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### Hierarchical approach of mitigating carbon influence in nano-porous

electro-catalyst with unique surface islands for efficient methanol resistive oxygen

#### reduction

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#### Abstract

For the first time, we report a facile one pot aqueous method for support-free Pd<sub>85</sub>Pt<sub>15</sub> nano-porous structures (NPoS) synthesis from PdMn nano-alloys at ambient conditions. A hierarchical approach was successfully employed through a simple "self-settlement" process with descending amounts of carbon to carbon-free electro-catalysts to improve catalyst utilization and avoid carbon degradation during operating conditions. Pd<sub>85</sub>Pt<sub>15</sub> NPoS exhibits enhanced methanol resistive oxygen reduction reaction (ORR) activity owing to the presence of highly active and unique surface PdPt islands compared to HiSPEC Pt/C catalysts. Accelerated durability tests of the support-free PdPt NPoS show enhanced durability in harsh acidic environment (1.0 N H<sub>2</sub>SO<sub>4</sub>) compared to HiSPEC Pt/C and DOE 2017-2020 durability target. Preliminary direct methanol fuel cell studies using hierarchically derived Pd<sub>85</sub>Pt<sub>15</sub> NPoS variants were performed at ultra-low Pt content. The effects of carbon content and catalyst layer thickness on fuel cell activities are well discussed.

Keywords: Nano-porous surface, surface-active Pd-Pt islands, methanol tolerant oxygen reduction reaction, direct methanol fuel cell, support-free ORR catalysts

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