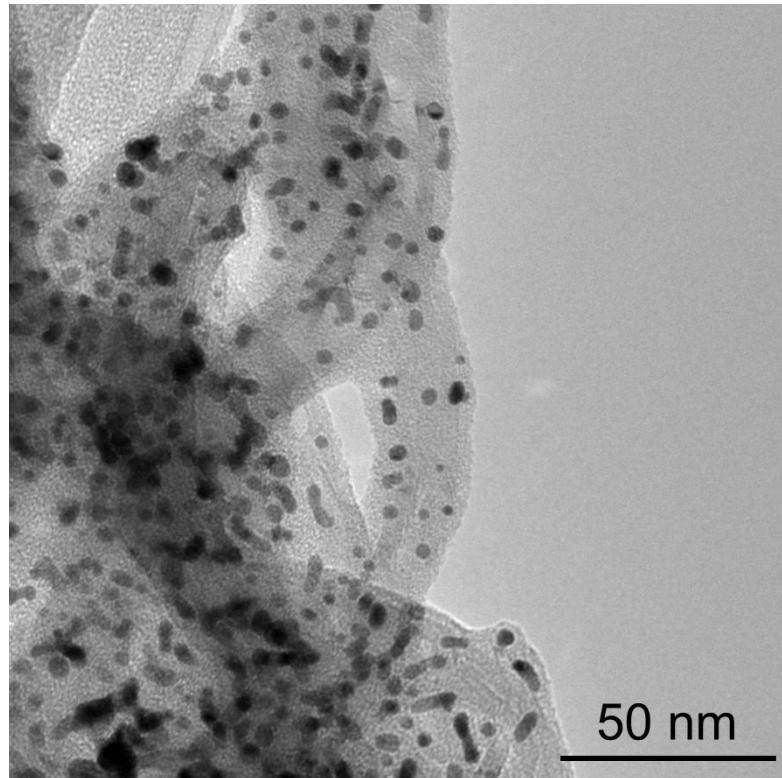


Figure S7. TEM image of Pt-MWCNT subjected to 20,000-cycle ADT.

Table S1. Performance comparison of CNT-supported Pt NP electrocatalysts reported in literatures

	CNT pretreatment	Immobilization of Pt NPs on CNT	ADT condition	ADT cycle number	Mean particle size /nm		ECSA /m ² g _{Pt} ⁻¹		Mass activity /A g _{Pt} ⁻¹		Specific activity /A m _{Pt} ⁻²		TOF** /atom _{Pt} ⁻¹ sec ⁻¹		Reference
					Initial	After ADT	Initial	After ADT	Initial	After ADT	Initial	After ADT	Initial	After ADT	
Pt/MU-MWCNT	Unzipping CNT with potassium permanganate	Glycol reduction of Pt ion on CNT	Potential sweep, 0.6–1.1 V, 50 mV sec ⁻¹	3000	1.95	-	103.4	82.2	174.5	93.3*	1.69	1.13	0.21	0.14	S1
Pt@CNx/CNT	Coating CNT with polyaniline and pyrolyzing it	Chemical reduction of Pt ion at same time as aniline polymerization on CNT	Potential sweep, 0–1.2 V, 50 mV sec ⁻¹	1500	3.1 ± 0.9	3.2 ± 0.7	74.3	77.0	111 (0.85 V)	108.5 (0.85 V)	1.5 (0.85 V)	1.41 (0.85 V)	0.19 (0.85 V)	0.18 (0.85 V)	S2
PtNW/F-CNT	Sulfur-doping functionalized CNT by annealing with phenyl disulfide at 1000°C	Polyol reduction of Pt ion on sulfur-doped CNT with autoclave	Potential sweep, 0–1.5 V	3000	Nanowire	-	17.2	16.0	272 (0.9 V)	220 (0.9 V)	15.8 (0.9 V)	13.8 (0.9 V)	2.00 (0.9 V)	1.75 (0.9 V)	S3
Pt/Ppy-MWCNT	Coating CNT with polypyrrole	Copolymerization of Pt complex and pyrrole on CNT	Square potential wave, 0.6 V (3 sec)–1.0 V (3 sec)	30000	1.5 ± 0.5	-	96.0	63.8*	166 (0.9 V)	124 (0.9 V)	1.7 (0.9 V)	1.9 (0.9 V)*	0.22 (0.9 V)	0.24 (0.9 V)	S4
Pt/N-CNT	Coating CNT with Nafion	Polyol reduction of Pt ion on CNT	Potential sweep, 0.6–1.2 V, 100 mV sec ⁻¹	6000	2.7	3.8	72.11	49.39	111 (0.9 V)	-	1.54 (0.9 V)	-	0.19 (0.9 V)	-	S5
Pt-Gd(5/1)-MWCNT	Coating CNT with polypyrrole	Copolymerization of Pt complex, pyrrole and Gd complex on CNT	Square potential wave, 0.6 V (3 sec)–1.0 V (3 sec)	30000	1.6 ± 0.9	-	68.8	36	350 (0.9 V)	120 (0.9 V)	5.0 (0.9 V)	3.4 (0.9 V)	0.63 (0.9 V)	0.43 (0.9 V)	S6
Pt-MWCNT	Nothing	Mixing Pt NP-dispersed IL and CNT at room temperature	Square potential wave, 0.6 V (3 sec)–1.0 V (3 sec)	20000	1.9 ± 0.3	4.2 ± 0.9	76.2	27.9	347.8 (0.9 V)	291.6 (0.9 V)	4.56 (0.9 V)	10.4 (0.9 V)	0.58 (0.9 V)	1.32 (0.9 V)	This work
Pt-SWCNT	Nothing	Mixing Pt NP-dispersed IL and CNT at room temperature	Square potential wave, 0.6 V (3 sec)–1.0 V (3 sec)	20000	1.9 ± 0.4	(2.6 ± 0.8 /6.9 ± 3.6)	100.2	51.4	353.3 (0.9 V)	374.8 (0.9 V)	3.52 (0.9 V)	7.29 (0.9 V)	0.45 (0.9 V)	0.92 (0.9 V)	This work

*: calculated from a bar graph in the literature

**: TOF is given by

$$\text{TOF (atom}_{\text{Pt}}^{-1}\text{sec}^{-1}) = \frac{\text{Specific activity (A m}_{\text{Pt}}^{-2})}{4 \text{ electron} \times e (\text{C electron}^{-1}) \times \text{surface atom number (atoms}_{\text{Pt}} \text{ m}_{\text{Pt}}^{-2})}$$

where the average of the atomic density for Pt(111), Pt(110), and Pt(100) (1.50×10^{15} , 0.92×10^{15} , and 1.28×10^{15} [atom cm⁻²], respectively) was used as the surface atom number.^{S7}

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