## Ligand-Controlled Co-Reduction *versus* Electroless Co-Deposition: Synthesis of Nanodendrites with Spatially Defined Bimetallic Distributions

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**Figure S1.** TEM images of time-based growth studies of Pd-Pt core-shell nanodendrites generated by heating  $Pd(acac)_2$  with  $Pt(acac)_2$  in oleylamine at 160 °C at (A) 30 min, (B) 2 hr, (C) 4 hr, and (D) 6 hr.



**Figure S2.** (A) TEM image of representative Pd-Pt core-shell nanodendrites generated by heating  $Pd(acac)_2$  with  $Pt(acac)_2$  in oleylamine at 160 °C and corresponding (B) EDX spectroscopy analysis of PdPt nanoparticles with \* denoting signals from the sample holder and Cu single arising from the grid.



**Figure S3.** TEM images single-metal Pd nanodendrites generated by heating with  $Pd(acac)_2$  in oleylamine at 160 °C. Aliquots were collected at (A) 5 min, (B) 15 min, (C) 30 min, and (D) 60 min.



**Figure S4.** TEM images of single-metal Pt nanodendrites of generated by heating with Pt(acac)<sub>2</sub> in oleylamine at 160 °C. Aliquots were collected at (A) 5 min, (B) 15 min, (C) 30 min, and (D) 60 min.



**Figure S5.** (A) STEM image of nanodendrites 6 hrs after co-reducing Pd(hfac)<sub>2</sub> with Pt(hfac)<sub>2</sub> in oleylamine at 160 °C. Elemental mapping of denotes the presence of (B) Pd (red), (C) Pt (green), and (D) combined Pd and Pt. (E) EDX line scan profile corresponding to the blue line in (D).



**Figure S6.** TEM images of Pd-Pt nanodendrites of mixed composition generated by heating  $H_2PtCl_6$  with Pd(acac)<sub>2</sub> in oleylamine at 160 °C at (A) 30 min, (B) 2 hr, (C) 4 hr, and (D) 6 hr.



**Figure S7.** (A) TEM image of Pd-Pt nanodendrites of mixed composition generated by heating  $H_2PtCl_6$  with  $Pd(acac)_2$  in oleylamine at 160 °C and corresponding (B) EDX analysis of Pd-Pt nanoparticles with \* denoting signals from the sample holder and Cu single arising from TEM grid.



**Figure S8.** (A) TEM image of Pd-Pt product obtained from injecting Pd seeds into an oleylamine solution containing  $H_2PtCl_6$  at 160 °C. (B) EDX analysis of product, where Si and Cu come from the TEM grid.



**Figure S9.** Characterization of a nanodendrite prepared at a Pd:Pt ratio of 2:1 (Pd(acac)<sub>2</sub> (0.055 mmol) and Pt(acac)<sub>2</sub> (0.0275 mmol) co-dissolved in oleylamine (22 mL) then heated from room temperature to 160 °C (ramp rate ca. 20 °C min<sup>-1</sup>). Product collected at 6 hours.). (A) TEM image of a PdPt nanodendrite of mixed composition with selected area used for STEM/EDX analysis. Elemental mapping of selected area denotes the presence of (B) Pd (red), (C) Pt (green), and (D) combined Pd and Pt signal. (E) EDX linescan from region denoted by the blue line in (D).



**Figure S10.** Characterization of a nanodendrite prepared at a Pd:Pt ratio of 1:1 (Pd(acac)<sub>2</sub> (0.055 mmol) and Pt(acac)<sub>2</sub> (0.055 mmol) co-dissolved in oleylamine (22 mL) then heated from room temperature to 160 °C (ramp rate ca. 20 °C min<sup>-1</sup>). Product collected at 6 hours.). (A) TEM image of a PdPt nanodendrite of mixed composition with selected area used for STEM/EDX analysis. Elemental mapping of selected area denotes the presence of (B) Pd (red), (C) Pt (green), and (D) combined Pd and Pt signal. (E) EDX linescan from region denoted by the blue line in (D).



**Figure S11.** Characterization of a nanodendrite prepared at a Pd:Pt ratio of 1:2 (Pd(acac)<sub>2</sub> (0.0275 mmol) and Pt(acac)<sub>2</sub> (0.055 mmol) co-dissolved in oleylamine (22 mL) then heated from room temperature to 160 °C (ramp rate ca. 20 °C min<sup>-1</sup>). Product collected at 6 hours.). (A) TEM image of a PdPt nanodendrite of mixed composition with selected area used for STEM/EDX analysis. Elemental mapping of selected area denotes the presence of (B) Pd (red), (C) Pt (green), and (D) combined Pd and Pt signal. (E) EDX linescan from region denoted by the blue line in (D).