Supporting Information
Ms. ID: nn-2016-04448k

22 August 2016

Efficient CO Oxidation Using Dendrimer-Encapsulated Pt Nanoparticles Activated with <2% Cu Surface Atoms

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**Figure S1.** (a) TEM images and (b) corresponding size distribution histogram of Vulcan carbon-supported Pt\textsubscript{147} DENs.

**Figure S2.** (a) TEM images of freshly-prepared, Vulcan-supported Pt\textsubscript{147}\textsubscript{Cu\textsubscript{partial}} DENs after immersion in deaerated 0.10 M NaOH but before CO oxidation, (b) Pt\textsubscript{147}\textsubscript{Cu\textsubscript{partial}} DENs after performing bulk CO oxidation using a RDE. CO oxidation conditions are -0.60 V vs. Hg/Hg\textsubscript{2}SO\textsubscript{4}, CO saturated 0.10 M NaOH, \(\omega = 1600\) rpm and \(t = 150\) s. (c) Corresponding size distributions of Pt\textsubscript{147}\textsubscript{Cu\textsubscript{partial}} DENs before and after CO oxidation.
**Figure S3.** Three consecutive CVs for Pt$_{147}$\_Cu$_{\text{partial}}$ DENs in deaerated 0.10 M NaOH. The scans started at -0.90 V and initially proceeded in the positive direction. The scan rate = 100.0 mV/s
Figure S4. XPS spectra of Pt$_{147}$ DENs, Pt$_{147}@Cu_{full}$ DENs and Pt$_{147}@Cu_{full}$ DENs after CO oxidation. The preparation of Pt$_{147}$ and Pt$_{147}@Cu_{full}$ DENs samples is described in the Methods section of the main text. To obtain the spectrum of Pt$_{147}@Cu_{full}$ DENs after CO oxidation, the following slightly modified procedure was used. First, Pt$_{147}$ DEN ink was dropcast onto a double-sided, XPS grade, conductive carbon tape attached to a GC-RDE. Next, Pt$_{147}@Cu_{full}$ DENs were prepared by depositing a full monolayer of Cu onto the Pt$_{147}$ DENs using the same UPD method described in the Methods. Next, the Pt$_{147}@Cu_{full}$ DENs-modified GC-RDE was cleaned using N$_2$-purged DI water, and then the electrode was immediately transferred to a CO-saturated, 0.10 M NaOH solution. Following CO oxidation at $-0.7$ V for 5 min and $\omega = 1600$ rpm, the carbon tape (with the adherent DENs) was removed from the GC-RDE, attached to a glassy carbon chip, and dried under flowing N$_2$ prior to XPS analysis. Note that resistance of the carbon tape was $\sim$1000 $\Omega$, and was therefore electronically compensated during electrochemical measurements.
**Figure S5.** Current-time traces for bulk CO oxidation using Pt\textsubscript{147}, Pt\textsubscript{147}@Cu\textsubscript{partial}, and Pt\textsubscript{147}@Cu\textsubscript{full} DEN-modified GC-RDEs in CO-saturated 0.10 M NaOH at -0.60 V vs. Hg/Hg\textsubscript{2}SO\textsubscript{4}. \(\omega = 1600\) rpm.

**Figure S6.** Calculated \(E_{b_{\text{OH}}}\) and \(E_{b_{\text{CO}}}\) at the three-fold Pt surface sites (blue arrows) in the Cu@core and Pt\textsubscript{147} models.