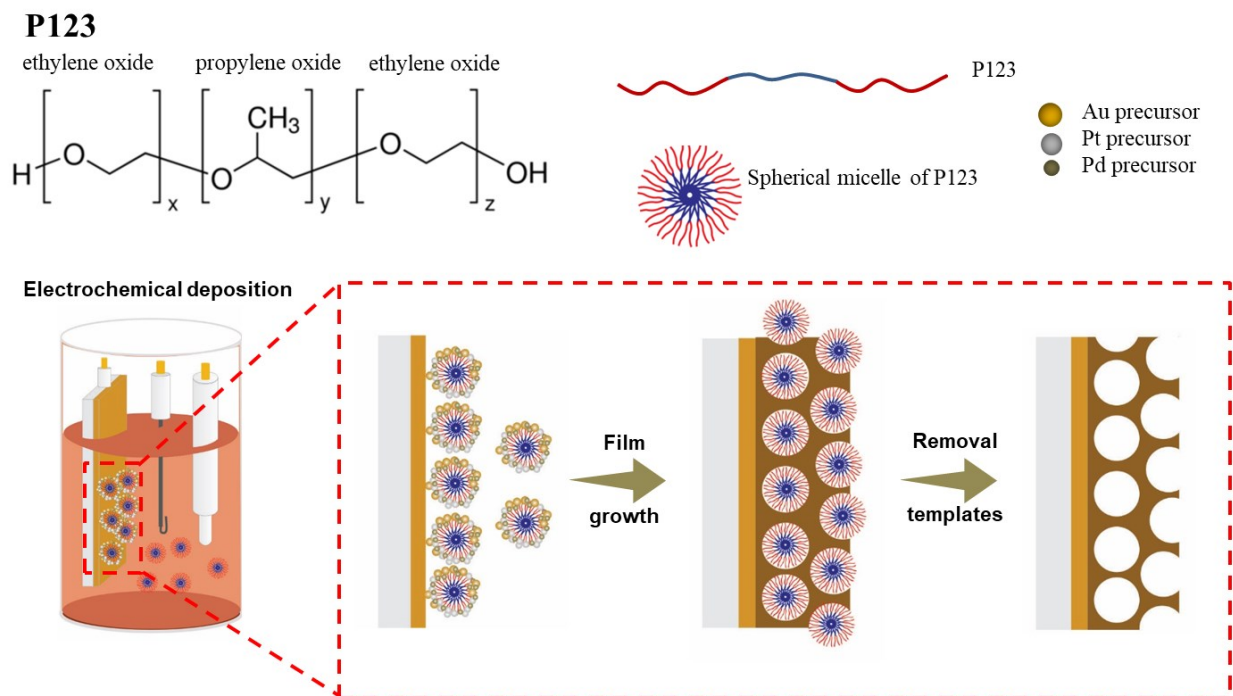
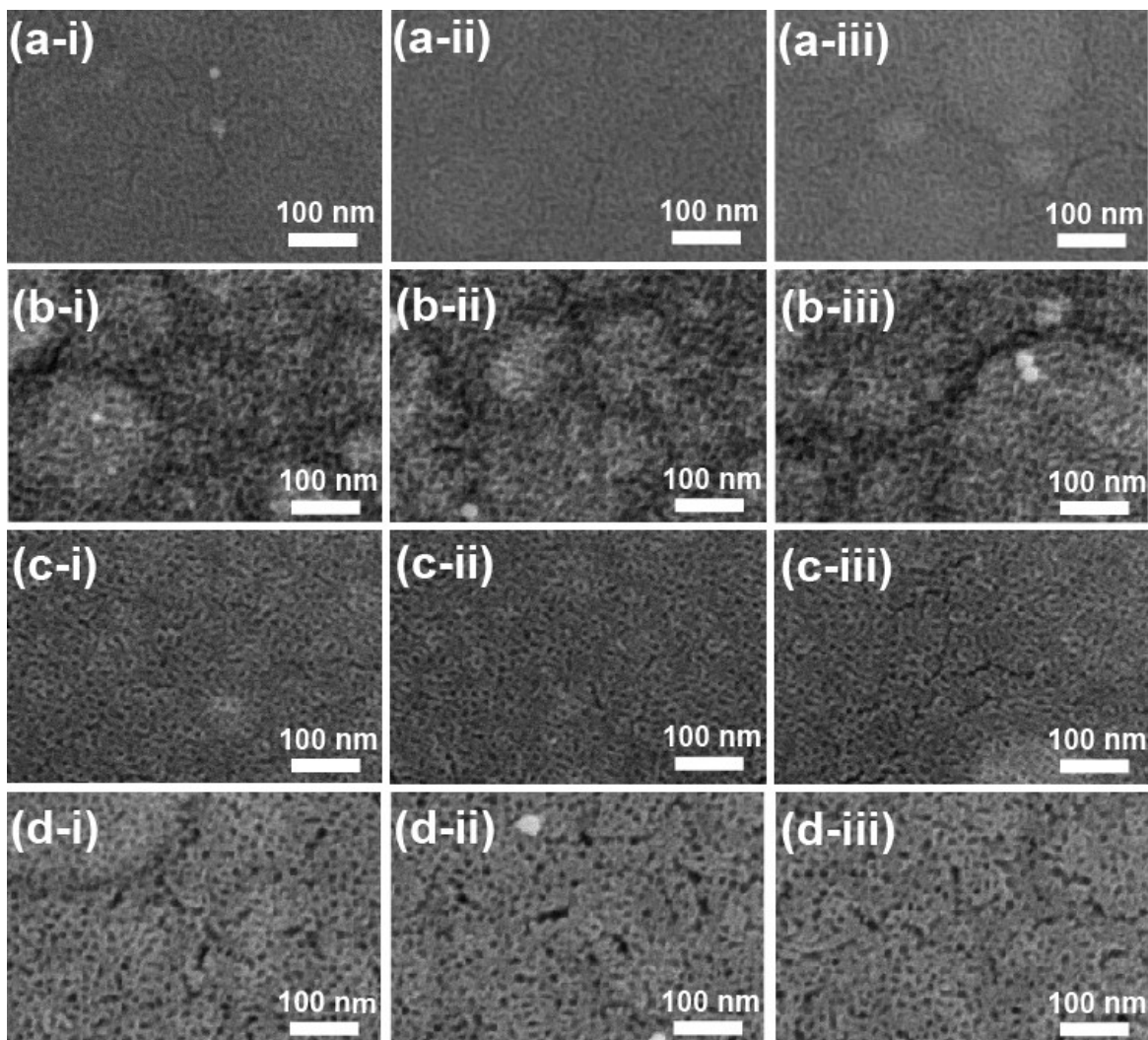


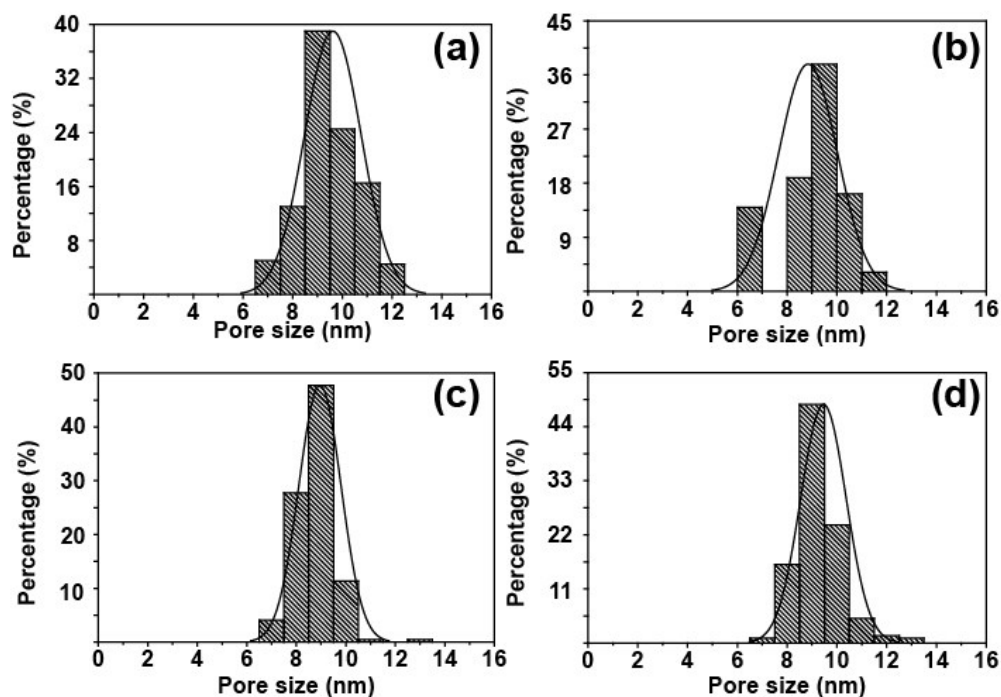
## Electronic supplementary information (ESI)



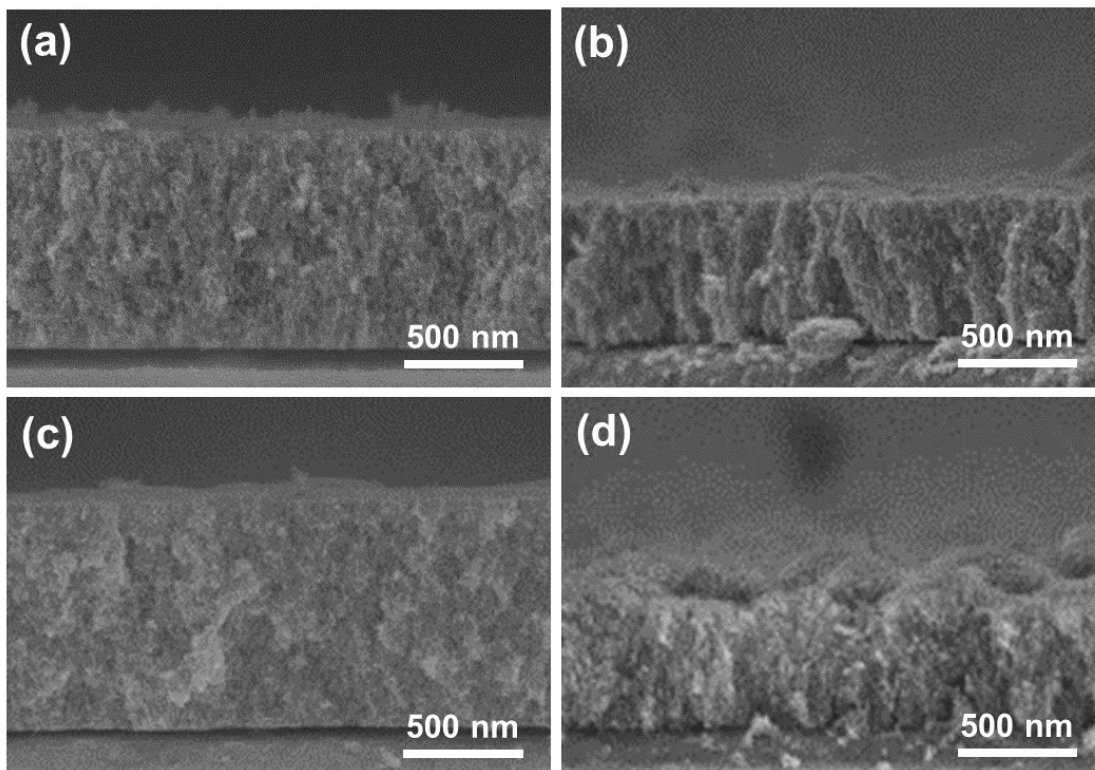
**Figure S1.** Schematic of fabrication of mesoporous PtPdAu alloy film by electrochemical deposition with the micelle assembly approach.



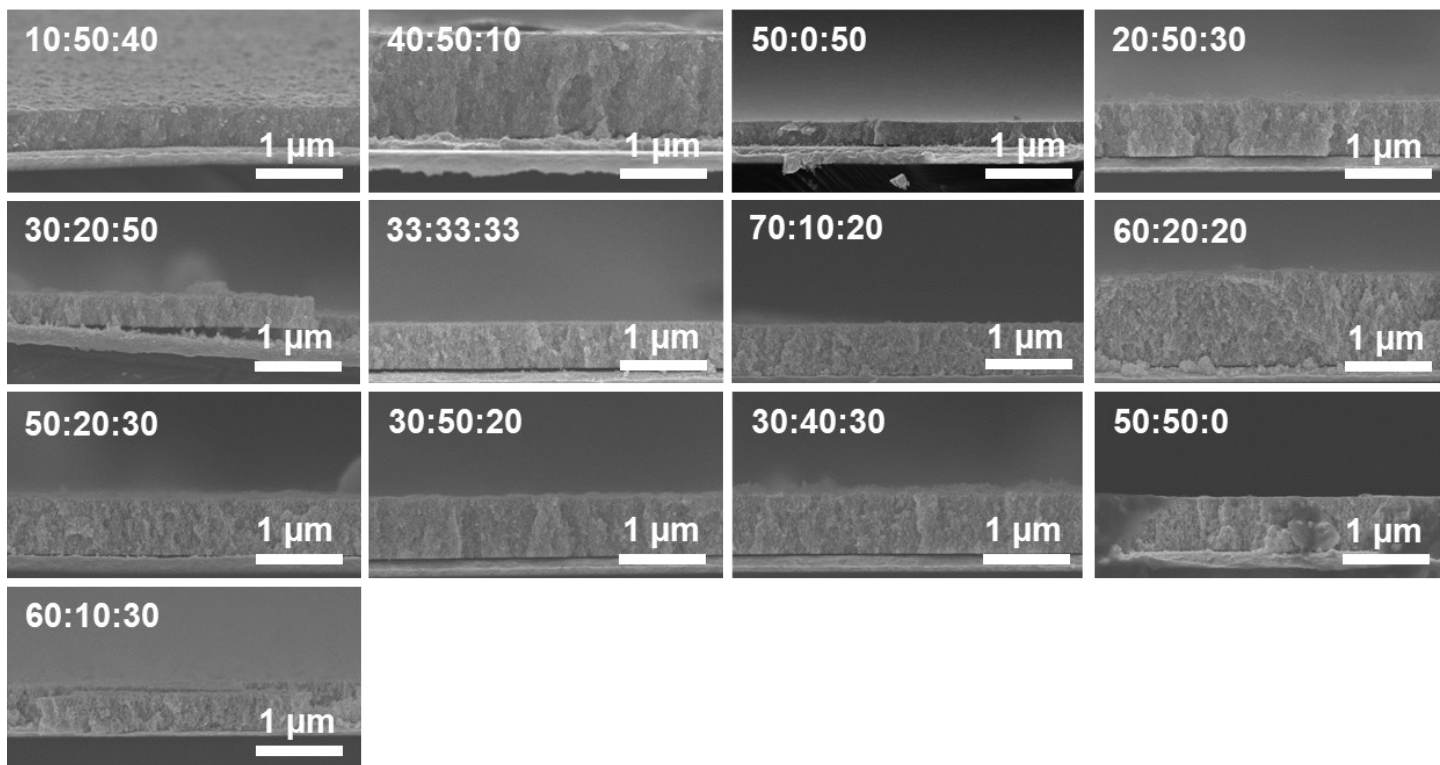
**Figure S2.** Top-view SEM images from 3 different samples (i, ii, and iii) of selected mesoporous PtPdAu films in the initial dataset with the chemical compositions of metal precursors  $[\text{PtCl}_4]^{2-}$ :  $[\text{PdCl}_4]^{2-}$ :  $[\text{AuCl}_4]^-$  of (a) 40: 40: 20, (b) 20: 40: 40, (c) 60: 30: 10, and (d) 30: 10: 60.



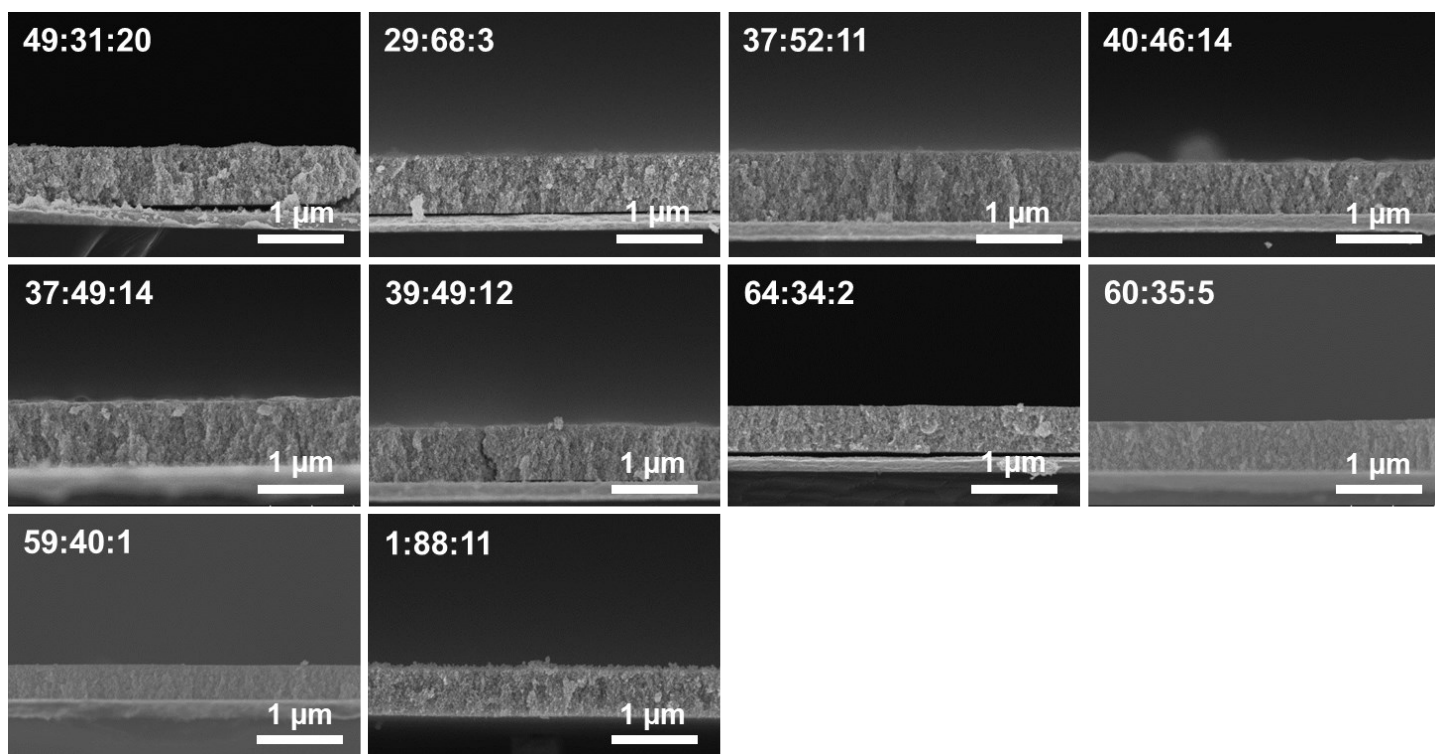
**Figure S3.** Mesopore size distribution of selected mesoporous PtPdAu films in the initial dataset with the chemical compositions of metal precursors [PtCl<sub>4</sub>]<sup>2-</sup>: [PdCl<sub>4</sub>]<sup>2-</sup>: [AuCl<sub>4</sub>]<sup>-</sup> of (a) 40: 40: 20, (b) 20: 40: 40, (c) 60: 30: 10, and (d) 30: 10: 60. Pore size distribution was taken from 3 samples for each condition with minimum 200 pores were measured.



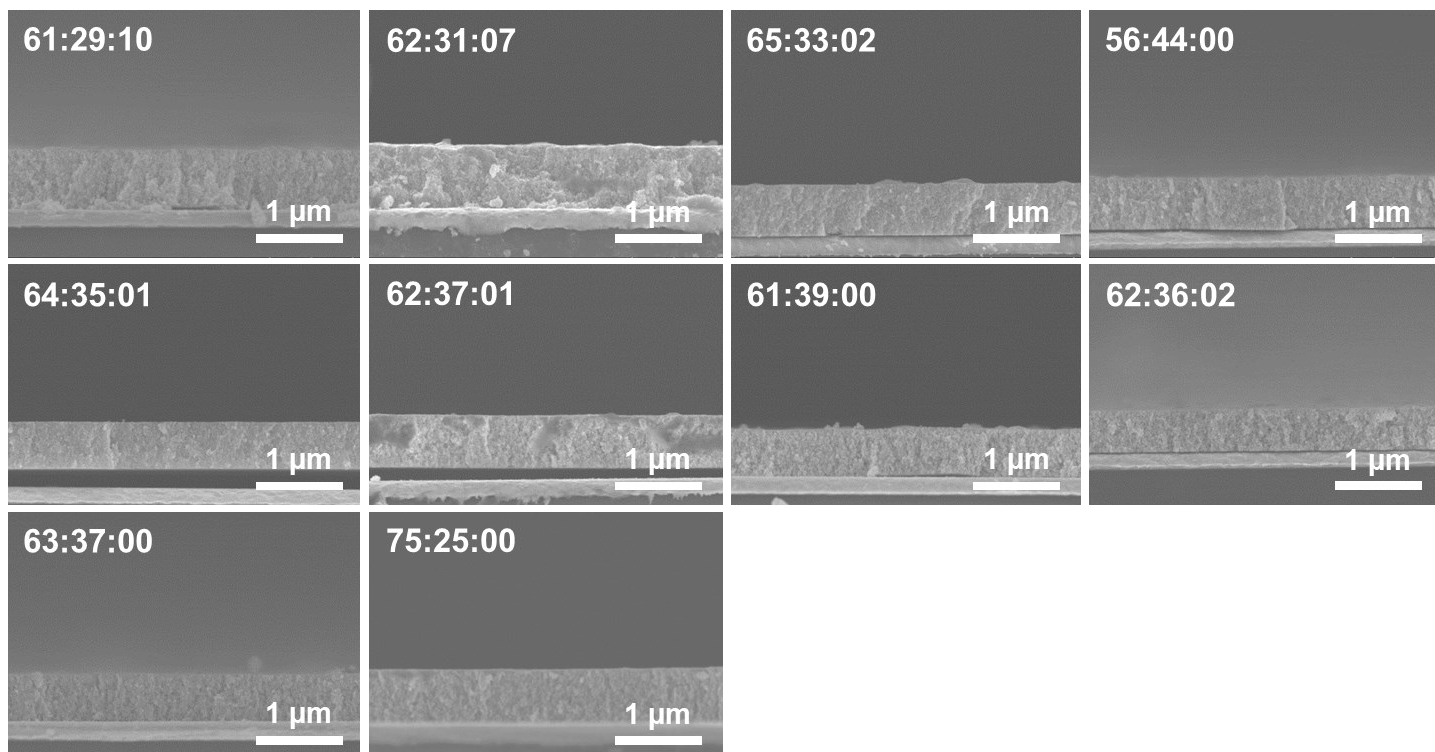
**Figure S4.** Cross-sectional view SEM images of selected mesoporous PtPdAu films in the initial dataset with the chemical compositions of metal precursors  $[\text{PtCl}_4]^{2-}$ :  $[\text{PdCl}_4]^{2-}$ :  $[\text{AuCl}_4]^-$  of (a) 40: 40: 20, (b) 20: 40: 40, (c) 60: 30: 10, and (d) 30: 10: 60.



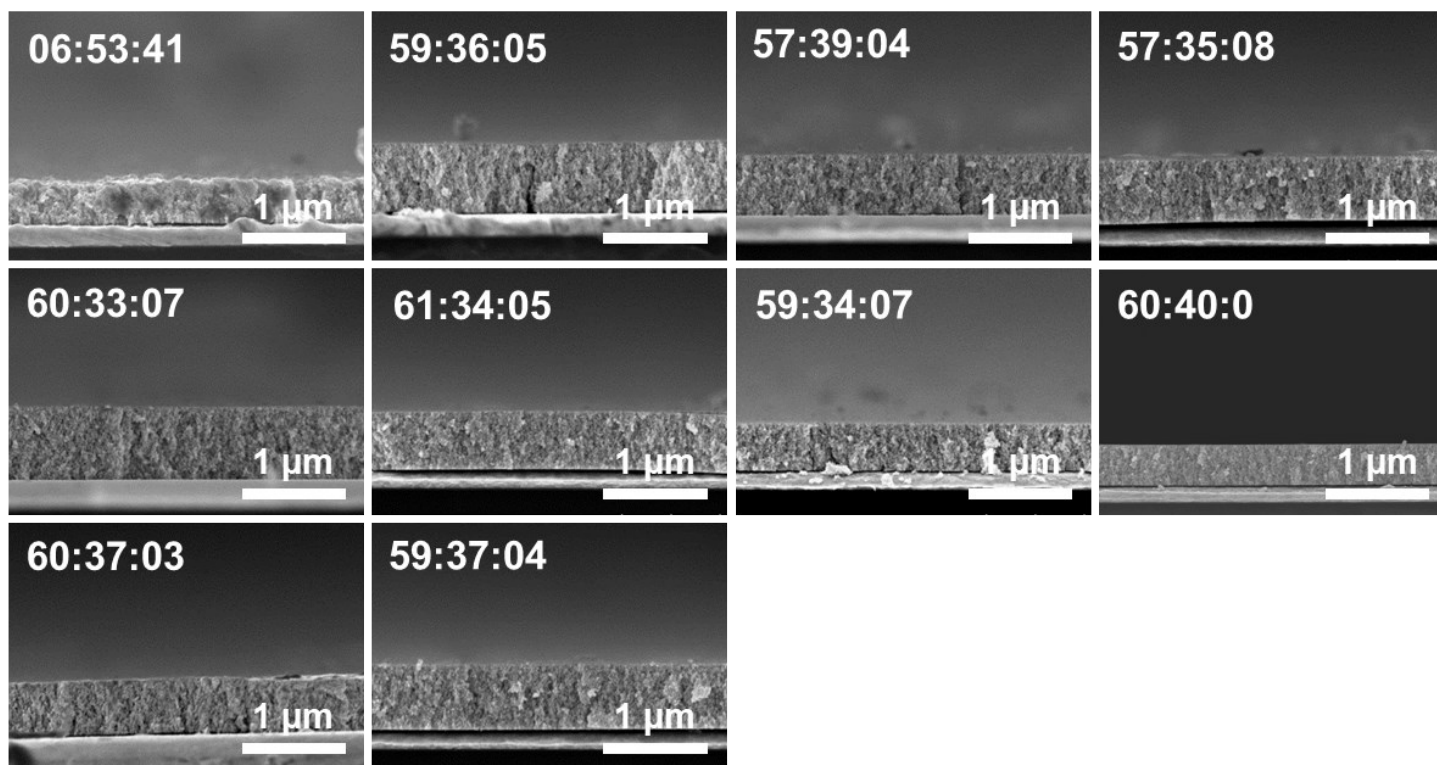
**Figure S5.** Cross-section SEM images of mesoporous PtPdAu produced by different compositional ratios of the precursor Pt:Pd:Au in the initial dataset.  $[\text{PtCl}_4]^{2-}:[\text{PdCl}_4]^{2-}:[\text{AuCl}_4]^-$  ratio in the initial electrolyte solution.



**Figure S6.** Cross-section SEM images of mesoporous PtPdAu produced by different compositional ratios of the precursor Pt:Pd:Au (dataset from 1<sup>st</sup> cycle).  $[\text{PtCl}_4]^{2-}:[\text{PdCl}_4]^{2-}:[\text{AuCl}_4]^-$  ratio in the initial electrolyte solution.

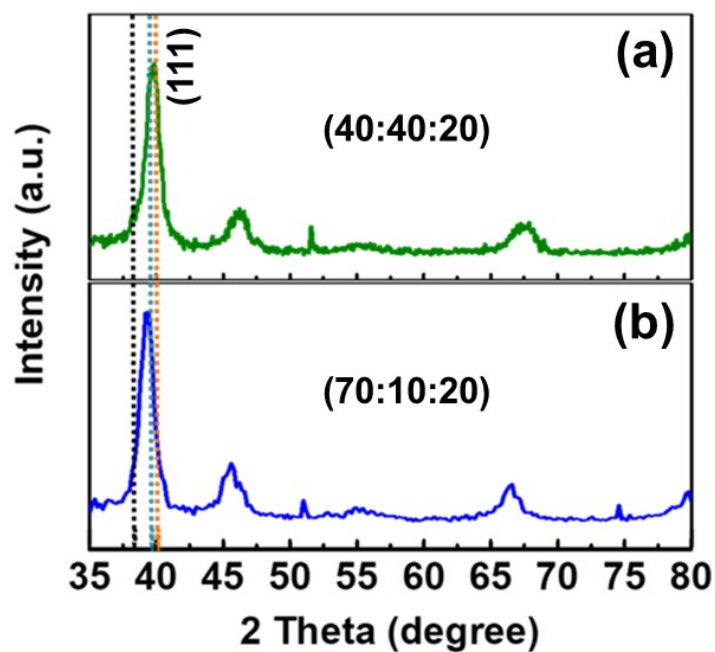


**Figure S7.** Cross-section SEM images of mesoporous PtPdAu produced by different compositional ratios of the precursor Pt:Pd:Au (dataset from 2<sup>nd</sup> cycle).  $[\text{PtCl}_4]^{2-}:[\text{PdCl}_4]^{2-}:[\text{AuCl}_4]^-$  ratio in the initial electrolyte solution.

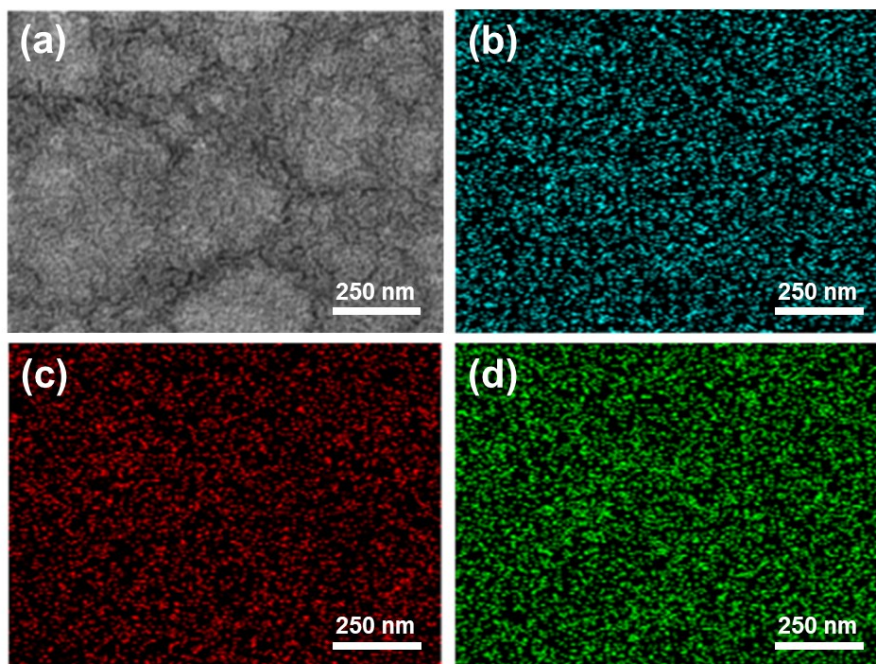


**Figure S8.** Cross-section SEM images of mesoporous PtPdAu produced by different compositional ratios of the precursor Pt:Pd:Au (dataset from 3<sup>rd</sup> cycle).  $[\text{PtCl}_4]^{2-}:[\text{PdCl}_4]^{2-}:[\text{AuCl}_4]^-$  ratio in the initial electrolyte solution.

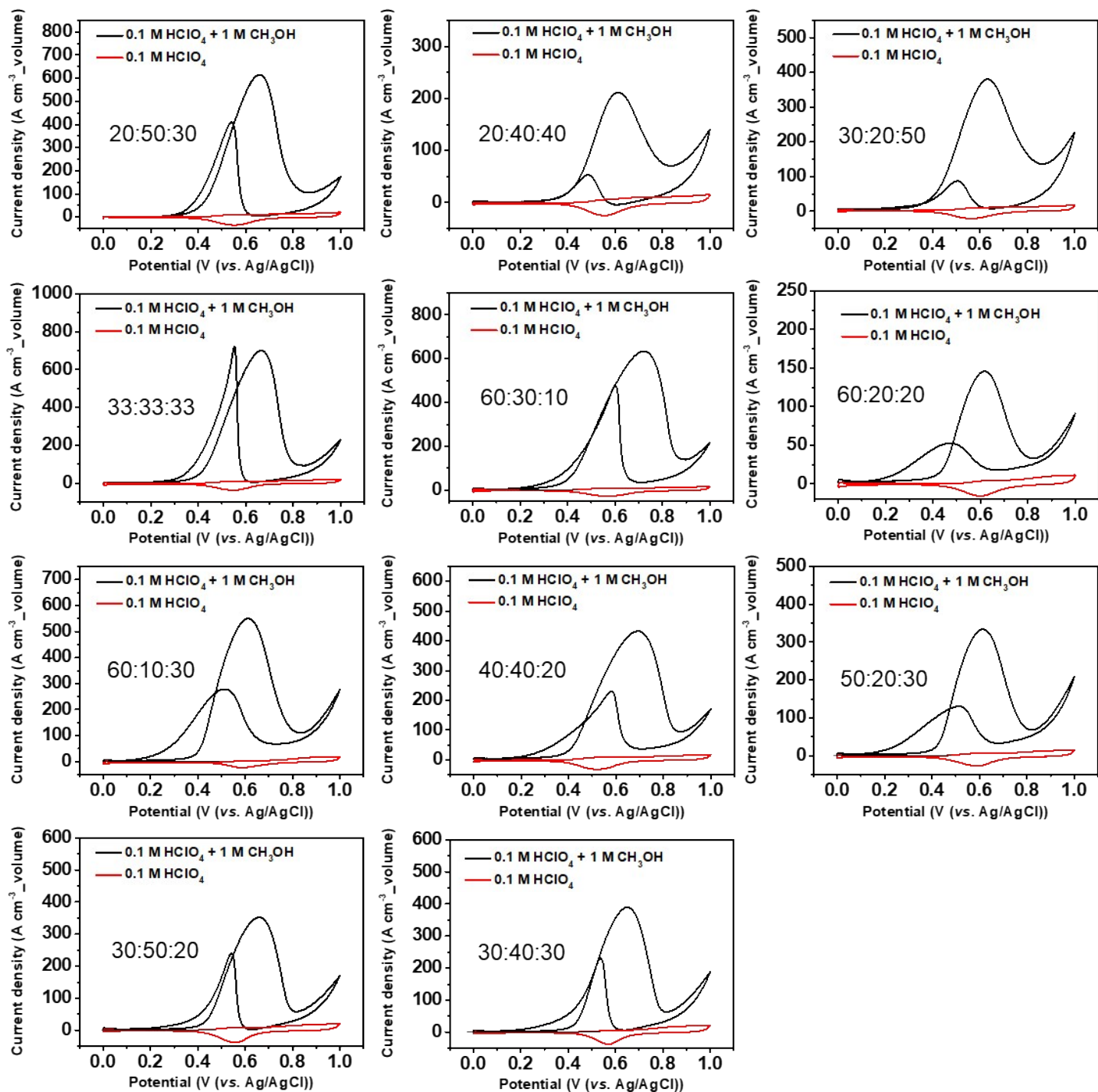




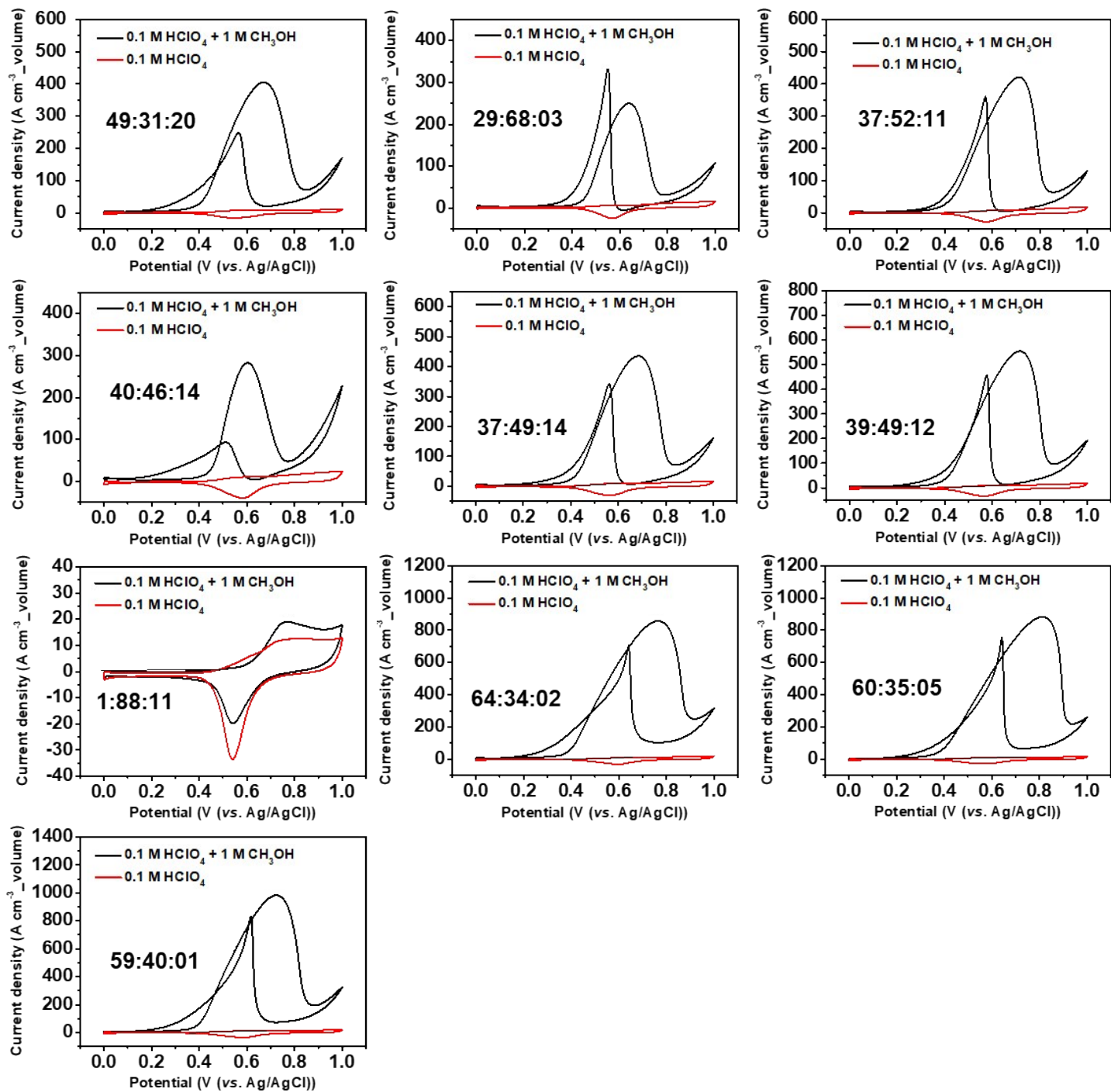
**Figure S9.** Wide-angle XRD patterns of selected mesoporous PtPdAu films in the initial dataset with the chemical compositions of metal precursors  $[\text{PtCl}_4]^{2-}$ :  $[\text{PdCl}_4]^{2-}$ :  $[\text{AuCl}_4]^-$  of (a) 40:40:20 and (b) 70:10:20. Dot lines represent: *fcc* Pt (JCPDS 05-0682), light blue; *fcc* Pd (JCPDS 04-0802), orange; *fcc* Au (JCPDS 04-0784), black.



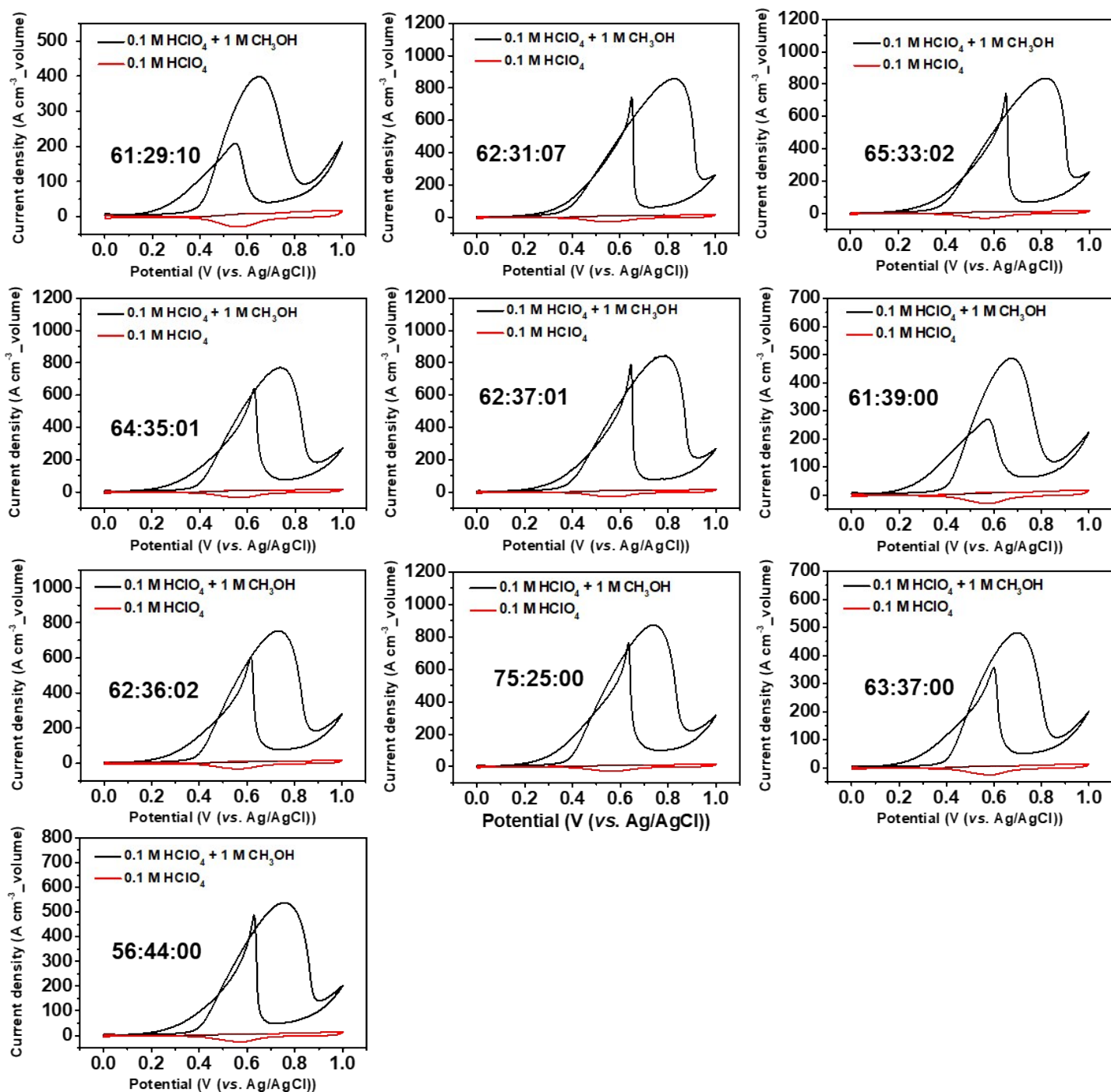
**Figure S10.** (a) Top-view SEM image and the corresponding EDX elemental mapping (b) Au, (c) Pd, and (d) Pt of mesoporous PtPdAu film fabricated from the electrolyte solution with chemical composition of  $[\text{PtCl}_4]^{2-}$ :  $[\text{PdCl}_4]^{2-}$ :  $[\text{AuCl}_4]^-$  of 33: 33: 33.



**Figure S11.** Cyclic voltammograms of methanol electrooxidation in 0.1 M HClO<sub>4</sub> + 1 M CH<sub>3</sub>OH at the scan rate of 5 mV s<sup>-1</sup> using mesoporous PtPdAu films in the initial dataset. The current density is normalized by the film volume.

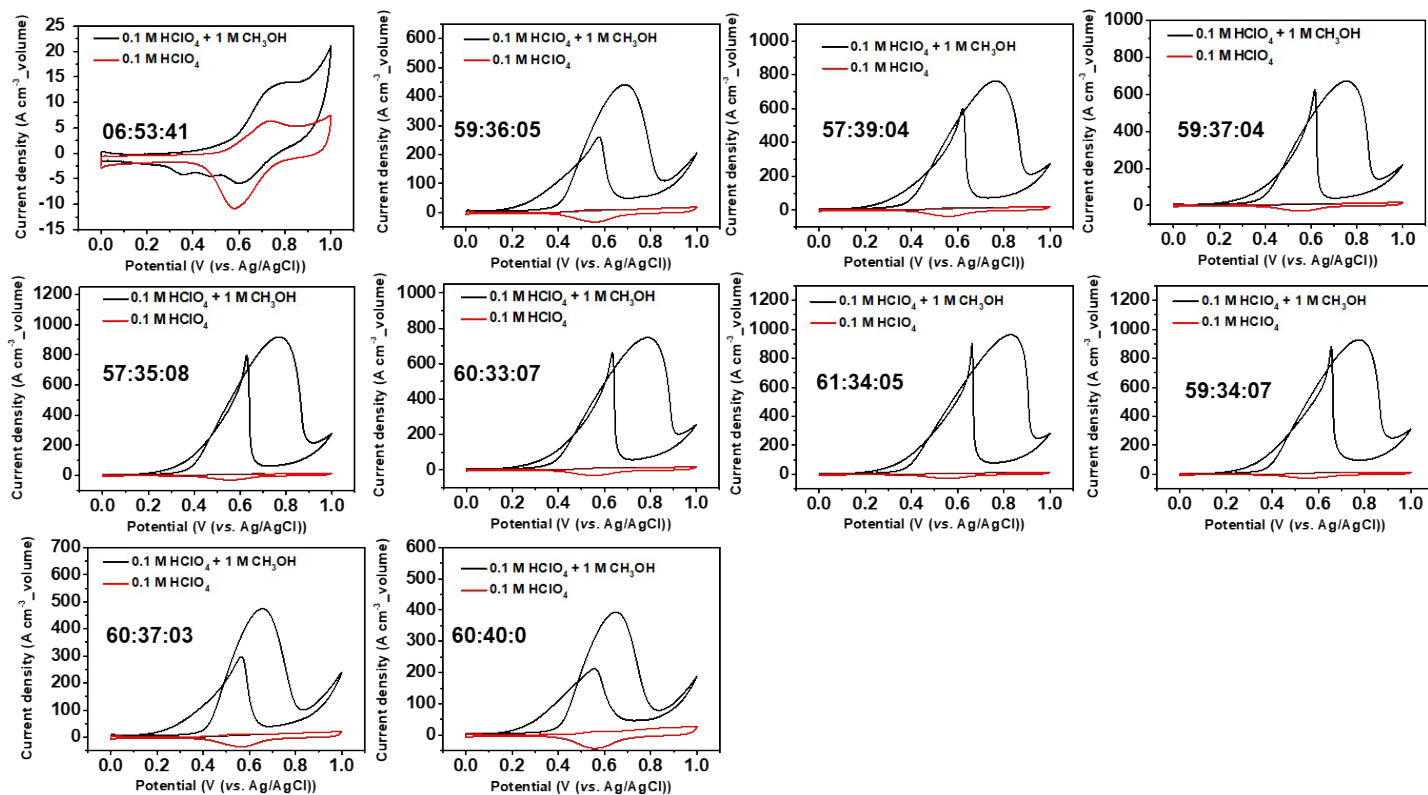


**Figure S12.** Electrocatalytic performance of mesoporous PtPdPAu with different compositions (dataset from 1<sup>st</sup> cycle) on MOR in 0.1 M HClO<sub>4</sub> + 1 M methanol at scan rate of 5 mV s<sup>-1</sup>. [PtCl<sub>4</sub>]<sup>2-</sup>: [PdCl<sub>4</sub>]<sup>2-</sup>: [AuCl<sub>4</sub>]<sup>-</sup> ratio in the initial electrolyte solution.

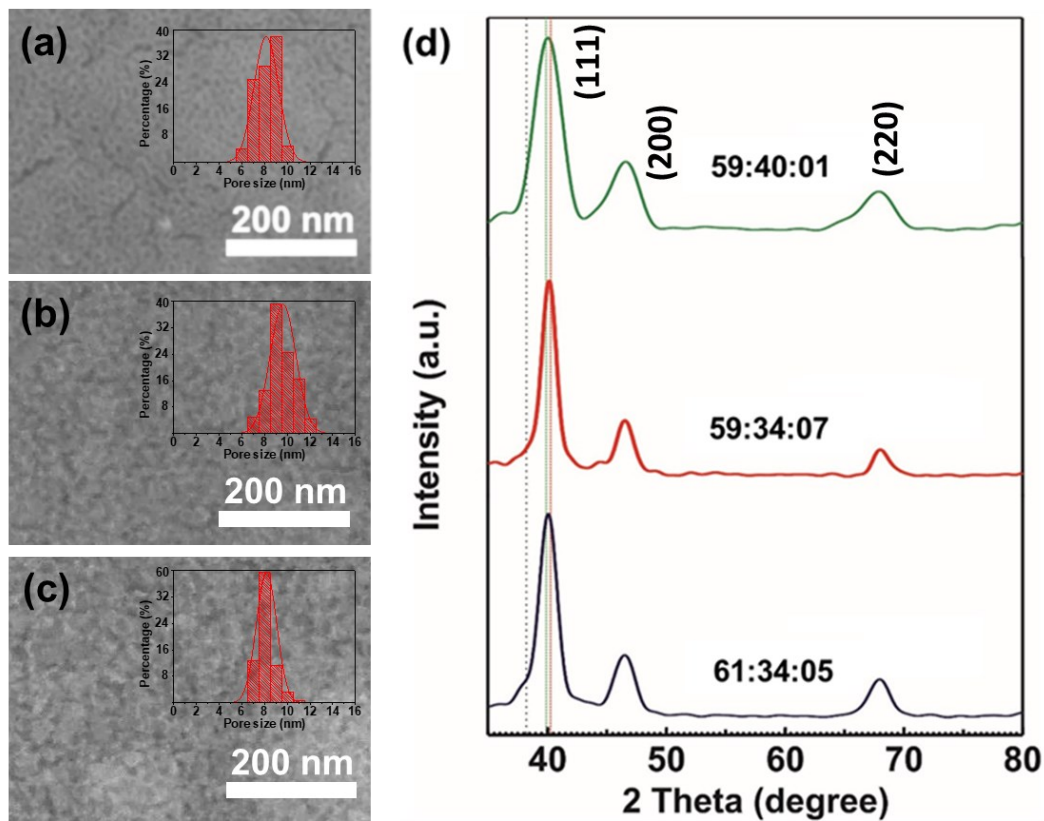


**Figure S13.** Electrochemical performance of mesoporous PtPdPAu with different compositions (dataset from 2<sup>nd</sup> cycle) on MOR in 0.1 M HClO<sub>4</sub> + 1 M methanol at scan rate of 5 mV s<sup>-1</sup>. [PtCl<sub>4</sub>]<sup>2-</sup>: [PdCl<sub>4</sub>]<sup>2-</sup>: [AuCl<sub>4</sub>]<sup>-</sup> ratio in the initial electrolyte solution.

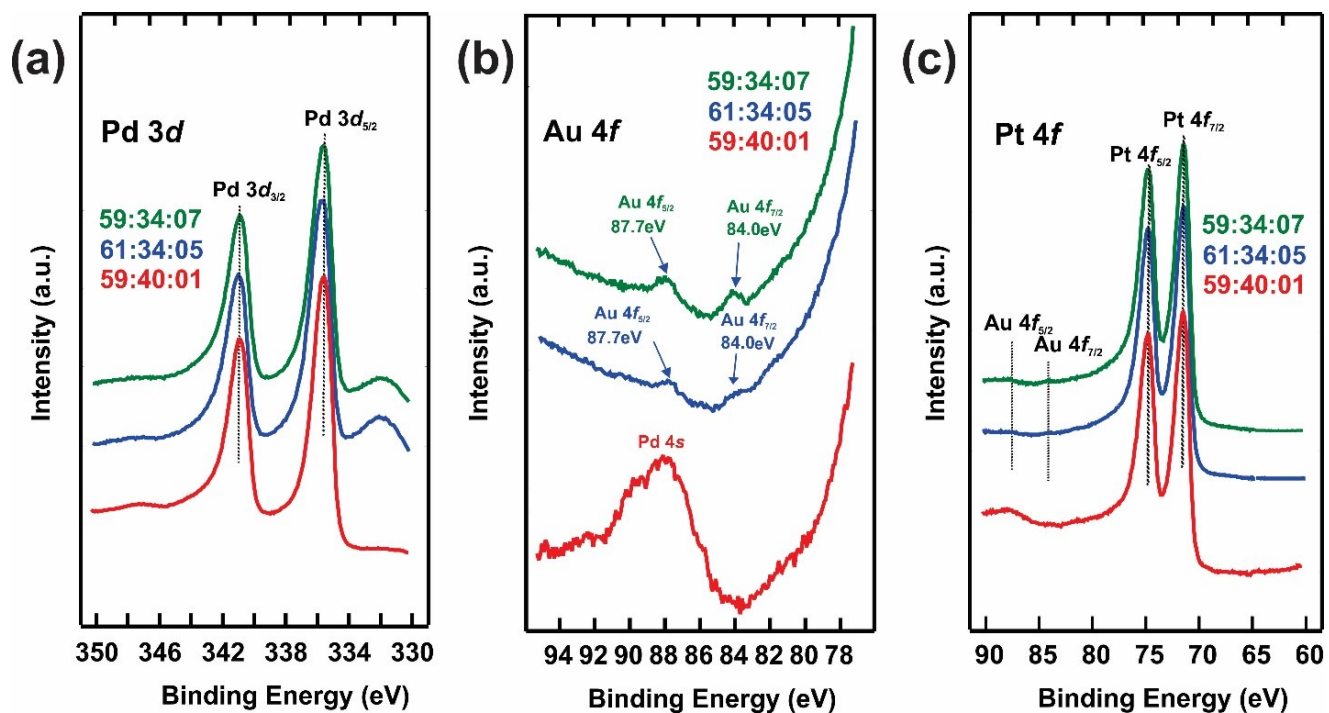




**Figure S14.** Electrochemical performance of mesoporous PtPdPAu with different compositions (dataset from 3<sup>rd</sup> cycle) on MOR in 0.1 M HClO<sub>4</sub> + 1 M methanol at scan rate of 5 mV s<sup>-1</sup>. [PtCl<sub>4</sub>]<sup>2-</sup>: [PdCl<sub>4</sub>]<sup>2-</sup>: [AuCl<sub>4</sub>]<sup>-</sup> ratio in the initial electrolyte solution.

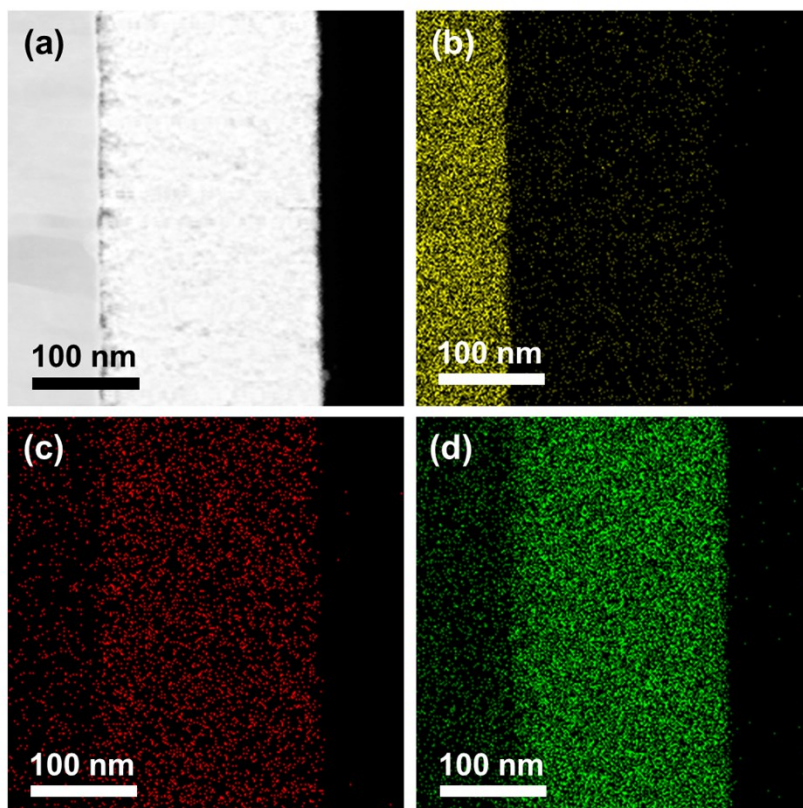


**Figure S15.** Top-view SEM images of mesoporous PtPdAu films fabricated from electrolyte solutions with  $[\text{PtCl}_4]^{2-}$ :  $[\text{PdCl}_4]^{2-}$ :  $[\text{AuCl}_4]^-$  of (a) 59: 40: 1, (b) 59: 34: 7, and (c) 61: 34: 5. (d) Wide-angle XRD patterns of mesoporous PtPdAu films. Dot lines represent: *fcc* Pt (JCPDS 05-0682), light blue; *fcc* Pd (JCPDS 04-0802), orange; *fcc* Au (JCPDS 04-0784), black.

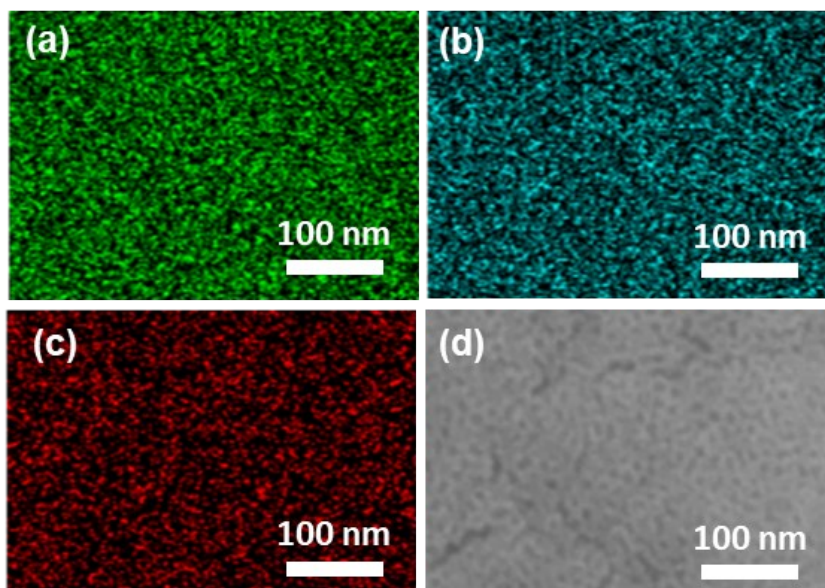


**Figure S16.** XPS spectra of mesoporous PtPdAu films fabricated from electrolyte solutions with  $[\text{PtCl}_4]^{2-}$ :  $[\text{PdCl}_4]^{2-}$ :  $[\text{AuCl}_4]^-$  of 59: 40: 1 (red line), 61: 34: 5 (blue line) and 59: 34: 7 (green line) on (a) Pd 3d, (b) Au 4f, and (c) Pt 4f.





**Figure S17.** (a) HAADF-STEM image and the corresponding EDX elemental mapping of (b) Au, (c) Pd, and (d) Pt of mesoporous PtPdAu film fabricated from the electrolyte solution with  $[\text{PtCl}_4]^{2-}$ :  $[\text{PdCl}_4]^{2-}$ :  $[\text{AuCl}_4]^-$  of 59: 40: 1.



**Figure S18.** EDX elemental mapping of (a) Pt, (b) Pd, (c) Au, and (d) Top-surface of mesoporous PtPdAu film fabricated from the electrolyte solution with  $[\text{PtCl}_4]^{2-}$ :  $[\text{PdCl}_4]^{2-}$ :  $[\text{AuCl}_4]^-$  of 59: 40: 1.

**Table S1.** Compositional ratios of metal precursors in the electrolyte solutions, and thickness and electrocatalytic results on methanol oxidation of the resulting films.<sup>[a]</sup>

Run	Compositions of metal precursors (Pt: Pd: Au) <sup>[b]</sup>	Film thickness [nm]	Onset potential <sup>[c]</sup> [V]	Peak potential <sup>[c]</sup> [V]	Peak current volume density [A cm <sup>-3</sup> ]
<b><i>Initial dataset</i></b>					
1	10: 50: 40	467	0.42	0.64	35.76
2	30: 10: 60	407	0.41	0.61	227.03
3	20: 50: 30	632	0.42	0.65	618.67
4	20: 40: 40	636	0.43	0.61	212.26
5	30: 20: 50	364	0.42	0.63	381.87
6	33: 33: 33	559	0.42	0.66	701.79
7	50: 0: 50	333	0.44	0.55	79.58
8	70: 10: 20	602	0.41	0.61	468.44
9	60: 30: 10	975	0.41	0.72	633.85
10	60: 20: 20	1110	0.43	0.62	145.95
11	60: 10: 30	449	0.40	0.61	550.11
12	40: 40: 20	923	0.40	0.70	433.37
13	40: 50: 10	1267	0.39	0.72	511.44
14	50: 20: 30	703	0.42	0.61	334.28
15	30: 50: 20	720	0.43	0.66	352.78
16	30: 40: 30	703	0.43	0.65	389.76
17	50: 50: 0	661	0.44	0.62	338.88
<b><i>Dataset from 1<sup>st</sup> cycle</i></b>					
18	49: 31: 20	728	0.41	0.67	405.32
19	29: 68: 3	677	0.45	0.64	250.01
20	37: 52: 11	838	0.42	0.71	420.31
21	40: 46: 14	635	0.46	0.60	284.05
22	37: 49: 14	847	0.42	0.68	437.89
23	39: 49: 12	677	0.41	0.71	556.92
24	1: 88: 11	610	n.a. <sup>[d]</sup>	n.a. <sup>[d]</sup>	n.a. <sup>[d]</sup>
25	64: 34: 2	567	0.39	0.76	858.32

26	60: 35: 5	670	0.38	0.81	885.85
27	59: 40: 1	457	0.39	0.72	983.47
<b>Dataset from 2<sup>nd</sup> cycle</b>					
28	61: 29: 10	711	0.40	0.65	399.02
29	62: 31: 7	720	0.36	0.83	863.68
30	65: 33: 2	618	0.37	0.82	842.02
31	64: 35: 1	576	0.38	0.74	769.66
32	62: 37: 1	635	0.375	0.79	843.83
33	61: 39: 0	533	0.40	0.67	488.50
34	62: 36: 2	508	0.39	0.73	757.66
35	75: 25: 0	449	0.39	0.74	879.32
36	63: 37: 0	584	0.39	0.69	481.99
37	56: 44: 0	627	0.38	0.76	540.94
<b>Dataset from 3<sup>rd</sup> cycle</b>					
38	6: 53: 41	452	n.a. <sup>[d]</sup>	n.a. <sup>[d]</sup>	n.a. <sup>[d]</sup>
39	59: 36: 5	676	0.40	0.69	441.05
40	57: 39: 4	611	0.38	0.69	767.41
41	59: 37: 4	660	0.38	0.76	671.42
42	57: 35: 8	647	0.38	0.75	921.06
43	60: 33: 7	726	0.37	0.77	751.96
44	61: 34: 5	597	0.38	0.79	968.73
45	59: 34: 7	503	0.38	0.83	929.33
46	60: 37: 3	561	0.41	0.65	475.34
47	60: 40: 0	483	0.41	0.65	392.61

<sup>[a]</sup>Electrochemical oxidation of methanol was measured in 0.1 M HClO<sub>4</sub> containing 1.0 M CH<sub>3</sub>OH.

<sup>[b]</sup>Molar ratios of [PtCl<sub>4</sub>]<sup>2-</sup>: [PdCl<sub>4</sub>]<sup>2-</sup>: [AuCl<sub>4</sub>]<sup>-</sup> in the electrolyte solutions.

<sup>[c]</sup>Reported with Ag/AgCl reference.

<sup>[d]</sup>Oxidation peak was too small to accurately determine the values (i.e., almost no activity).