**Supporting Information**

# Enhanced CO Oxidation and Cyclic Activities in Three-Dimensional Platinum/Indium Tin Oxide/Carbon Black Electrocatalysts Processed by Cathodic Arc Deposition

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## 1) Comparisons in electrochemical performance of Sn or ITO-involved catalysts

**Table S1.** Processing and performance of the reported electrocatalysts incorporating Sn or ITO (with the electrocatalytic performance of the current study in the last row)

| Catalyst | Preparation method | ITO loading (%) | Pt loading (%) | ITO size (nm) | ECSA ($m^2g^{-1}$) | Mass activity ($A\ g^{-1}$) | Ref. |
|---|---|---|---|---|---|---|---|
| Pt-ITO-graphene | Chemical reduction | 75% | 20 | 10-12 | 67 (36.4) | 108 (84) | 1 |
| Pt/Sn-$In_2O_3$ | Chemical reduction | - | 20 | 17 | - | - | 2 |
| Pt/In-$SnO_2$ | Chemical reduction | - | 20 | 13 | - | - | |
| Pt-ITO-CNT | Chemical reduction | 30 | 20 | 10-20 | 24 | 19 | 3 |
| Pt/ITO | Chemical reduction | - | | 22 | 74.4 (61.5) | 243 | 4 |
| Pt/ITO | Chemical reduction | - | 20 | 5 | 83.1 | 621 | 5 |
| Pt/ITO | Co-precipitation | - | 40 | 20-30 | 22 | ~150 | 6 |
| Pt/ITO/CB | Arc deposition | 12.2 | 9.3 | 5.6 | 129.4 | ~296 | This study |

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[2] Liu, Y.; Mustain, W. E. Stability limitations for Pt/Sn–$In_2O_3$ and Pt/In–$SnO_2$ in Acidic Electrochemical Systems. *Electrochim. Acta* **2014**, *115*, 116-125.

[3] Park, S.; Shao, Y.; Viswanathan, V. V.; Liu, J.; Wang, Y. Electrochemical Study of Highly Durable Cathode with Pt Supported on ITO-CNT Composite for Proton Exchange Membrane Fuel Cells. *Ind. Eng. Chem. Res.* **2016**, *42*, 81-86.

[4] Zhao, S.; Wangstrom, A. E.; Liu, Y.; Rigdon, W. A.; Mustain, W. E. Stability and Activity of Pt/ITO Electrocatalyst for Oxygen Reduction Reaction in Alkaline Media . *Electrochim. Acta* **2015**, *157*, 175-182.

[5] Liu, Y.; Mustain, W. E. High Stability, High Activity Pt/ITO Oxygen Reduction Electrocatalysts. *J. Am. Chem. Soc*. **2013**, *135*, 530-533.

[6] Wang, G.; Niangar, E.; Huang, K.; Atienza, D.; Kumar, A.; Dale, N.; Oshihara, K.; Ramani, V. K. Indium Tin Oxide as Catalyst Support for PEM Fuel Cell: RDE and MEA Performance. *ECS Trans.* **2015**, 69, 1179-1205.

## 2) Effects of different number of discharge pulses in the CAD process of ITO

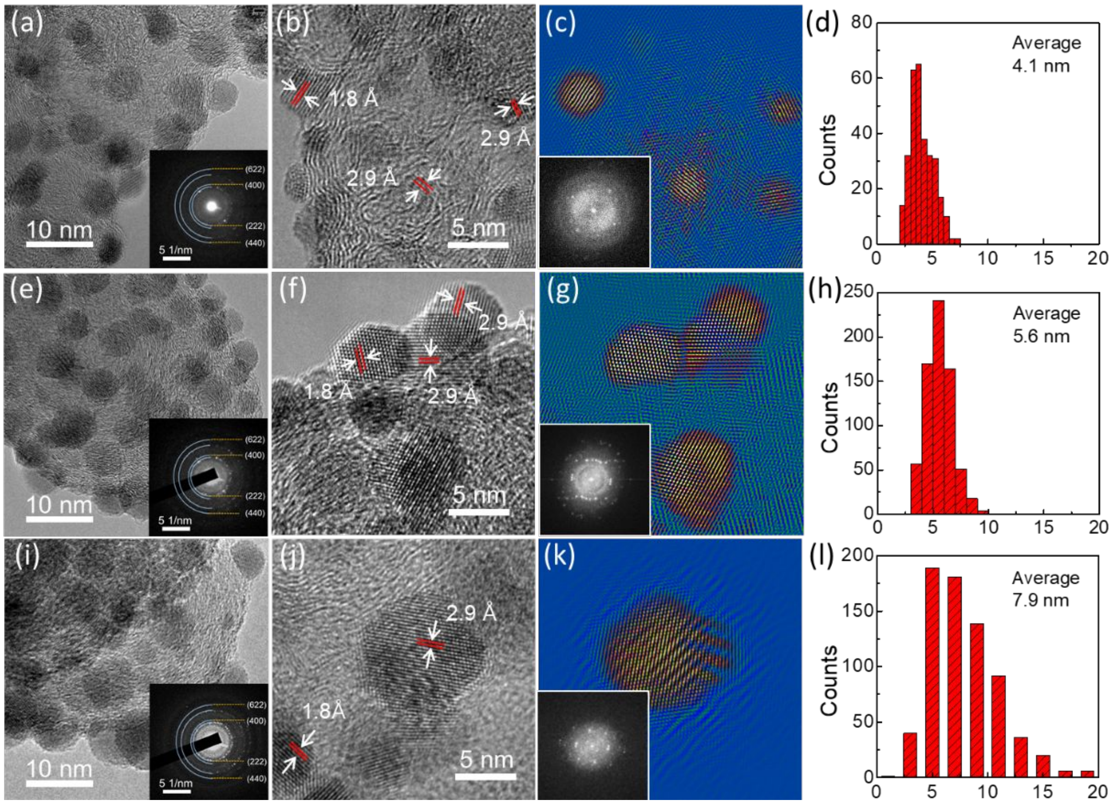


**Figure S1.** TEM, HR-TEM, and FFT filtered images and particle size distributions of ITO nanoparticles on carbon black support, which were processed by CAD with different discharge pulse rates of (a-c) 100,000, (e-g) 200,000, and (i-k) 300,000 with (d,h,l) the corresponding particle size distributions. The average particle size was noticed at the top of the distribution plots.

**3) Phase analysis and Sn concentration of the ITO-modified CB samples**

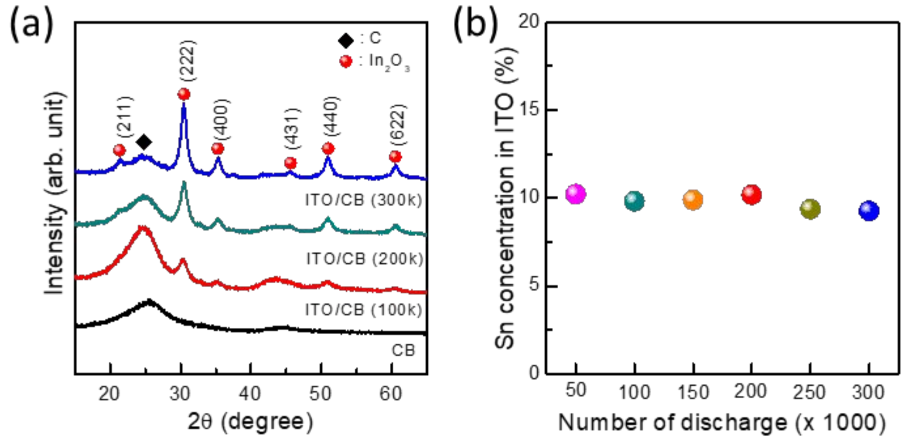


**Figure S2.** (a) XRD patterns of ITO-deposited CB samples with different discharge pulse rates of 100,000, 200,000, and 300,000 during the CAD process (the pattern of the CB support is included for reference) and (b) the variation of the Sn concentration as a function of the discharge number in ITO nanoparticles.

**4) TGA curves and the relation of ITO content with the number of discharge**

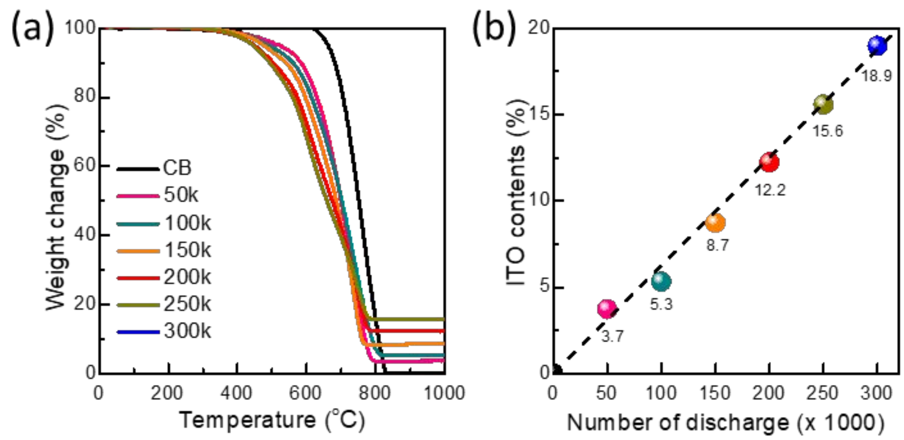


**Figure S3.** (a) TGA curves of ITO-modified CB samples with different number of discharge pulses and (b) the relation of ITO contents with the number of discharges.

## 5) TGA curves of the Pt/ITO/CB catalysts

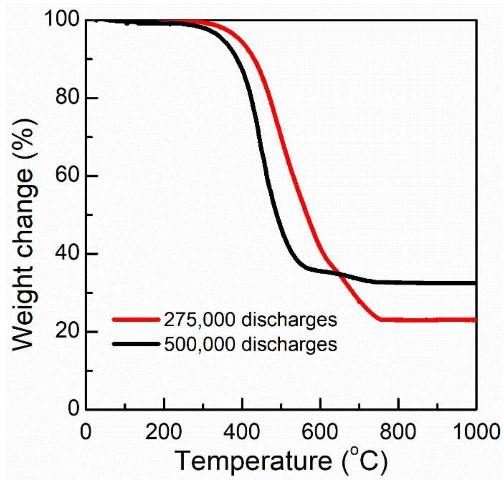


**Figure S4.** TGA curves of the Pt/ITO/CB samples processed with 275,000 and 500,000 discharges for the Pt synthesis.

## 6) XRD patterns and XPS spectra of the electrocatalysts

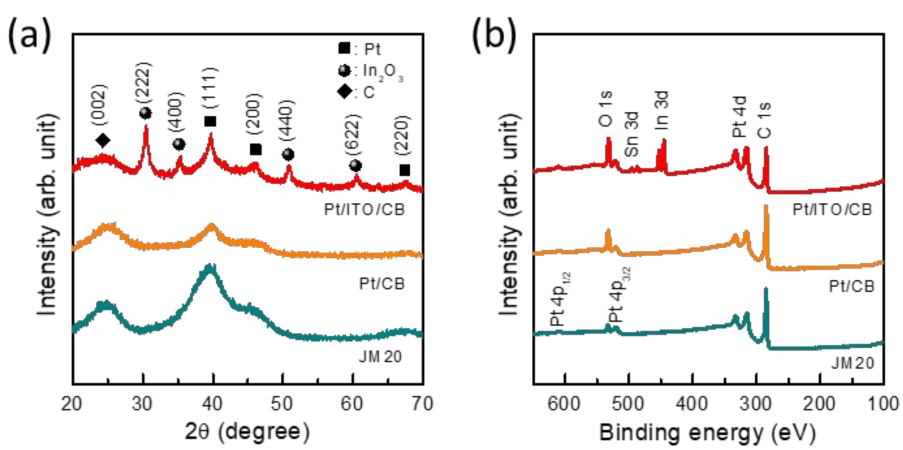


**Figure S5.** (a) XRD patterns and (b) XPS spectra of the JM20, Pt/CB, and Pt/ITO/CB electrocatalysts

## 7) Additional TEM images, and the size distributions of Pt nanoparticles

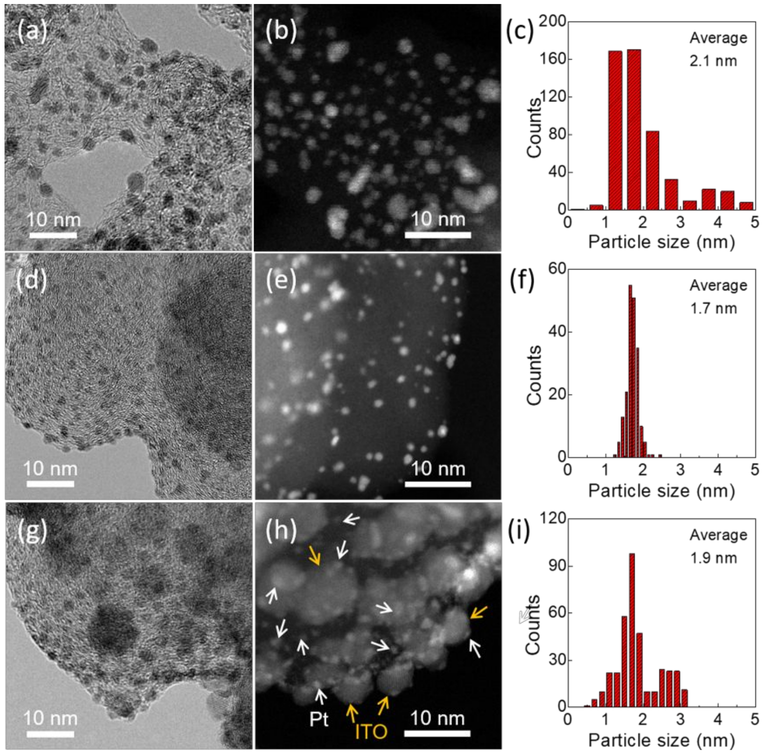


**Figure S6.** TEM, atomic-resolution HAADF images and particle size distributions of (a-c) JM20, (d-f) Pt/CB, and (g-i) Pt/ITO/CB. Particle size distributions were extracted from the TEM images, with the average Pt particle size of ~2.1 nm for JM20, ~1.7 nm for Pt/CB, and ~1.9 nm for Pt/ITO/CB.

## 8) TEM image and particle size distribution of Pt/CB after ADT

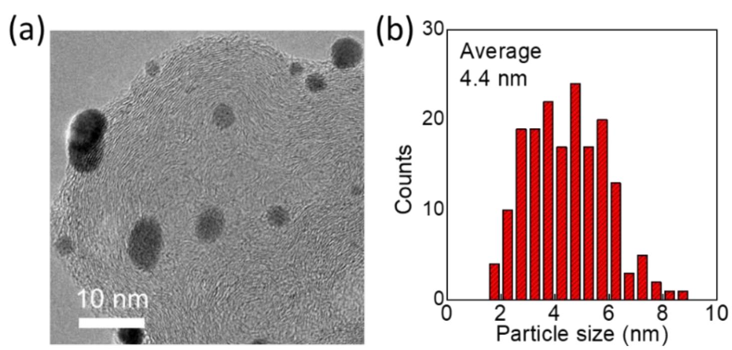


**Figure S7.** (a) TEM images and (b) the size distribution of Pt nanoparticles for the Pt/CB catalyst after the 3,000-cycle ADT evaluation.

## 9) Rotation rate-dependent ORR curves and the Koutecky-Levich plots

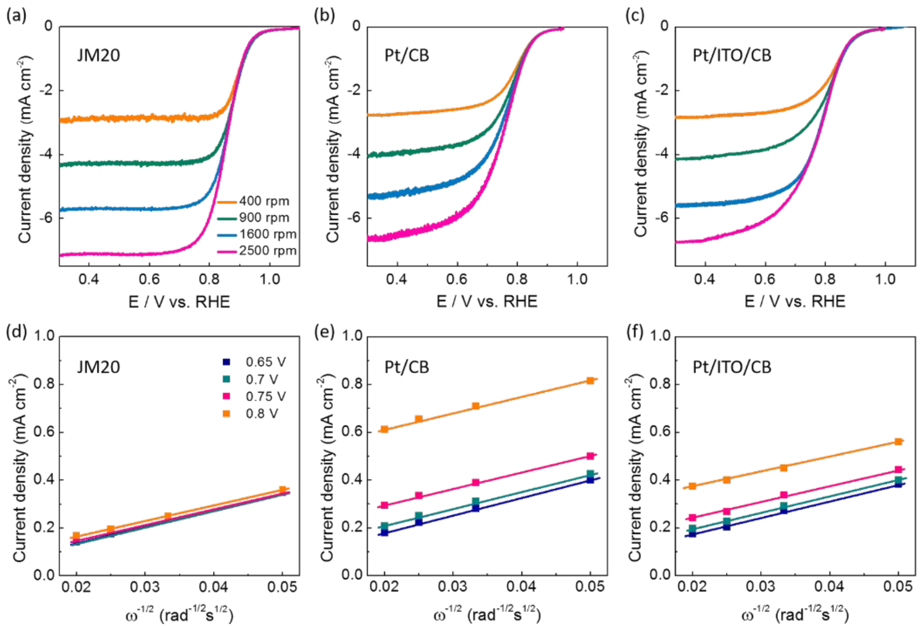


**Figure S8.** (a-c) Rotation rate-dependent current-potential curves and (d-f) the corresponding Koutecky-Levich plots at different potentials for JM20, Pt/CB, and Pt/ITO/CB.

## 10) CO stripping results of CB and ITO/CB

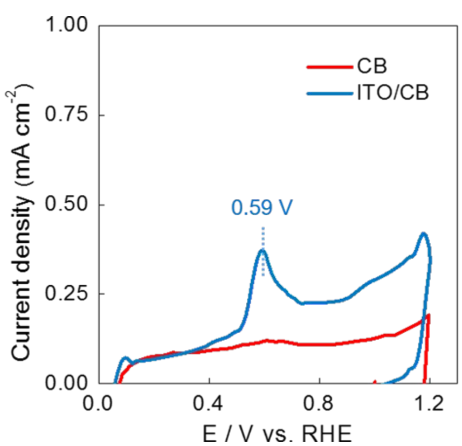


**Figure S9.** CO stripping voltammetry curves of CB (red line) and ITO/CB (blue line) support materials.

**11) CO stripping results of the reported RuPt-based electrocatalysts**

**Table S2.** CO stripping test results for the reported PtRu-base electrocatalysts

| Catalyst | Manufacturer | $E_P$(CO) V | Solution | Ref. |
|---|---|---|---|---|
| C/PtRu (60) | Alfa Aesar | 0.621 | 0.5 M $H_2SO_4$ | 1 |
| CB/PtRu (60) | Alfa Aesar | 0.68 | 0.5 M $H_2SO_4$ | 2 |
| PtRu/C (60) | Johnson Matthey | 0.51 | 0.5 M $H_2SO_4$ | 3 |
| Pt-Ru/CB (60) | Johnson Matthey | 0.56 | 0.5 M $H_2SO_4$ | this study |

(1) Yu, X.; Zhang, Q.; Ling, Y.; Yang, Z.; Cheng, H. Promoted Stability and Electrocatalytic Activity of PtRu Electrocatalyst Derived from Coating by Cerium Oxide with High Oxygen Storage Capacity. *Appl. Surf. Sci.* **2018**, *455*, 815-820.
(2) Luo, F.; Zhang, Q.; Qu, K.; Guo, L.; Hu, H.; Yang, Z.; Cai, W.; Cheng, H. Decorated PtRu Electrocatalyst for Concentrated Direct Methanol Fuel Cells. *ChemCatChem* **2019**, *11*, 1238-1243.
(3) Tian, M.; Shi, S.; Shen, Y.; Yin, H. PtRu Alloy Nanoparticles Supported on Nanoporous Gold as an Efficient Anode Catalyst for Direct Methanol Fuel Cell. *Electrochim. Acta* **2019**, *293*, 390-398.